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PROGRESS REPORT

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FOREWORD

The Reactor Development Program Progress Report, issued monthly, is intended to be a means of reporting those items of significant technical progress which have occurred in both the specific reactor projects and the general engineering research and development programs. The report is organized in a way which, it is hoped, gives the clearest, most logical 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:

August 1966	ANL-7249
September 1966	ANL-7255
October 1966	ANL-7276
November 1966	ANL-7279
December 1966	ANL-7286
January 1967	ANL-7302

REACTOR DEVELOPMENT PROGRAM

Highlights of Project Activities for February 1967

EBWR Plutonium Recycle

After three months of operation at 42 MW, the EBWR was shut down on February 11, 1967 to remove regular and special fuel pins. These will be analyzed for specific isotopes by the Pacific Northwest Laboratory. The fuel in the plutonium zone had an average exposure of approximately 1300 MWd/tonne.

During this shutdown period the pile oscillator will be attached to the center control rod and will be used to measure the reactor transfer function between 42 and 70 MW. After the transfer-function measurements are made, the reactor will be operated at 70 MW or the highest stable power attainable below 70 MW.

The next shutdown is scheduled for late April in order to remove additional sample pins.

EBR-II

Based on surveillance measurements of selected depleted uranium elements in inner blanket Row 7, the depleted uranium in this row was replaced with subassemblies containing stainless steel rods. Thirty-one of these latter subassemblies were loaded into the basket and then into Row 7. At the same time, a detailed study to determine swelling in the uranium in Rows 7 and 8, and to correlate this with exposure time and temperature, is under way before starting Run No. 25. Meanwhile neutron flux wire irradiations have been conducted at low powers to obtain measurements of the distribution rates of fission and plutonium production in the radial blanket.

Fuel Cycle Facility

Radial blanket subassemblies from Rows 6, 7, and 8 of the reactor are being examined as part of the continuing surveillance program. Thus far, seven rods from Rows 6 and 7 have been measured for cladding OD, slug OD, length, and density. Central slugs (that is, in positions of highest flux) showed significant diametral increase, slight increase in length, and some accompanying decrease in density. A maximum increase of 3 mil in the diameter of the cladding was measured, but no evidence of uranium-cladding contact has been observed. All 6th row blanket subassemblies,

which have been replaced by driver or experimental subassemblies, were cleaned of sodium and stored in steel-lined pits in the waste burial ground. A few have been reserved for surveillance.

ZPR-3

Experiments with Assembly 48 relevant to the FFTF analysis and design were completed. These included radial traverses with poison rods in place and in the clean core with no poison rods, and interaction effects between poison rods with varying boron-10 content.

Reloading of the reactor for Assembly 48B is under way. This is the basic Assembly 48 with a central zone containing plutonium with a higher Pu^{240} content. A series of experiments will be run for comparisons with the original Assembly 48.

ZPPR

Pouring of concrete in various areas has been accomplished. The topping slab for the main floor of the support wing, walls of the vault-workroom area, the 29 required concrete pile caps, and the first of six pours for the reactor pit and cell floor are completed. All structural steel and roof decking for the support wing have been placed, and suspect waste retention tanks and pumps have been installed. Fifty-two of 54 steel caps for the piling posts have been welded on. Prestressed reinforced-concrete panels for the roof have been set in place.

AARR

Specifications for the primary pumps were sent to vendors for bids. Title II detailed design has begun.

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I. PLUTONIUM UTILIZATION--EBWR

A. Research and Development

1. Operation of Plutonium-fueled EBWR

After three months of operation at 42 MW, with a generation output of 4.5 MWe, EBWR was shut down on February 11, 1967 to remove 17 regular and special fuel pins from the northwest quadrant of the plutonium zone. The pins will be analyzed for specific isotopes by the Pacific Northwest Laboratory. The fuel had an average plutonium zone exposure of approximately 1300 MWd/tonne.

The power generated by the plant was as follows:

	<u>Thermal Power, MWt</u>	<u>Electrical Power, MWe</u>
During February 1967	10,314	1,176
Total since start of Plutonium Recycle Program	90,299	10,044

During this shutdown period the pile oscillator will be attached to the center control rod and will be used to measure steam quality and the reactor transfer function between 42 and 70 MW in 10-MW steps. After these measurements are made, the reactor will be operated at 70 MW or the highest stable power attainable below 70 MW.

The next shutdown is scheduled for late April in order to remove additional sample pins.

II. LIQUID-METAL FAST BREEDER REACTORS

A. EBR-II

1. Operations

Flux wire irradiations were conducted to obtain distribution measurements of fission and plutonium production rates in the radial blanket of the reactor. Six special flux wire subassemblies were irradiated in two groups, three for 100 kW and three for 1000 kW (Run No. 25C). The special subassemblies were irradiated in Rows 7, 9, 11, 13, 14, and 15 of the reactor. Data from these runs are being analyzed.

During routine fuel handling following Run 25B it was noted that the sense rod on the oscillator drive shaft (a device to indicate the position of the oscillator rod head in the gripper jaws) was binding and required manual assistance to move. The sense rod was also observed to move with the jaw-actuating sleeve. This indicated the presence of some foreign material or possible galling between the sense shaft and the jaw-actuating sleeve. These symptoms could also be indicative of a broken sense-rod bellows. It was decided to leave the rod drive in place to observe the behavior of the sense rod, perhaps through Run No. 25. Subsequently, indications were that the sense rod was becoming inoperative, and the operation of the jaw drive became extremely difficult. The oscillator mechanism was therefore removed for inspection and replacement of defective parts. A standard control rod and control-rod drive have been inserted in this location. A new oscillator drive gripper, lower shaft end, and bellows assembly are being fabricated. A study is also under way to determine if the jaw-actuator angles require changing to achieve better "open-close" operation.

Based on surveillance measurements of depleted uranium in the outer row of the inner blanket (Row 7), the depleted uranium in this row was replaced with subassemblies containing stainless steel rods. A detailed study is under way to determine the amount of swelling in the uranium from Rows 7 and 8, and to correlate these measurements with exposure and temperature (see Sect. II.A.6). These data, along with future surveillance measurements should provide a firm basis for scheduling blanket-subassembly replacement. To provide sufficient storage in the fuel-storage basket for removal and replacement of the 7th row blanket subassemblies, 24 depleted uranium subassemblies were removed and transferred to the Fuel Cycle Facility for disposal. Thirty-one stainless steel subassemblies have been transferred into the storage basket and loaded into Row 7.

Experimental subassembly XG06 was removed at the request of the experimenter (GE) and replaced by a C2000-type half-worth subassembly (see Sect. II.A.4).

A survey of potential experimenters interested in utilizing instrumented subassemblies in EBR-II has been completed.

Fabrication of a new backup Fuel Unloading Machine (FUM) gripper is about 40% complete. Another revision to the jaws of the present FUM gripper was completed and operation at the present time seems to be satisfactory. Design of a new manually operated jaw actuator is in progress.

Fabrication of the new sodium-level transmitter for the primary tank is in progress. A test is being conducted on the load cell that will be used on this unit.

2. Reactor Improvements

a. Irradiation Subassembly, Mark C37. To determine hole sizes for orifices that will bypass part of the total coolant flow through the subassembly, a second series of hydraulic tests was performed (see Progress Report for January 1967, ANL-7302, pp. 13-14). The tests started with orifices consisting of six holes, each 0.125-in. in diameter, and progressed in convenient increments of diameter up to 0.593 in. Measurements of the resulting flow velocities were made by pitot-static instrumentation installed in the test subassembly. The instrumentation was found to be very accurate; except for readings at low velocities, the error was less than 1% for each hole size tested. From these data, the quantity of bypass flow was calculated and the percentage of total flow was determined. With knowledge of the neutron flux and the heat-generation characteristics of the material to be irradiated, an experimenter will be able to choose a hole size to give coolant flows that will yield the test temperature he desires.

A new series of tests will be conducted to verify proper mixing of the bypass flow with the coolant flowing from the specimen. This mixing is important to prevent undesirably high temperature differentials along the walls of the outer hexagonal tube and at the lifting fixture located above the mixing section.

b. Improved Gripper and Holddown Force-limiting Device. Design effort is continuing. Principal emphasis is on refining the force-limiting devices (see Progress Report for January 1967, ANL-7302, pp. 14-15) to provide continuous measurements of the forces applied by the gripper and holddown. The deflections of the force springs will be monitored by the linear-motion resistance potentiometers. Electrical signals from the potentiometers will provide information at the Fuel Handling Console for stopping the drives and indicating the value of the applied forces. An adjustable tripping point and an adjustable zero-offset will be provided so that the stopping and alarm signals can be set to operate at some desired

value of applied force above that required to drive the mechanism through its packing gland. It is planned to provide for automatic setting of the zero offset ("zero calibration") while the mechanism is running through a region of travel where the drive is loaded only by the packing gland. There would also be provisions for locking out this automatic feature and setting the zero manually while observing the force required to drive through the packing gland. Total force limits, independent of the zero-setting, will also be provided.

The automatic zero-setting feature will probably be provided by using an instrument-type servomechanism repeater. The servo will position a zero-offset potentiometer in response to the signal from the linear potentiometer on the mechanism during the zero-calibration period. At the end of the calibration period further motion of the zero-offset potentiometer will be prevented either by decoupling it with a small magnetic clutch or by shutting off the servo. Therefore, the zero-offset setting will remain until the next calibration period. This method is being chosen because it provides an offset setting that is retained until the next calibration regardless of the time interval or any intermediate control-panel shutdown or other loss of electrical power.

Two important considerations in the design of the force-limiting system are reliability and continuity of operation. Redundancy will be employed. Since the main characteristic of the system is that it respond to a certain amount of spring deflection, the deflection will be measured by at least two separate transducers whose signals will be carried back to the Fuel Handling Console on separate channels, where they will be compared. If the signals are different, an alarm will be initiated. In case of failure of one channel, the other can be used alone temporarily to provide the necessary signals for continuing operations. A possible arrangement is to have two separate linear-motion potentiometers mounted on the mechanism, each measuring the spring deflection, and one feeding a servo repeater with the zero-setting feature and the other feeding a fast responding electronic amplifier. In addition, each potentiometer has a second identical resistance element for other simple indication or use as a replacement in case the main element fails.

c. Irradiated Fuel Monitor. Fabrication of mechanisms is continuing (see Progress Report for January 1967, pp. 11-12); approximately 15% of the work is completed. Difficulties in purchasing parts and extended delivery dates might delay completion until May.

d. Rotating Plug Seal Cleaning System. Further wear and life tests of the prototype pump for the tin-bismuth eutectic alloy are being carried out (see Progress Report for January 1967, pp. 12-13). The pump has been subjected to repeated heating and cooling cycles during simulated operation with favorable results; bearings have not worn excessively and the small (1/4-in.-dia) impeller driveshaft has not distorted.

Methods to separate the oxide material from the tin-bismuth eutectic alloy are being investigated. A "skimming" system is being developed to take advantage of the tendency of the oxides to float on the surface of the alloy. A motorized skimmer bowl is being designed to test the feasibility of this concept in a mockup.

e. Instrumented Subassemblies. Work on the development of the instrumented subassembly system is continuing (see Progress Report for November 1966, ANL-7299, pp. 5-6, for a general description, and the Report for January 1967, ANL-7302, pp. 15-18, for further details).

(i) Operating Mechanism. To permit the rotating plug to move for indexing purposes during normal EBR-II fuel-handling operation, it will be necessary to raise the instrumented subassembly approximately 8 ft. For this purpose a mechanism is being designed that involves a unitized structure with an electric drive motor, a lead-screw and nut, a guided elevator plate, an instrument-lead terminal box, and an extension-tube seal and bellows. The mechanism, which can be placed in the space occupied by any one of the 12 existing control-rod drives, will be supported by the center column that supports the control-rod drives.

The drive motor operates the lead-screw and nut attached to the guided elevator plate, which supports the instrumented-subassembly package consisting of the instrument-lead terminal box, the extension tube, and the instrumented subassembly. The extension-tube seal and bellows provide a gastight seal between the extension tube and the rotating plug. The seal consists of a packing gland which is clamped tight for operation, or loosened for unloading, by means of a seal drive. The bellows permit relative vertical movement between the seal and the extension tube.

Safety features are included in the design of the mechanism so that excessive friction forces will automatically stop the motion of the subassembly system. The force required to raise or lower the instrumented subassembly will be observed at all times by means of load-transducer instruments; thus friction forces greater than normal inside the reactor vessel and cover or in the seal region will be indicated. The actuation of appropriately installed limit switches at the seal and the elevator plate region will arrest further motion.

3. Surveillance

a. Surface Anomalies on Blanket-element Tubing. Because of anomalies observed in the surface appearance of the outer hexagonal can of standard subassemblies after they were removed from the reactor and cleaned in steam and water, the metallurgical effects of cleaning (with water) EBR-II blanket-element tubing after exposure to sodium have been

investigated. After being cleaned and stored in the Air and Argon Cells, most subassemblies have a bright, typically stainless steel appearance. In contrast, several out of several hundred subassemblies exhibit an unexplained pattern of alternate bright and dark surface areas.

On the basis of other materials evaluations (see Progress Reports for January 1967, pp. 42-46; for August 1966, ANL-7249, pp. 34-35, and for intervening months) it was suspected that the water-cleaning operation after sodium exposure could produce results of the nature observed. Therefore, to investigate the anomaly, a previously unused tubular sample of EBR-II blanket-element 304 SS jacketing was exposed (pretreated) in 1200°F, flowing sodium for 5 days, washed in alcohol for 1 hr to react surface sodium residue, allowed to stand in ordinary laboratory air atmosphere for 60 days, and then was immersed in distilled water at 180°F for 7, 14, 21, and 29 days. An as-received (not exposed to sodium) sample was also immersed in water as a control specimen.

After 29 days of immersion in water, the specimen that had been previously exposed to sodium was cut up and wall sections at six locations were examined metallographically. The specimens were polished but not chemically etched.

Results include: (a) The 29-day control sample that had no sodium exposure showed no surface discoloration or visual evidence of surface or subsurface corrosion; (b) the sodium-treated specimen showed visible evidence of surface corrosion, with alternate dark and bright areas visually similar to surface effects observed on blanket jackets. Examination of the wall sections showed that the microstructures of the sodium-exposed and water-cleaned 304 SS samples were similar in nature to sodium-exposed samples subjected to a modified Strauss solution (described in Progress Reports for August 1966 through January 1967). Apparently, it takes distilled water about 30 days to produce the same visual effects on 304 SS microstructure that can be accomplished in 15 min in modified Strauss solution.

The depth and extent of the corrosion of a sodium-exposed and water-cleaned specimen were measured. The extent of 29-day water corrosion on the sodium-pretreated specimen of 304 SS was measured by means of a standard 1/8 x 1/8-in. square-grid-counting technique. Penetration of the outer surface was about 0.006 in. deep; penetration of the inner surface was about 0.005 in. deep. From these measurements the rate of corrosion in water was determined to be 8.6×10^{-6} in./hr; it has been reported¹ that 304 SS in 160°F distilled water normally corrodes at a rate approximately 0.01×10^{-6} in./hr. Comparison of these measurements with those from

¹Polar, J. P. (Climax Molybdenum Co.), A Guide to Corrosion Resistance, p. 257, Caxton Printers, Ltd., Caldwell, Idaho (1961).

tests in which a modified Strauss solution was used instead of water revealed that the depths by which water corrodes sodium-affected grain boundaries is the same as that by which the Strauss solution etches out the carbide grain boundaries that have interacted or been infused with sodium. The only difference in the tests is the duration of exposure to the cleaning agent, in one case Strauss solution for 15 min and in the other distilled water for 29 days.

4. Experimental Irradiations

The status of all experimental irradiations in the reactor and the approximate exposure at month's end is shown below:

Sub	Date Loaded	Capsule Content and Number of Capsules ()	Experimenter	Approximate Accumulated Exposure (MWd)	Goal Exposure (MWd)
XG02	7/16/65	UO ₂ -20 w/o PuO ₂ (1) Stainless Dummies (18)	GE	9,744	13,600
XG03	7/16/65	UO ₂ -20 w/o PuO ₂ (2) Stainless Dummies (17)	GE	9,744	19,450
XG04	7/16/65	UO ₂ -20 w/o PuO ₂ (2) Stainless Dummies (17)	GE	9,744	39,000
XG05	9/3/65	UO ₂ -20 w/o PuO ₂ (9) U-15 w/o Pu-10 w/o Zr (1) U-15 w/o Pu-10 w/o Ti (1) UC-20 w/o PuC (3) Structural (5)	GE ANL ANL ANL GE	9,317	14,750
XA08	12/13/65	UC-20 w/o PuC (8) Structural (11)	ANL	7,495	19,800
XO10	3/24/66	UO ₂ -20 w/o PuO ₂ (4) Structural (11) Structural (4)	GE ANL PNWL	6,675	19,600
XO11	5/9/66	UO ₂ -20 w/o PuO ₂ (7) UO ₂ -20 w/o PuO ₂ (9) 304 SS-30 w/o PuO ₂ (1) 304 SS-20 w/o PuO ₂ (1) 304 SS-20 w/o UO ₂ (1)	ANL GE PNWL PNWL PNWL	5,104	8,300
XO12	8/10/66	UO ₂ -20 w/o PuO ₂ (19)	NuMEC	2,975	20,600
XO14	7/17/66	Structural (5) Graphite (2) Structural (7) Structural (5)	PNWL PNWL GE NRL	3,674	a
XO15	11/15/66	PuO ₂ -UO ₂ (11) PuO ₂ -UO ₂ (2) (Pu,U)C (4) MK-IA (Metal) (2)	NuMEC GE ANL ANL	1,320	11,000
XO16	1/13/67	Structural (9) Structural (10)	ANL GE	0	3,000
XO17	11/15/66	PuO ₂ -UO ₂ (11) (Pu,U)C (3) MK-IA (Metal) (5)	NuMEC UNC ANL	1,320	6,500

Sub	Date Loaded	Capsule Content and Number of Capsules ()	Experimenter	Approximate Accumulated Exposure (MWd)	Goal Exposure (MWd)
XO18	12/6/66	Structural (3)	GE	630	21,300
		Structural (1)	PNWL		
		Structural (2)	ANL		
		Structural and Heavy Metal Fission Yield Samples (1)	ANL		
XO19	1/13/67	UO ₂ -20 w/o PuO ₂ (7)	GE	0	7,500 ^b
		(U _{0.8} Pu _{0.2}) ^C (3)	UNC		
		Structural (8)	PNWL		
		Graphite (1)	PNWL		
XO20	1/13/67	UO ₂ -20 w/o PuO ₂ (9)	GE	0	7,500 ^b
		(U _{0.8} Pu _{0.2}) ^C (3)	UNC		
		Structural (4)	PNWL		
		Structural (2)	ANL		
		Graphite (1)	PNWL		
XO21	2/27/67	Structural (7)	PNWL	0	21,500
XO22	2/27/67	Structural (7)	PNWL	0	8,000

^aMaximum attainable before reaching experimental subassembly capacity.

^bAre being considered for further exposure.

Subassemblies XO21 and XO22 were assembled, flow-tested, and installed in Rows 2 and 7, respectively. These irradiations utilize the Mk-B-7 experimental subassembly, of which there are now three in the reactor (XO18 being the other).

During the current extended shutdown, GE decided to remove subassembly XG06, which had achieved about 5 a/o burnup, so that these samples could be examined and evaluated prior to the time they submitted unencapsulated (singly clad) elements for irradiation. The twelve UO₂-20 w/o PuO₂ fuel samples in XG06 ranged in axial average pin exposure from a maximum of about 5.2 a/o to a minimum of 4.6 a/o. In addition, two ANL metal-alloy pins were included in XG06 and had reached exposures of about 4.8 and 4.5 a/o burnup. The remaining five test capsules contained structural materials of Incoloy-800, and 304, 316L, 321, and 347 stainless steel. The average exposure for these structural materials capsules was about 3.3×10^{22} nvt (total flux). All the GE capsules were shipped from the EBR-II site on February 21.

Inner blanket stainless steel subassembly A-800 and outer blanket stainless steel subassembly U-1600 were calibrated in the flow-test loop.

5. Vertical Assembler-Dismantler (VAD)

Assembly of the VAD was started. One of the major assembly operations--alignment and doweling of rails into place on the framework--has been completed. Other units of the machine are being added as they

are received or assembled. The partially completed machine has been placed in a floor pit which simulates the one in the Air Cell. Manipulators, a crane, and a window mockup have also been installed in preparation for the remote installation and operational tests to follow.

While assembly of many units is being carried forward, lack of a number of key components delays completion of several of the larger sub-assemblies. Attempts to expedite deliveries have been only moderately successful. As a consequence, in order to make continued effective progress, it has been necessary to resort to such expedients as: the omission of plating and other finishes, the omission of inspection operations, and the partial assembly of some of the units. In addition, we have arranged a bench test of the performance of the servo vertical drive units because this cannot yet be carried out on the machine.

Air-flow tests of two prototype elbow flowmeters have been concluded. These devices are equivalent in characteristics to those of air-velocity pitots now in use on the EBR-II Subassembly Dismantler cooling system. The elbows have the advantage of being less delicate and less easily clogged than the pitots. They are being incorporated into the VAD cooling-system piping.

6. Fuel Cycle Facility (FCF)

a. Examination of Irradiated Radial Blanket Rods. As part of the continuing surveillance program, examinations are being made of material from radial blanket subassemblies in the Rows 6, 7, and 8 to determine the effect of the irradiation which it has received. Adjacent blanket rods are also being examined to estimate the probable effects of increased neutron and thermal fluxes resulting from core expansion. Measurements of cladding OD, uranium slug OD, length, and density have been completed for 3 of 19 rods from 6th row subassembly A-701 (0.26 a/o maximum, 0.09 a/o average estimated burnup) irradiated in position 6A5, and for 4 rods from 7th row subassembly A-760 (0.15 a/o maximum, 0.05 a/o average estimated burnup) irradiated in position 7F4. Similar data are being obtained for an additional 7th row subassembly A-752 (position 7A3) and for 8th row subassembly U-1049 (position 8A5).

Significant radial swelling (diametral increases of up to 28 mils) was observed on the central 3 of 5 slugs from rods contained in the 6th and 7th row subassemblies. Slight increases in length (up to ~0.2 in.) and decreases in density from 18.9 g/cc to as low as about 17.0 g/cc were also observed for these slugs. Measurements of cladding OD indicated up to about a 3-mil increase in diameter in blanket elements on the side of the 6th row blanket subassembly toward the reactor center,

but slug diameters do not indicate that uranium and cladding were in contact at these points, at least in the cold, as-measured condition. Little or no increase in diameter was apparent from cladding measurements on the side of the 6th row subassembly away from the reactor center, or on rods from the 7th or 8th row subassemblies examined.

b. Operations. All 6th row blanket subassemblies, now replaced by fueled subassemblies and irradiation experiments in the expanded core, were cleaned of sodium and stored in the steel-lined pits in the waste burial ground. This provided space in the reactor storage basket for uranium radial blanket subassemblies in Row 7 that were removed and replaced by stainless steel reflector subassemblies. A few 6th row blanket subassemblies were reserved for examination. Experimental subassembly XG06 was dismantled for removal and shipment of irradiation capsules. Process operations were carried on routinely, as summarized in Table I.

TABLE I. Production Summary for February 1967

	February	Total This Year
Subassemblies received:		
Core Control, Safety (for processing)	4	20
Other	25	31
Subassemblies dismantled (for processing)	14	20
Subassemblies dismantled (for examination, etc.)	10	20
Subassemblies fabricated: reprocessed	14	29 ^a
unirradiated	0	4
Reprocessed subassemblies transferred to reactor	7	20
Elements decanned:		
From irradiated subassemblies	1171	
Rejects	0	
Other (list)	0	
Total Decanned	1171	

^aThis includes 15 reported as fabricated in January (see Reactor Development Program Progress Report for January 1967, ANL-7302, p. 23). One of these was actually produced in December, but after the period used for reporting in 1966. Henceforth, all production figures will be for the entire month.

TABLE I (Contd.)

	<u>Melt Refining</u>			
	<u>Irradiated Fuel</u>	<u>Recycle Material</u>	<u>New Fuel</u>	<u>Other (see remarks)</u>
No. of runs	8	8		
Average pour yield, %	91.8	86.9		
	<u>Processing</u>			
Injection-casting runs (total number)				13
Elements processed:				
Accepted	1442			
Rejects	90			
Elements welded:	1418	rewelded: 4		
Elements leak tested:				
Accepted	1411			
Rejected	16			
Elements bonded (including recycle)	1945			
Elements bond tested:				
Accepted	1515			
Rejected	430			
Elements to surveillance:		97 from 10 subassemblies		
	<u>Waste Shipments</u>			
Cans to burial ground	13			13
Oxide and glass scrap to ICP (No. of shipments)				8

Remarks: All of the EBR-II 6th row depleted uranium blanket subassemblies (23 units) were received, washed, temporarily stored, and then routed to the burial storage area. Seven blanket subassemblies were dismantled and received initial surveillance before shipment to TREAT and TAN for additional surveillance.

B. Physics Development

1. ZPR-3

Assembly 48, a 400-liter, plutonium-fueled assembly with a softened neutron spectrum representative of a large power reactor, has been studied for several months. Additional data for U^{235} and U^{238} fission distributions from foil irradiations in the heterogeneity experiments have been obtained.

After completion of the Fast Flux Test Facility (FFTF) experiments in a slightly revised assembly (48A), discussed below in Sect. II.B.1.b, the assembly was changed back to Assembly 48. Radial traverses of material worths were made in the P-row of this assembly. Traverse data were obtained for stainless steel, empty sample carrier, plutonium, uranium-235, depleted uranium, boron-10, tantalum, and sodium. The current measurements are being evaluated and they supplement the axial traverses and the

central worth measurements reported previously. This constituted the final set of measurements for Assembly 48.

a. Foil Irradiations. The relative U^{238} capture and fission rates and U^{235} fission rates were measured at seven positions through the central drawer (P-16) at a distance of 1 in. from the core midplane in the three heterogeneous configurations of Assembly 48. The results are shown in Figs. 1, 2, and 3.

Depleted (0.2%) and enriched (93.1%) uranium foils were irradiated at each position and the U^{238} captures determined by counting the U^{239} gamma activity of the depleted uranium foil as described by Davey.² The fission product activity in depleted uranium is determined in this method and used in conjunction with the fission product activity of the enriched uranium foil, summed over the same gamma energies to determine the relative fission rates of U^{238} and U^{235} .

b. Assembly 48A. The initial set of experiments which were relevant to the analysis and design of the Fast Flux Test Facility (FFTF) by Pacific Northwest Laboratory was completed (see Reactor Development Program Progress Reports for December 1966, ANL-7286, and January 1967, ANL-7302).

Fig. 1. Relative Reaction Rates through the Normal Central Drawer of ZPR-3 Assembly 48

Fig. 2. Relative Reaction Rates through the Bunched Central Drawer of ZPR-3 Assembly 48

²Davey, W. G., The Ratio of U^{238} Captures and U^{235} Fission Cross Sections in Fast Reactors, ANL-7034 (May 1965).

Fig. 3. Relative Reaction Rates through the Unbunched Central Drawer of ZPR-3 Assembly 48

for the various combinations, enrichments, and geometries are given in Table II. The core was loaded as shown in Fig. 4 for these experiments. The nonlinearity of the degree of interaction with enrichment can be seen, as well as the effect of bunching the poison material. The geometry of the poison-rod loadings was the same as shown in the December report. The boron-10 content was varied by changing the ratio of natural to enriched B_4C while retaining the same total volume of B_4C .

The Inconel mockup used here was as close as possible to the composition expected in the FFTF. It contained 18.64 v/o Na, 50.69 v/o Ni, 7.97 v/o Fe, 15.88 v/o Cr, and 6.82 v/o void. The composition of the poison rods was 26.86 v/o Na, 45.27 v/o stainless steel, 21.04 v/o B_4C , and 6.82 v/o void.

(i) Subcritical Measurements of k_{eff} . Further studies were made to determine the effect of the distortion of the flux distribution on the linearity of k_{eff} subcritical measurements. Computations (similar to those described in Progress Report for January 1967, ANL-7302, pp. 27-28) indicated that differences in the

As indicated in the December report, criticality was achieved with two poison-rod mockups loaded into the Inconel reflector at the core edge on opposite sides of the core in positions P-9 and P-23 (see Fig. 4). Radial traverses have been made in the P-row of the assembly with the poison rods in place, as well as in the clean core with no poison rods. Material-worth traverses were made for stainless steel, empty sample carrier, plutonium, depleted uranium, and boron-10 (plus tantalum in the clean core). Reaction-rate traverses were made for plutonium and depleted uranium fission counters, and a BF_3 proportional counter. The material-worth data are still being processed. The reaction rate data for the plutonium and BF_3 counters are shown in Figs. 5 and 6.

The December report indicated measurements were under way to determine the interaction effect between poison rods. These data are now complete and have been processed. The rod worths

Fig. 4. Cross Section of ZPR-3 Loaded for Rod Interaction Experiments

Fig. 5

Radial Profiles of Plutonium Fission Rate through Assembly 48A with and without Mockup B₄C Rods

Fig. 6

Radial Profiles of Boron-10 Reaction Rate through Assembly 48A with and without Mockup B₄C Rods

TABLE II. Measured Worths of B₄C Control Rods
(Inhours $\pm 2\%$ -- 1013.4 Ih per 1% $\Delta k/k$)

Location	B ¹⁰ per Rod (g)			
	200	680	1100	1100 (bunched)
P-9	205.72	321.25	422.59	358.74
P-23 ^a			417.52	
P-9, P-23			832.00	
L-10			397.25	
T-10			387.12	
L-10, T-10			764.10	
L-10, T-10, P-23			1203.92	
L-10, T-10, P-9			1111.67	
N-9		277.67	354.69	
R-9			354.69	
N-9, R-9		539.13	683.03	
N-9, R-9, P-23			1106.63	
N-9, R-9, P-9		763.10	951.58	
P-16 ^b	746.86		1747.36	1529.82

^aP-23 is symmetric with P-9.

^bCore was completely blanketed with U²³⁸ (1002.5 Ih per 1% $\Delta k/k$).

measurements of neutron counting rate result from the distortion of the flux distribution by a highly absorbing region as well as from the attenuation of the flux through that region.

However, the results of the overall investigation indicate that the relation between the inverse of multiplication and Δk subcritical is linear to within 4% for 4% Δk subcritical. Since the series of control-rod experiments performed for the FFTR was maintained to well within 2% Δk subcritical, the errors associated with the inverse multiplication technique are therefore expected to be less than 2%. The error can be taken to be linear to Δk , providing the external counters are not close to shadow of the control rods.

(ii) Preparation of a Cross-section Set. The effort to prepare fine and coarse energy-group cross-section sets continues (see ANL-7302, p. 33). Several 6-group structures have been investigated and are being modified to improve the agreement with reactivity effects obtained with the fine 22-group set. The group structure of the collapsed sets is to be based on several conditions: (1) the set should reproduce the worths of materials having spectral- and leakage-dependent components within some acceptable margin of error; (2) the general energy dependence of the fine-group real flux and adjoint function should be reasonably reproduced by the coarser energy-group structure. The material worths of interest are those used in the design of fast power reactors in addition to the plutonium and uranium fertile and fissile isotopes. This computational technique is, in effect, adjoint weighting the averaged cross sections but is accomplished by doing direct k_{eff} calculations.

c. Assembly 48B. Assembly 48B is being loaded. This will consist of the basic Assembly 48 with a central zone containing plutonium with a higher Pu^{240} content. This zone will be 12 in. in length and approximately 15 in. in diameter. For this series of experiments, 17.2 kg of plutonium with 22% Pu^{240} have been obtained. The average Pu^{240} content of the fuel in this zone will change from 6.0 to 17.6%. A series of experiments which are sensitive to the higher Pu^{240} will be run and a comparison with results from Assembly 48 can then be made.

2. ZPR-6

a. Voiding Measurements. The effects of sodium voiding on the fission densities in U^{235} and U^{238} and the capture density in U^{238} were investigated in Assembly 5. Enriched and depleted uranium foils (93 and 0.2%, respectively) were irradiated in the central drawer. Two irradiations were performed: one with the sodium "in" and another with the sodium "out" from a central region of 25 cm in radius. The sodium was removed from the entire axial height of the core. The foils were placed at intervals of 2 in. along the axial length at approximately the midsection

of the central drawer. The materials adjacent to the foils were 1/8-in.-thick depleted uranium plate on one side and 1/2-in.-thick sodium cans in the case of sodium "in," or 1/2-in.-thick empty SS cans in the case of sodium "out," on the other side. The depleted foils were placed in one-half of the reactor and the enriched foils in the other half. In each irradiation a U^{235} monitor foil was placed at the radial core-reflector interface away from the perturbed region. The relative activities of the fission products were determined by scintillation counting. Coincidence counting was used to determine the relative captures in U^{238} . Absolute determination of the amount of fissions in U^{238} and U^{235} and capture in U^{238} was also performed on a few foils by radiochemical techniques. The results from the radiochemical measurements were used to normalize the results obtained from scintillation counting. The results are shown in Table III; the uncertainty in each number is about 5%.

TABLE III. The Effects of Sodium Voiding on the Absolute Fission Densities in U^{238} and U^{235} and the Capture Density in U^{238}

Z (cm)	Foil No.	Absolute Fissions/g U^{238}		Absolute Captures/g U^{238}		Absolute Fissions/g U^{235}	
		with Na ($\times 10^9$)	w/o Na ($\times 10^9$)	with Na ($\times 10^{10}$)	w/o Na ($\times 10^{10}$)	with Na ($\times 10^{11}$)	w/o Na ($\times 10^{11}$)
	1	3.95	3.94	1.92	1.63	1.387	1.210
5.08	2	3.93	3.92	1.92	1.61	1.374	1.209
10.16	3	3.93	3.93	1.88	1.59	1.377	1.204
15.24	4	3.85	3.86	1.87	1.57	1.350	1.185
20.32	5	3.73	3.65	1.80	1.51	1.295	1.142
25.40	6	3.63	3.54	1.74	1.45	1.246	1.110
30.48	7	3.34	3.40	1.66	1.40	1.193	1.045
35.56	8	3.31	3.20	1.61	1.31	1.126	1.000
40.64	9	2.94	2.95	1.47	1.22	1.047	0.927
45.72	10	2.69	2.69	1.32	1.14	0.975	0.851
50.80	11	2.40	2.42	1.21	1.03	0.869	0.768
55.88	12	2.10	2.12	1.07	0.91	0.847	0.685
60.96	13	1.79	1.76	0.93	0.81	0.662	-
66.04	14	1.37	-	0.76	-	-	-

The results show: (1) the fissions per gram of U^{238} were not affected by the removal of sodium; (2) the captures per unit weight of U^{238} decreased by about 15% when the sodium was voided; (3) the fissions in U^{235} decreased by about 12% upon removal of the sodium.

b. Doppler Measurement. Installation and checkout of the modified ZPR-6 Doppler-measurement equipment in Assembly 5 have been completed. The first sample measured in the present series was the 1-in. natural UO_2 sample. The results were identical with those observed in ZPR-6 Assembly 4Z, within statistics, confirming the reliability of other Doppler measurements made with Assembly 4Z. After appropriate data adjustments, the natural UO_2 Doppler measurements made in the zoned ZPR-9 Assemblies 11 and 12 also agreed with the measurements made on the full-size Assembly 5 within statistics.

3. ZPR-9

Material-sample worths and Doppler coefficients for U^{238} and U^{235} have been measured for Assembly 13. The worth measurements are tabulated in Table IV, together with the corresponding worths calculated by use of the Argonne 801 cross-section set and MACH 1, a one-dimensional diffusion code. Doppler-coefficient calculations for U^{238} and U^{235} , using the same cross-section set and code, indicate that the calculated and experimental results agree within $\sim 10\%$. Since the spectrum in Assembly 13 is considerably softer than a typical fast spectrum, a large fraction of the Doppler effect for both U^{235} and U^{238} comes from the resolved resonance region. These measurements should allow comparisons to be made with theory, without adding the uncertainty of statistical averaging techniques which must be used when working in the unresolved resonance region.

TABLE IV. Central Worths in Assembly 13
(1% = 438.861 lh)

Sample	Weight (g)	SS Can Weight (g)	Net Worth (lh)	Experimental (lh/kg)	Calculated (lh/kg)
Mo	598.8	-	-11.621 ± 0.030	-19.41 ± 0.05	-23.99
Ni	546.1	-	-6.335 ± 0.014	-11.60 ± 0.03	-10.28
Fe	484.3	-	-2.996 ± 0.014	-6.186 ± 0.03	-5.300
Be	114.3	-	$+4.480 \pm 0.007$	$+39.20 \pm 0.06$	+45.47
C	102.0	-	$+1.073 \pm 0.004$	$+10.52 \pm 0.04$	+7.676
Al	167.1	-	-0.191 ± 0.025	-1.143 ± 0.150	-1.212
Na	51.38	56.42	-0.068 ± 0.037	-1.323 ± 0.720	+1.542
W	1108	-	-25.56 ± 0.35	-23.07 ± 0.32	-
Ta	924.7	27.54	-45.35 ± 0.35	-49.04 ± 0.38	-
B	75.2	20.6	-61.95 ± 0.43	-824 ± 6	-4150
Poly	58.988	-	$+25.757 \pm 0.005$	$+436.65 \pm 0.08$	+406.73
Poly	13.70	72.32	$+6.425 \pm 0.043$	$+469.0 \pm 3.1$	+407
Depleted Uranium	1155	-	-9.829 ± 0.006	-8.510 ± 0.005	-13.06
Enriched Uranium	18.24 17.00 U^{235}	- 72.34	$+2.453 \pm 0.010$	$+134.5 \pm 0.5$ $+144.3 \pm 0.6$	+106.9 +115.7 ^a
SS	481.0	-	-3.828 ± 0.014	-7.958 ± 0.029	-6.086
SS Can	72.38	-	-0.651 ± 0.003	-8.994 ± 0.041	-
B ₄ C	59.62	64.70	-42.52 ± 0.35	-713.2 ± 5.9	-3247

^aAdjusted for U^{238} present in the sample.

Assembly 14, the second in a series of four polyethylene softened spectrum assemblies, has replaced Assembly 13. The new assembly has the same volume percent of polyethylene as in Assembly 13 but with

1.75 v/o of U^{238} replaced by carbon. Material-sample worths have been measured (see Table V). Doppler-coefficient measurements are currently being made.

TABLE V. Central Worths in Assembly 14
(1% = 435.691 Ih)

Sample	Weight (g)	SS Can Weight (g)	Net Worth (Ih)	Experimental (Ih/kg)	Calculated (Ih/kg)
Mo	598.8	-	-12.143 ± 0.001	-20.28 ± 0.00	-25.424
Ni	546.1	-	-6.548 ± 0.010	-11.99 ± 0.02	-10.674
Fe	484.3	-	-2.947 ± 0.007	-6.084 ± 0.015	-5.294
Be	114.3	-	$+5.273 \pm 0.013$	$+46.13 \pm 0.11$	+53.360
C	102.0	-	$+1.393 \pm 0.009$	$+13.66 \pm 0.08$	+9.752
Al	167.1	-	-0.054 ± 0.001	-0.325 ± 0.006	-0.2782
Na	51.38	56.42	$+0.006 \pm 0.003$	$+0.108 \pm 0.049$	+2.523
W	1108	-	-30.50 ± 0.35	-27.53 ± 0.32	-
Ta	924.7	27.54	-53.24 ± 0.35	-57.58 ± 0.38	-
B	75.2	20.6	-73.30 ± 0.25	-975 ± 3	-4600
Poly	58.988	-	$+28.75 \pm 0.35$	$+487.39 \pm 5.93$	+459.86
Poly	13.70	72.32	$+7.017 \pm 0.042$	$+512.2 \pm 3.1$	+459.9
Depleted Uranium	1155	-	-10.292 ± 0.014	-8.911 ± 0.012	-14.02
Enriched Uranium	18.17 16.93 U^{235}	72.34	$+2.496 \pm 0.006$	$+137.4 \pm 0.3$ $+147.4 \pm 0.4$	+109.4 +118.5 ^a
SS	481.0	-	-3.877 ± 0.002	-8.059 ± 0.004	-6.179
SS Can	72.38	-	-0.695 ± 0.002	-9.600 ± 0.034	-
B ₄ C	59.62	64.70	-45.13 ± 0.35	-759.0 ± 5.9	-3608

^aAdjusted for U^{238} present in the sample.

4. ZPPR

a. General. The 3-in. concrete topping slab for the main floor of the support wing has been poured. All structural steel and roof decking for the support wing have been placed. The suspect-waste retention tanks and pumps have been installed. All walls of the vault-workroom area have been poured and the precast, prestressed reinforced-concrete panels for the roof have been placed. Fifty-two of the 54 steel caps for the piling posts have been welded on. All the 29 required concrete pile caps have been poured. The first of six concrete pours for the reactor pit and cell floor has been made.

The present status of the reactor components is as follows:

Items Received during February. The particulate air monitor for the mound-area exhaust stack was received without the operating and maintenance manual. Checkout will be delayed until the manual is received. The matrix tie-down parts and the reactor knees were received and accepted. The replacement clutch for the slow speed motor of the reactor table drive was received. The reactor control cable stands were received and accepted.

Matrix Drawers. The remaining 280 front drawers are in the final stage of fabrication.

Poison Safety Rod Drives. The first rod was received at the Idaho site. The rod performed satisfactorily, and the vendor was instructed to fabricate the remaining rod drives.

Personnel Shields. The vendor is ordering materials and is scheduling the fabrication of the personnel shields.

Matrix Alignment. Work is progressing on the plates. The order for the holding fixtures was placed with Gate City Steel Co., Pocatello, Idaho.

Source Drives. The vendor requested a delay in shipping until early April 1967 because of late arrival of purchased items.

Rod Drive Mounting Plates. The first rough machine cut on the newly cast plate was not deep enough to check for defects properly. The plate was returned to the machine shop for proper machining.

Fueled Control and Safety Rod Drawers and Ball Joint Assemblies. The items were rejected and have been returned to the vendor for repairs to conform with the specifications.

Poison Safety Blades and Sheaths. The blades and sheaths were shipped on February 16 from the Scottsonizing plant to the vendor for inspection prior to shipping to Argonne.

Boron Fabrication. The bids have been received and are being reviewed.

Stack Gamma Monitor. The stack gamma monitor is completed except for the audible alarm unit, which is a stock item and has been back-ordered.

b. Programmatic Requirements. The ZPPR experiments will require an estimated 2900 kg of plutonium with 11.5% Pu²⁴⁰ as finished product for fuel elements. An additional requirement is for approximately 190 kg of 25 to 30% Pu²⁴⁰, it is estimated that 500 kg of finished product of 11.5% Pu²⁴⁰ will be required in forms other than metal plates. Originally withdrawal for the starting plutonium requests were made to the AEC Production Division for supplying this total requirement in various fiscal years. It is now apparent that a more thorough analysis of the withdrawal requirements for both quantity and schedule must be prepared. This information will be based upon continuing contracts with two commercial fuel-element fabricators, in addition to a portion of the work being performed at Rocky Flats Division, Dow Chemical Company. This schedule has been prepared and will be forwarded to the AEC.

c. Ingoting of Plutonium Supply Metal. A contract has been arranged with Isochem Inc., Richland, Washington, for the blending of the required plutonium to ANL specifications. A primary requirement is to blend this material to a constant 11.5 w/o Pu²⁴⁰ $\pm$ 0.05 w/o. Isochem has on hand 600 kg of buttons. They have commenced work and estimate that they will complete 200 kg during the month of February and 400 kg during the month of March. This material will be cast into approximately one-kg ingots and supplied to the contracted fuel-element fabricators under an AEC-CH supply agreement.

Isochem will continue producing approximately 200 kg per month through fiscal 1967, provided this total quantity is released to them by the AEC. The first 200 kg of ingots produced will be scheduled for fuel fabrication at Rocky Flats Division, Dow Chemical Company; the next 400 kg will be scheduled for use at NUMEC. ANL representatives are working closely with Isochem during this production period.

d. In-house Facilities. In order to assure a sufficient supply of the fuel elements to meet the programmatic needs for fuel elements containing 1100 kg of plutonium to be shipped to ANL by March 1968 for use in the first experiment in May 1968, it has been decided that an in-house facility should be used for part of the fuel-element fabrication. ANL investigated the possibility of having work done at Isochem, Rocky Flats Division of Dow Chemical Company, or at ANL. ANL recommended selection of Rocky Flats Division, Dow Chemical Company, as the source for the in-house fabrication, and this recommendation was approved by the AEC for the production of fuel elements with 700 kg of total plutonium. Authorization to commence work was given to Rocky Flats. The present budget estimate for the 700 kg is \$1,221,000 (including quality control and nuclear material management), with delivery being completed by March 1968.

Rocky Flats was requested to schedule the delivery of this quantity on a one-shift basis so that if the commercial producers run into

production problems or delivery delays Rocky Flats can produce additional quantities by working added shifts. If necessary, Rocky Flats probably could produce the entire requirements for fuel elements containing 1200 kg of plutonium by March 1968. They have agreed to this proposal and have submitted manufacturing schedules for fuel elements containing either 700 kg or 1200 kg of plutonium to be completed by March 1968.

e. NUMEC. ANL is negotiating with NUMEC for delivery of 555 kg of total plutonium in fuel elements by March 1968. NUMEC is preparing a price breakdown, as well as the prices for extras and adjustments in the bidding assumptions requested by the Laboratory and the Commission in the fuel-element-contract and supply-contract negotiations. Similar information is being prepared on various option quantities. If the contract negotiations are successful with NUMEC, and AEC approval is given, it is expected that NUMEC will produce approximately 50% of the ANL requirements of 2900 kg total plutonium in fuel elements. NUMEC has produced similar fuel elements for the SEFOR critical experiments and is the only commercial organization that has production experience with plutonium metal fuel plates of the type required by ANL. In competing for the contract, NUMEC decided to establish sufficient capability to produce the entire quantity needed within a period less than the Laboratory's final delivery requirements.

The AEC Plutonium Supply Contract for the plutonium to be used in the fabrication of the ZPPR fuel elements is presently being negotiated with NUMEC and it is expected that both fuel-element contract negotiations and supply-contract negotiations can be consummated by March 15, 1967.

f. Second Commercial Source. The AEC has decided that a second commercial source should be available for some portion of the ANL requirements to insure competition, increase commercial experience for plutonium work, and to insure an adequate supply of fuel for ZPPR. The AEC-CH has stated that they will act as the prime contractor in the establishment of the second commercial source and therefore will solicit proposals from various industrial organizations. A selection board has been appointed by AEC-CH to recommend the second commercial supplier and the type of contract to be used.

g. ZPPR Fuel Jackets. ANL has taken the responsibility of furnishing fuel jackets and end plugs to the various fabricators of the fuel elements. This was done in order to insure a uniform quality of both the vacuum-melted metal tubing and the forming of this tubing into the rectangular shapes required. The contract for the tubing has been let to the Pacific Tube Company, with partial delivery estimated in February 1967. However, it appears that the first delivery will be made in April 1967. The special tooling required for the forming of the rectangular shape from the

round tubing has been completed by the ANL Central Shops. This special tooling will be used by Central Shops for forming the rectangular jackets or will be supplied to a commercial vendor if a contract can be successfully negotiated. If necessary, ANL Shops will shape a minimum amount necessary to satisfy the early production requirements for jackets to be used by Rocky Flats.

h. Nonfissile Materials. A contract has been approved for supply of 24,000 kg of depleted uranium plates by the National Lead Company. A small portion of these plates will be delivered during FY 67 with the remainder furnished in FY 68.

A contract has been approved for 70,000 kg of uranium blocks with the National Lead Company, 25% to be delivered in FY 67 and 75% in FY 68. These blocks will be furnished in the "as cast condition" in order to achieve the lowest possible fixed price.

ANL has received a proposal from NUMEC for the fabrication of 195,000 U_3O_8 plates. This contract has been approved, and it is expected that all plates will be delivered in FY 67.

i. Sodium and Sodium Carbonate. ANL will procure the necessary stainless steel jackets for both the sodium and sodium carbonate requirements. Sixty-five thousand feet of stainless steel tubing are needed. The tubing will be formed to the rectangular shape, either commercially or at ANL. Specifications have been written for the jacket tubing, jacket forming, sodium filling for 3500 liters, and sodium carbonate filling of 1500 liters. The tubing procurement should be completed during FY 67, the filling of both sodium and sodium carbonate will be in FY 68. These specifications have been submitted to Idaho for approval.

A quantity of 2 x 2-in. and 1 x 1-in. depleted uranium blocks are being manufactured by the National Lead Company and will be delivered in FY 67.

C. Component Development

1. Sodium Technology and Development

a. Fission-gas Pressure Transducer. Literature reviews and a survey of the state of the art indicate that three basic types of pressure transducers hold greatest promise for application in the Fast Flux Test Facility (FFTF): a null-point system, a capillary (NaK-filled) system with a bellows sensor to strain-gauge output, and a strain-gauge type of sensor.

(i) Null-point System. Atomics International (AI) has developed a null-point pressure transducer for fission-gas measurements. The sensor

is 0.35 in. in diameter, 0.4 in. long, and constructed from two 0.25-in. stainless steel welded bellows with the closed ends adjacent to each other; each open end is welded to the housing. One bellows sees the fission-gas pressure and the other sees the signal gas. With the contact point mounted at the closed end junction of the two bellows, an electrical pickup probe makes contact when increased fission-gas pressure moves the bellows. The probe serves both as a contact device and as a thermocouple, since the temperature is measured at the point of contact. Thus the transducer can be temperature compensated. Accuracy for measuring fission-gas pressure is reported as about 1% at 100 psig. AI has tested this device to 1000°F at 100 psig, and tests are being made under irradiation conditions in a SNAP reactor and in a Canadian reactor in which maximum temperature is 750°F.

Mine Safety Appliance Co. is also developing for ANL a null-point device whose probe is an insulated wire that makes contact with a closed end of a single bellows. Tests at 1200°F and 100 psig are reported to have 2% accuracy.

(ii) NaK-filled System. A bellows-operated, NaK-filled pressure transmitter was developed at ANL for TREAT experiments. Devices of this type have operated up to 1000°F and 300 psig; much of the work has been concerned with pressure pulses and microsecond response times. Tested accuracy was better than 1% full scale. Presumably, this type of device with a 0.25-in. bellows could be operated at 1200°F.

This device will be redesigned for use as a fission-gas pressure transducer. A 20-ft-long capillary tube, which will lengthen response time, will be attached to the bellows and to a strain gauge. The strain gauge could be placed either outside the reactor or in the reactor at a location where temperature and radiation are low.

(iii) Strain-gauge Systems. Tests have continued on a Statham PA812-100 thin-film strain-gauge pressure transducer. The zero drift at about 600°F (see Progress Report for January 1967, ANL-7302, p. 38) was found to be caused by a permanent decrease in the insulation resistance between the strain-gauge bridge and transducer body. At termination of the initial test, the insulation resistance had dropped from over 200 megohms to 3 kilohms, but transducer sensitivity remained excellent. Destructive examination indicated that slight reorientation of internal wires could have caused the problem. The components are being tested further to learn whether this condition was, in fact, the cause of the zero-shift drift at the high temperature.

b. Method for Locating Failed Fuel. To find a method for determining which subassembly within a shutdown reactor core has one or more failed fuel pins, the feasibility of sensing the fission-gas release

from the subassembly will be investigated. To prove the principle, a scheme will be demonstrated for efficiently disengaging noble gases from a continuously circulating liquid-sodium system at 1200°F. The equipment will include devices for disengaging the gas, trapping sodium vapor, viewing, monitoring radiation, and for injecting gas.

Tests with a transparent model, in which helium and water simulate fission gas and sodium, show that an under-sodium gas pickup tube is not likely to improve the sensitivity of fission-gas accumulation to the inlet of a sodium-inert gas vapor trap.

Modifications have begun on an existing 1200°F sodium-sump tank and sodium-vapor trap assembly that had been used to measure vapor-trap efficiencies. Small amounts of helium will be injected into the region of argon above the 1200°F sodium-free surface, and a helium detector will be used to determine the smallest quantity of helium which can be detected.

c. Signal Lead Connectors for Sodium Service. To develop remote connectors for electrical and fluid signal leads for in-core instrumentation, connector lead-wire assembly designs and test procedures will be reviewed, changes made to meet FFTF design criteria, new connector concepts considered, out-of-pile test programs established, and methods for terminating the sensor leads inside the reactor vessel, both above and within the sodium, studied and developed.

The renovation and reassembly of the gasket and connector test equipment are nearly complete; a strip-chart recorder was added to the system to monitor the thermocouples continuously.

d. In-core Flowmeter. So that sodium flow through individual subassemblies or process tubes can be measured, technology will be established to assure that small magnetic, turbine, or other types of flowmeters can be obtained for use in severe temperature and radiation environments. To demonstrate performance and reliability, prototype flowmeters will be tested out-of-pile and in-pile.

Initial evaluation of flowmeters and preparation of the preliminary status report are continuing.

Thermal tests of Alnico-V magnetic material are continuing. A rod magnet having a length-to-diameter (L/D) ratio of 6 was thermally stabilized by the procedure outlined previously (see Progress Report for January 1967, ANL-7302, pp. 39-40). However, after the third thermal cycle, noticeable changes in remanence were still occurring. This contrasts with the results obtained from cylindrical flowmeter magnets that have an equivalent L/D of 2.15, where no changes in remanence could be noted after the second thermal cycle. The supposedly stabilized rod magnet

has accumulated 270 hr at 1200°F. When checked at room temperature, the remanence had not changed during the first ~150 hr; although a slight decrease was measured after 270 hr, it is within the gaussmeter accuracy. Further aging of this magnet is continuing.

A second rod magnet of the same type will be given sufficient initial thermal cycles that changes in remanence will not be detectable after the second-from-the-last thermal cycle.

A magnetometer is being procured for continuous and more precise measurements of magnetic moment at high temperatures; it will be used for both pre- and post-irradiation tests. Design has begun of all other necessary equipment, such as the furnace, rotating mechanism, and controls.

Attempts to purchase stock rod magnets of Alnico-V, -VI, and -VIII from seven magnet vendors have been unsuccessful.

Drawings are being prepared for modifying the sodium-flowmeter calibration loop so its operating temperature can be raised from 900 to 1200°F; materials are being ordered.

The ceramic electrical insulators were received, conditioned, and used to complete fabrication and assembly of a preliminary prototype magnetic flowmeter. The prototype will be installed in the calibration loop, and long-term signal stability will be investigated.

For irradiation tests of magnet samples, it has been decided to adapt an EBR-II Mark B7 subassembly. Ranges of test magnet sizes and L/D ratios have been selected:

Diameter (in.)	Length (in.)	L/D	Volume (in. ³ /π)
1/4	2	8	0.032
3/8	1½	4	0.053
1/2	1	2	0.063
5/8	5/8	1	0.061

Rod magnets were selected because they should be readily available, inexpensive, and easily installed in the irradiation capsules. Plans have begun for the arrangement of samples in individual irradiation capsules. Calculations are continuing of both neutron-induced gamma activity and heating in sample magnets of Alnico-V, -VI, and -VIII. The gamma activity will determine the hot-cell required for postirradiation examination and tests; the gamma heating will influence the irradiation-capsule design required to achieve test temperatures.

e. Fuel-pin Thermocouple. Thermocouples are being developed to indicate fuel-pin temperatures up to 2800°C. For this development, previous experience with fuel-pin thermocouples is being evaluated, technology will be established to assure satisfactory design and performance, thermocouple assemblies and materials will be tested and evaluated, extensively, in-pile and out-of-pile tests will be conducted, and thermocouple specifications and fabrication techniques will be formulated so that bids can be obtained for sensors developed.

A preliminary report on previous experience is being prepared.

High-fired 99.9% thoria insulators have been received; tests will begin after a spectrochemical analysis is received.

Crushable 99.8% thoria insulators have been tested up to 2400°C with the results shown in Fig. 7. The initial samples shorted out at ~2400°C because the thoria had crystallized, forming voids in the insulator. Within some of these voids were "fingers" of pure tantalum, creating a low-

resistance circuit between the center wire and the outer sheath. (This type of failure has also occurred with some thermocouple assemblies having a similar type of thoria.) The failure is believed to be attributable to the welding procedure used during manufacture. Residual oxygen or water vapor enters and remains in the insulators during manufacture; during welding, tantalum oxide forms locally at the hot spots. Then, when the test specimens are heated in an inert gas, the tantalum oxide migrates and decomposes, leaving a pure tantalum bridge in the voided areas of the coaxial resistivity probe and causing a short circuit. Tests are planned to verify this postulated mode of failure.

For study of the compatibility of tantalum and W-25% Re with UO_2 , further test specimens are being assembled; they will be tested in stagnant helium and argon at ~1 atm.

For preliminary in-pile tests, the following reactors are being considered: the Materials Test Reactor (MTR), Engineering Test Reactor (ETR), CP-5 Reactor, High Flux Isotope Reactor (HFIR), and the Brookhaven High Flux Beam Reactor (HFBR).

Fig. 7. Resistivity of Crushable Thoria (99.8% pure)

D. Fuel Development

1. Metal Fuels

a. Irradiation of Uranium-Plutonium Alloys. Sixteen encapsulated fuel elements with U-Pu-Zr pins severally jacketed in V-20 w/o Ti, Types 304 and 316 stainless steel, and Hastelloys -X and -X-280 were non-destructively examined last month after operation at maximum jacket temperatures of 630°C to burnups of 4.7 to 5.1 a/o. All fuel elements were intact and essentially unchanged in diameter, but the fuel pins had increased in length an average of 1.9%, had become elevated about 0.20 in. above the bottom closures, and had, in most cases, sustained one or two separations. There was an average of 0.21-in. bowing of 40-in. capsules and 0.12-in. bowing of the fuel elements inside. The void volume had become redistributed as fine porosity in the sodium.

Postirradiation examinations have continued this month of these fuel elements which had been removed from the EBR-II irradiation subassembly XA07. These elements, which measured 34 in. in length, were fueled with U-15 w/o Pu-9 w/o Zr or U-14 w/o Pu-12 w/o Zr. The transverse macrosections of the fuel showed distinct bands thought to be correlated with the phase transformations of this alloy.

Five elements of this group will not be destructively examined, but are being withheld for transient irradiations in the TREAT reactor as well as elevated-temperature furnace tests.

The elements ND23 and NC23, which were being irradiated in subassembly XG06, are being disassembled at ANL-Idaho in preparation for shipment to ANL-Illinois. These elements are U-Pu-Zr and U-Pu-Ti jacketed in V-20 w/o Ti tubing. They had been irradiated at maximum jacket temperatures of 540°C and had reached a final estimated burnup of 5.0 a/o. Two other elements remain in the reactor in subassembly XG05; they are similar to those removed and will be irradiated to an estimated 7.5 a/o burnup. Additional details on the design of the fuel rods and the irradiation conditions are summarized in Table VI.

TABLE VI. Status of Metal-fuel (Clad in V-20 w/o Ti) Irradiations in Progress in EBR-II

Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions			
		Fuel Composition (w/o)	Effective Density (%)	Cladding OD (in.)	Cladding Thickness (in.)	Max kW/ft	Max Cladding Temp (°C)	Burnup to Date	
								a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ ^a
XG06	ND23	U-15 Pu-10 Zr	63.9	0.209	0.016	10.0	540	4.9	11.7
XG05	ND24	U-15 Pu-10 Zr	63.6	0.208	0.015	10.0	535	4.9	11.8
XG05	NC17	U-15 Pu-10 Ti	66.7	0.204	0.016	10.0	540	5.1	11.3
XG06	NC23	U-15 Pu-10 Ti	63.3	0.209	0.016	10.0	540	5.0	11.1

^aBased on effective density.

2. Oxide and Carbide Fuels

a. Irradiations of Ceramic and Cermet Fuels in EBR-II. Mixed-oxide fuel specimens received for examination in January were reported (see Progress Report for January 1967, ANL-7302, p. 48) to include two of $\text{UO}_2\text{-20 w/o PuO}_2$ jacketed in Type 304 stainless steel and irradiated at maximum jacket temperatures of 555 and 565°C to corresponding burnups of 2.7 and 2.9 a/o (U + Pu). Axial growth of the oxide fuel columns, which had axial center voids, 0.0625 in. in diameter, was about 1%. Postirradiation examination of the two mixed-oxide elements continued this month in the Metallurgy Alpha-Gamma Hot Cell. Additional elements remained in the EBR-II for further irradiation, as listed in Tables VII and VIII.

TABLE VII. Status of Irradiations of $\text{UO}_2\text{-20 w/o PuO}_2$ in Capsule X011 in EBR-II

Specimen Number	Design Parameters				Operating Conditions			
	Effective Density (%)	Cladding Composition	Cladding OD (in.)	Cladding Thickness (in.)	Max kW/ft	Max Cladding Temp (°C)	Burnup to Date	
							a/o (U + Pu)	fiss/cc x 10 ^{-20a}
SOV-3	83.1	304 SS	0.296	0.021	24.2	610	3.2	6.6
SOV-7	85.1	304 SS	0.296	0.021	26.1	630	3.1	6.5
SOV-1	79.9	304 SS	0.296	0.021	22.2	590	3.1	6.1
HOV-10	84.8	Hastelloy-X	0.296	0.016	26.4	615	3.0	6.3
HOV-15	79.8	Hastelloy-X	0.295	0.016	23.5	590	3.0	6.0
HOV-4	80.0	Hastelloy-X	0.295	0.016	25.1	600	3.1	6.2
TVOV-1	76.8	V-20 w/o Ti	0.298	0.022	23.2	575	3.1	5.8

^aBased on effective density.

TABLE VIII. Status of Type 304 Stainless Steel-clad Cermet Fuel Irradiations in Capsule X011 in EBR-II

Specimen Number	Design Parameters				Operating Conditions			
	Fuel Composition (w/o)	Effective Density (%)	Cladding OD (in.)	Cladding Thickness (in.)	Max kW/ft	Max Cladding Temp (°C)	Burnup to Date	
							a/o (U + Pu)	fiss/cc x 10 ^{-20a}
5P-9	SS-40PuO ₂	98.0	0.301	0.015	12.0	495	3.7	2.8
5P-12	SS-27PuO ₂	98.7	0.294	0.015	7.9	450	3.7	1.8
5U-14	SS-27UO ₂	97.9	0.298	0.013	6.2	435	2.8	1.4

^aBased on effective density.

Elements undergoing examination were mixed oxide-elements SOV-5 and SOV-6. A description of these elements and their irradiation histories are given in Table IX.

TABLE IX. ANL Ceramic-fuel Irradiations Removed from EBR-II Experimental Subassembly X009 for Examination

Specimen Number	Design Parameters					Operating Conditions			
	Fuel Composition (w/o)	Effective Density (%)	Cladding Composition	Cladding OD (in.)	Cladding Thickness (in.)	Max kW/ft	Max Cladding Temp (°C)	Estimated Burnup	
								a/o (U + Pu)	fiss/cc x 10 ^{-20a}
SOV-5	$\text{UO}_2\text{-20 PuO}_2$	83.4	304 SS	0.296	0.021	19.0	595	2.7	5.6
SOV-6	$\text{UO}_2\text{-20 PuO}_2$	83.3	304 SS	0.296	0.021	19.8	605	2.9	6.0
SMV-1	UC-20 PuC	80.0	316 SS	0.306	0.025	25.3	675	2.7	7.2
SMP-1	UC-20 PuC	81.4	316 SS	0.306	0.024	18.9	600	2.1	5.6
VMV-1	UC-20 PuC	85.9	Vanadium	0.301	0.024	29.5	680	3.1	8.9

^aBased on effective density.

Elements SOV-5 and SOV-6 contained vibratorily compacted hyperstoichiometric powders derived from Dynapak material supplied by Battelle Northwest Laboratory. Both elements were examined by neutron radiography before removal from the capsules. Destructive tests are scheduled only for SOV-6, because of similarity in irradiation conditions and in postirradiation appearance. Rod No. SOV-5 has been kept intact for anticipated transient irradiations in the TREAT reactor.

The stainless steel jackets of both elements were intact, no fission-product activity being detected in the irradiation capsules. The physical appearance of the jackets was good. Density decreases of jacketed fuel columns were less than 0.03%, and dimension measurements disclosed no significant changes. The neutron radiographs indicated that fuel columns within the jackets elongated about 0.06 in., or 0.54%.

Fission-gas release in element SOV-6 was approximately 65% of the amount produced. The corresponding element plenum pressure at operating temperature at the end of the irradiation was calculated to be 224 psi.

The diameter of the axial cavity in SOV-6 (previously noted in neutron radiographs of both elements) was 0.070 to 0.074 in., as measured on metallographic cross sections. Figure 8 illustrates a typical cross section showing the axial cavity and circumferential region of columnar grains. An outer region of partially sintered material has two zones in which grain growth occurred to a limited degree in the inner zone. The alpha autoradiograph shows no evidence of plutonium migration. A region of higher density adjacent to the cavity was visible on the neutron radiographs of SOV-5 and SOV-6. Fuel densification during formation of the axial cavity can account for the band on the neutron radiograph.

Fig. 2. Cross Section of Element SOV-6

Metallic inclusions were observed in apparently random locations in the mixed-oxide structure in a cross section at the midplane. A concentration of inclusions of similar appearance was found at the bottom of the axial cavity at the lower end of the fuel rod, at a location corresponding to a peak in a gamma scan for ruthenium-103 activity. Increased density had also been observed in the neutron radiographs of both elements in the same area.

b. Mixed-carbide Fuel Irradiations in EBR-II. Of three specimens of UC-20 w/o PuC received in January for examination, two were jacketed in Type 316 stainless steel and irradiated at maximum jacket temperatures of 570 and 640°C, respectively, to corresponding burnups of 2.7 a/o and 2.1 a/o (U+Pu). The third specimen was jacketed in vanadium and irradiated at 640°C to a burnup of 3.1 a/o. All elements were intact.

Postirradiation examination of three mixed-carbide elements continued. An additional 15 mixed-carbide elements, as shown in Table X, remained in EBR-II for further irradiation.

TABLE X. Status of UC-20 w/o PuC Fuel Irradiations in Progress in EBR-II

Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions			
		Effective Density (%)	Cladding Composition	Cladding OD (in.)	Cladding Thickness (in.)	Max kW/ft	Max Cladding Temp (°C)	Burnup to Date	
								a/o (U + Pu)	fiss/cc x 10 ^{-20(a)}
XG05	SMV-2	83.8	304 SS	0.296	0.021	21.6	645	4.8	13.2
XG05	HMV-5	80.0	Hastelloy-X	0.295	0.015	24.5	670	5.0	13.1
XG05	NMV-11	82.9	Nb-1 w/o Zr	0.284	0.012	22.3	645	5.0	13.7
XO08	NMP-2	81.6	Nb-1 w/o Zr	0.284	0.013	18.6	545	3.0	8.2
XO08	NMV-4	80.0	Nb-1 w/o Zr	0.283	0.013	27.9	635	4.4	11.6
XO08	NMV-7	80.0	Nb-1 w/o Zr	0.283	0.013	24.7	605	4.0	10.6
XO08	NMV-12	85.5	Nb-1 w/o Zr	0.284	0.014	27.7	635	4.4	12.4
XO08	HMV-1	80.0	Hastelloy-X	0.295	0.015	25.3	640	3.9	10.4
XO08	HMV-4	80.0	Hastelloy-X	0.295	0.016	27.9	670	4.3	11.4
XO08	HWMP-1	81.4	Hastelloy-X + W	0.296	0.020	17.9	555	3.0	8.1
XO08	HWMP-1	82.9	Hastelloy-X + W	0.295	0.018	27.4	685	4.4	12.0
XO15	NMP-1	81.6	Nb-1 w/o Zr	0.284	0.013	17.6	535	0.50	1.3
XO15	NMV-3	80.0	Nb-1 w/o Zr	0.284	0.013	27.1	625	0.74	2.0
XO15	HMV-2	80.0	Hastelloy-X	0.295	0.015	18.5	670	0.81	2.1
XO15	TVMV-1	83.7	V-20 w/o Ti	0.298	0.021	23.8	620	0.65	1.8

^aBased on effective density.

The three mixed-carbide elements undergoing examination contained vibratorily compacted physically mixed powders (SMV-1), vibratorily compacted solid-solution powder (VMV-1), and pelletized solid-solution powder (SMP-1). Further details, including irradiation histories of these elements, are given in Table IX. The Type 316 stainless steel and unalloyed vanadium jacket materials maintained their integrity as shown by neutron radiography prior to capsule opening and by the absence of fission-product gases in the irradiation capsules.

Dimensional changes of the elements with stainless steel jackets were insignificant. The vanadium-jacketed element, however, elongated 0.31% and the diameter enlarged 0.83%. Postirradiation neutron radiography

shows that the vibratorily compacted fuel column in the vanadium jacket has deflected the restrainer rod. The density of the vanadium-jacketed element reflected the dimension changes by decreasing 1.46%. Density decreases of the stainless steel-jacketed elements were of the order of only 0.3%.

Only the pellet element, SMP-1, has been punctured for collection of plenum gases. Preliminary estimates indicate that fission-gas release from the mixed-carbide pellet fuel was less than 2% of the amount theoretically generated.

c. Fluidized-bed Process for Preparation of (U,Pu)C. A series of fifteen runs was completed in which (U, 15 w/o Pu)C was prepared in a

TABLE XI. Analysis of (U,Pu)C
Prepared by Fluidized-bed Technique
(Run conditions: 6 v/o CH₄-94 v/o H₂,
2 atm pressure, 750-800°C)

	Run Number	
	14	15
Carbon (w/o)	4.95	4.66
Oxygen (ppm)	270	290
Nitrogen (ppm)	360	150
Hydrogen (ppm)	30	>200
Product Weight (g)	316	630

fluidized-bed reactor on a 300- to 600-g scale (see Progress Report for December 1966, ANL-7286, p. 26). In this series of runs, 3/8-in.-dia rods were hydrided to form a powder, which was then carbided by a CH₄-H₂ gas stream at 550 to 800°C.

The product analysis for the last two runs is given in Table XI. The carbon content of the final products of these runs was estimated to be about 4.78 w/o by the method described in ANL-7286, p. 26. This result differed by -0.17 w/o and +0.12 w/o from the chemical analyses of the products in Runs 14 and 15, respectively (see Table XI). In repeated runs under identical conditions, it should be possible to control the carbon content of the product to within 0.10 w/o and to obtain even closer control by blending of batches. The low oxygen and nitrogen contents of the products for these runs are typical for the fluidized-bed technique. When carbiding was complete, the products of both runs were purged with helium at 750 to 800°C to remove hydrogen. The removal of hydrogen was found to be more efficient in Run 14 than in Run 15.

The results of these runs, together with those reported previously, demonstrate that a high-purity product of closely controlled stoichiometry can be produced by the fluidized-bed technique.

Efforts are now being directed at preparing facilities for pressing and sintering fuel pellets required for irradiation testing. The work will be done in a high-purity helium glovebox that is connected to the box in which the fluidized-bed synthesis will be carried out.

Knowledge of the reactivity of the fluidized-bed carbide powder with the glovebox atmosphere is important because the powder will be exposed to the glovebox atmosphere during subsequent fabrication operations. To test the reactivity of the fluidized-bed carbide, samples of (U,Pu)C powder were placed on an analytical balance and exposed to a glovebox atmosphere of helium. The change in weight was noted over a period of time. To duplicate the atmospheric conditions prevailing in the glovebox during handling of carbide powder, short-term tests were made in which the (U,Pu)C powder was exposed to the purest helium atmosphere attainable in the glovebox system (1 ppm H_2O and 3 ppm O_2). There was no weight gain in two separate tests of 3.2- and 7.5-hr duration for the powder from Run 14, and only a small weight gain in a 3.0-hr test for the powder from Run 15.

Longer tests were made in which the samples were exposed to a helium atmosphere which was less pure; it contained between 30 and 400 ppm oxygen, and about the same water content (~ 1 ppm) as in the atmosphere used in the shorter runs.

Again, the weight gain for the product from Run 14 was much less than that for the product from Run 15; after five days, the weight gain for Run 14 was 0.06 w/o, whereas for Run 15 it was about 0.20 w/o. The low reactivity of the carbide from Run 14 may have been due to its low hydrogen content. The (U,Pu)C powder from Run 14 contained only 30 ppm hydrogen, and that from Run 15, which reacted more rapidly, contained more than 200 ppm hydrogen (Table XI). However, the difference in carbon contents of the specimens may also have been a factor in the difference in reactivities.

The role of hydrogen in the reaction of impurities in the helium atmosphere will be investigated in future work. However, it has been demonstrated that (U,Pu)C containing even as much as 200 ppm hydrogen could be exposed to the glovebox atmosphere for a short time without significant contamination.

3. Fuel Cladding and Structure

a. Development of Refractory-metal Alloys for Service in Oxygen-contaminated Sodium. A continuing (refreshed autoclave) test was reported (see Progress Report for January 1967, ANL-7302, p. 51) in which selected vanadium alloys had gained weight in sodium at 550°C (cold trapped at 120°). After six weeks of exposure, only weight gains were found. After eight weeks it was apparent that the thin corrosion coating on the specimens was partially lost. This was noted particularly for V-20 w/o Ti. Apparently, some in-leakage of oxygen had occurred, since the analysis of sodium taken at that

time showed 10.6 wt ppm oxygen as contrasted with previously obtained values of about 9. This slight rise in oxygen level seemed to have triggered the change in corrosion behavior.

The test was continued for an additional two examinations (over four weeks) to see if the weight-gain behavior would be re-established. The specimens continued to lose weight at low rates. The total penetration of the alloys will now be evaluated by microscopic examination and micro-hardness traverses.

A dynamic test is under way at 550°C in sodium flowing at 6.1 m/sec (cold trapped at 120°C). The alloys exposed are the same as those previously tested in the 650°C experiment: V, V-5 w/o Cr, V-20 w/o Ti, and V-15 w/o Ti-7.5 w/o Cr (as cold rolled and annealed). The same order of resistance is noted at 550 as at 650°C, but the spread of values is much less (see Table XII).

TABLE XII. Corrosion of Vanadium Alloys at 550°C
in Sodium Flowing at 6.1 m/sec

Alloy	Weight Gain (mg/cm ²) at Indicated Time (days)		
	6.9	20.9	53.8
V	0	0.06	0.08
V-5 w/o Cr	0	0.032	0.064
	0	0.043	0.065
V-20 w/o Ti	0.089	0.20	0.25
	0.079	0.21	0.26
V-15 w/o Ti-7.5 w/o Cr	0.077	0.15	0.22
	0.088	0.17	0.22
Oxygen Level	-	7.8 ppm	9.7 ppm

The various testing systems are being modified for in-place vacuum distillation analysis of the sodium for oxygen. A large vacuum manifold has been installed and analyses will be attempted shortly. For a period of time, both the distillation method and the mercury amalgamation method will be used simultaneously to permit comparison of values.

b. In-reactor Corrosion of Jacket and Structural Materials by Sodium. The first three groups of twelve cylindrical specimens were removed from the EBR-II core after approximately seven months of sodium exposure (see Progress Report for January 1967, ANL-7302, pp. 51-53). During this period, the reactor was at full power for 119 days,

and these cylinders, of 9.5 mm dia by 19.2 mm long (5.73 sq cm surface area), were exposed to reactor sodium at 510 or 530°C and 3.7-m/sec velocity. The structural materials--Type 304 stainless steel (980°C annealed), Incoloy 800 (980°C annealed), Hastelloy-X (hot rolled) and Inconel 600 (900°C annealed)--showed neither measurable weight change (<0.05 mg/cm²) nor dimensional change (<0.1 mm). In the case of jacket materials, the unalloyed vanadium (950°C annealed) was severely pitted, with a dimensional change of 2 mm in diameter and approximately 50% weight loss (620 mg/cm²). However, the V-20 w/o Ti and V-40 w/o Ti had small weight losses (4 to 12 mg/cm²), amounting to dimensional changes of approximately 0.5 mil per surface. The results are summarized in Table XIII. Micro-examination of these two vanadium alloys indicated the absence of a hardened layer on the surface. A hardened layer is normally observed in all ex-reactor experiments (in the case where weight losses as well as weight gains are encountered), and when this brittle layer reaches appreciable thickness it can degrade the stress-rupture properties of these alloys.

TABLE XIII. EBR-II Corrosion Data

Capsule No.	VMV-1		SMV-1		SMP-1	
Group No.	V		IV		III	
Sodium at Full Power						
Temperature (°C)	530		510		530	
Velocity (m/sec)	3.7		3.7		3.7	
	Specimen No.	Wt Loss (mg/cm ²)	Specimen No.	Wt Loss (mg/cm ²)	Specimen No.	Wt Loss (mg/cm ²)
Material						
Vanadium (950°C Ann)	D6	620	-	-	-	-
V-20 w/o Ti (900°C Ann)	-	-	H5	5.9	H1	12.2
V-40 w/o Ti (900°C Ann)	T3	10.4	T1	3.7	-	-

The potential importance of this difference between in-reactor and ex-reactor behavior of vanadium alloys in liquid sodium is obvious, and the possible reasons are being investigated. The initial result of a seven-day test in ex-reactor sodium (550°C and ~10 wt ppm oxygen) shows that a hardened layer is formed on specimens that had been previously exposed to in-reactor sodium. The results of the seven-day test are summarized in Table XIV; longer exposures are in progress.

c. Effect of Mechanical Properties on Nickel-base Alloys in 650°C Sodium. The Division of Reactor Development and Technology, AEC, requested the Applied Corrosion Group of the Metallurgy Division to evaluate the behavior in sodium of welded Incoloy 800 as a candidate heat-exchanger material for LMFBR. AEC needs a Ni-Cr-Fe weld-metal that is resistant to stress-corrosion cracking in coolant water at approximately 350°C and at the same time has acceptable corrosion, stress rupture, and

creep properties in reactor sodium at about 650°C. Several grades of Incoloy 800 weld-metal meet the stress corrosion requirement in coolant water, but behavior in 650°C sodium has not been determined.

TABLE XIV. Microhardness of EBR-II Specimens

Material	Exposure	Microhardness--Knoop (15-g Load)					Hardened Layer
		Distance from Edge @					
		5 μ	10 μ	15 μ	25 μ	75 μ	
V-20 w/o Ti (H1)	EBR-II						None
V-20 w/o Ti (H5)	EBR-II						None
V-40 w/o Ti (T1)	EBR-II						None
V-40 w/o Ti (T3)	EBR-II						None
V-20 w/o Ti (H1)	EBR-II + 7 days in ex-reactor sodium	1481	637				10 μ thick
V-40 w/o Ti (T1)		508				<5 μ thick	

Creep tests at 650°C on tensile specimens of 0.125-in. dia and 1.5-in. gauge length are to be conducted in sodium (<20 wt ppm oxygen) and in vacuum for comparison. Since Nimonic 80A (21 w/o Cr, 1 w/o Al, 2.5 w/o Ti, and remainder Ni) is known to be susceptible to degradation of creep properties by sodium,³ this material was selected as the first material to be tested in order to demonstrate that the proposed experimental procedure was capable of detecting differences in behavior. The control specimens were given the standard double-anneal heat treatment: 1080°C/8 hr/air-cooled and 700°C/16 hr/air cooled. Preliminary results indicated that Nimonic 80A, stressed to 46.2 kg/mm², prematurely ruptured after about 228 hr in 650°C sodium, whereas a corresponding specimen has not yet ruptured after 575 hr in 650°C vacuum. We are now ready to evaluate the behavior of welded Incoloy 800 in sodium by the same experimental technique.

4. Fuel Reprocessing

a. Processes for Fast Reactor Fuels. Pyrochemical processing of fast reactor fuels of the ceramic type (e.g., oxide or carbide) clad with stainless steel is being investigated. A conceptual flowsheet currently under investigation has been described previously (see Progress Report for September 1966, ANL-7255, pp. 30-31). The process utilizes liquid

³ Nikitin, V. I., Adsorption-induced Effect of Liquid Metals on the Deformation Process of Solid Metals, Fizika metallov i metallovedeniye 15, No. 6, 900-907 (1963).

metal-molten salt extractions and salt-transport separations⁴ for the separation of fissile and fertile constituents of the fuel from the fission products. Recent laboratory and engineering investigations of the current flowsheet are discussed below.

The first step after the decladding of mixed oxide ($\text{PuO}_2\text{-UO}_2$) fuels is the reduction of the oxide to metal (plutonium and uranium) by a Cu-33 w/o Mg alloy in contact with a molten salt (see ANL-7255, pp. 30 and 31). The alkali, alkaline earth, and rare earth fission products and the solid MgO formed as a byproduct of the reduction reaction remain in the waste salt. The plutonium, uranium, and noble and refractory-metal fission products remain in the copper-magnesium alloy; the plutonium is in solution and the uranium is present as a precipitated metal phase. Since consideration must be given to the possible coprecipitation of fission products with uranium, a laboratory experiment was carried out to determine

Fig. 9. Distribution of Cerium between 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl Salt and Zinc-Magnesium Alloys at 600, 700, and 800°C

if zirconium would coprecipitate with uranium in the reduction step. In this experiment, U_3O_8 in the form of small-diameter pins⁵ was reduced by liquid Cu-Mg alloy which contained dissolved zirconium. The reductions were carried out at 600, 700, and 800°C (temperatures at which the solid phases present are α -, β -, and γ -uranium, respectively). The results indicate that zirconium does not coprecipitate significantly with β - or γ -uranium, but that it may do so with α -uranium. During this study the solubilities of uranium in liquid Cu-33 w/o Mg alloy were found to be 0.023, 0.076, and 0.22 w/o at 600, 700, and 800°C, respectively. Distribution coefficients for cerium ($K_d = \text{w/o Ce in salt} / \text{w/o Ce in metal}$) between liquid Mg-Zn alloys and molten 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl were measured at 600, 700, and 800°C over a range of magnesium concentrations in the metal phase. The data, which are presented in Fig. 9, show that the distribution coefficient is temperature dependent at low magnesium concentrations,

⁴ Salt-transport separations are based upon the selective transfer of fissile and fertile materials from one liquid-metal solution (donor) to another (acceptor) by cycling a molten salt phase which acts as a carrier between the two metal solutions. Noble and refractory-metal fission products remain in the donor alloy.

⁵ The pins were cast from U_3O_8 which had been pretreated with a salt mixture composed of 15 m/o MgCl_2 -37.5 m/o CaCl_2 -42.5 m/o NaCl -5 m/o CaF_2 .

whereas at high magnesium concentrations temperature has little effect on the distribution coefficient. These data indicate that any trace amounts of cerium not removed in the preceding oxide-reduction step would follow the plutonium in the salt-transport step. The data obtained in this study will also be used to estimate the thermodynamic properties of dilute solutions of cerium in liquid magnesium.

A laboratory-scale salt transport experiment using newly developed apparatus that minimizes the possibility of cross contamination

Fig. 10. Salt-transport Apparatus:
Double Crucible and Agitators

between the donor alloy and the acceptor alloy was completed. The apparatus (see Fig. 10), consisting of a double crucible and agitators, is installed in an automated rocking furnace. Within the furnace, the molten salt and liquid donor alloy are stirred together and allowed to settle. The salt is then top-poured into an adjacent crucible, which contains the acceptor alloy, by tilting the furnace. After the salt and the acceptor alloy are stirred and the two phases are allowed to separate, the salt is top-poured back into the crucible containing the donor alloy. This sequence of operations can be repeated as many times as needed to effect the desired transfer of the material from the donor alloy to the acceptor alloy. In the salt-transport experiment which utilized this technique, cerium (used as a stand-in for plutonium) was transferred from a Cu-33 w/o Mg donor alloy through 50 m/o MgCl_2 -30 m/o NaCl-20 m/o KCl to a Zn-5 w/o Mg acceptor alloy. After five cycles, 93% of the cerium charged to the Cu-Mg alloy had been transferred to the Zn-Mg alloy. It is estimated that an additional five cycles would be needed to transfer 99.5% of the cerium. The copper content of the Zn-Mg alloy remained constant at about 0.088 w/o during the last three cycles, thereby indicating that copper transfer to the acceptor alloys is very low with this procedure.

Further laboratory work on the solubility of liquid magnesium in molten salts has been carried out (see Progress Report for December 1966, ANL-7286, p. 32). When magnesium metal reacts with MgCl_2 , a "reduced magnesium" species is formed; this species might be either Mg^+ or Mg_2^{2+} . To determine which species is formed, the concentration of

"reduced magnesium" was measured at 800°C in molten MgCl_2 which was in contact with liquid Cu-Mg alloys of different compositions. If Mg^+ is the reduced species the square of its activity in the molten salt would be directly proportional to the activity of magnesium in the alloy; if Mg_2^{2+} is the reduced species its activity in the molten salt would be directly proportional to the activity of magnesium in the alloy. The magnesium activity in Cu-Mg alloys is known.⁶ The activity of the reduced species is assumed to be proportional to its concentration in the MgCl_2 . Since the concentration of the reduced species in the molten MgCl_2 was directly proportional to the activity of magnesium in the alloy, the reduced species appears to be Mg_2^{2+} .

Engineering studies are being carried out to develop the technology required for plant-scale application of pyrochemical processes.

In one study, a stainless steel loop utilized for testing equipment (see Progress Report for December 1966, ANL-7286, p. 33) has been operated for an additional 328 hr using a molten salt containing UCl_3 (3 m/o UCl_3 -48 m/o MgCl_2 -29 m/o NaCl -20 m/o KCl) at 540°C. Total running time for the loop is now 1348 hr. Evaluation of results from the run with UCl_3 is not yet complete.

In support of engineering studies for the pyrochemical process a high-temperature mixing dynamometer was built and operated. The tests were conducted in single-phase systems using flat paddles agitating either mercury, water, cadmium, No. 10 oil, or a liquid lead-bismuth eutectic alloy in baffled tanks. An equation to estimate mixing power has been developed and is of the form

$$P = 9.68 \text{ HD}^4 \text{N}^3 \rho / g_c,$$

where

P = mixing power;

H = vertical dimension of agitator blade;

D = agitator diameter;

N = agitator speed (revolutions per unit time);

ρ = liquid density;

g_c = constant.

Values of mixing power can be calculated with an estimated accuracy of $\pm 15\%$ from the relationship. A mean value of density would have to be estimated for a two-phase system.

⁶Hultgren, R., et al., Selected Values of Thermodynamic Properties of Metals and Alloys, John Wiley and Sons, New York, N.Y. (1963), p. 676.

A laboratory-scale experiment was carried out to investigate the possible use of a pyrochemical process for the recovery of plutonium from sodium fluoride traps. The fluoride volatility process employs sodium fluoride traps to collect and purify the plutonium hexafluoride product stream (see Progress Report for December 1966, ANL-7286, pp. 53 and 54). An experiment was carried out in which a sample of NaF containing plutonium and uranium was melted with NaCl and MgCl₂, and then contacted with a liquid Zn-10 w/o Mg alloy at 800°C. Greater than 99% removal of the plutonium and uranium from the salt was achieved. This result indicates that a pyrochemical procedure can be successfully utilized to recover plutonium from NaF traps.

In other work, two 12.5-kg double ingots of enriched U²³⁵-5 w/o fissium have been cast for use in an investigation of the effect of aluminum and silicon on the performance of Mark-IA EBR-II fuel.⁷ Both ingots were made following the procedure⁸ previously used for the preparation of Mark-IA fuel. The first casting was made to conform to this composition. The second casting contained 900 ppm aluminum and 350 ppm silicon which were added to the alloy.

E. Design Concept Analyses and Advanced Systems Evaluation

1. 1000-MWe Study

The study contract with the Babcock and Wilcox Co., which was executed on February 3, provides that the work be done on a cost-plus-fixed-fee basis from October 15, 1966 through May 17, 1968. Progress reporting and planning on the contract will be by means of the Program Evaluation and Review Technique (PERT).

The objectives of the B&W study are: (a) to prepare a reference design of a 1000-MWe LMFBR nuclear steam-supply system, observing specified ground rules and based on an estimation of the state of technology attainable in 1980, and (b) to identify the research and development programs required, and to establish their relative merit by means of a cost-benefit study.

B&W reported that its Task I work, which consists of developing and evaluating four plant concepts and selecting a reference design from the best features of each concept, is progressing on Concepts 1, 2 and 3. Work on Concept 1 is essentially complete, Concept 2 is about 75% complete, and work on Concept 3 is in its initial stages. In establishing Concepts 2 and 3, B&W has used its Parametric Study Code, which has as inputs economic and plant parameters for both the nuclear steam supply system and the energy conversion system. The three concepts are:

⁷ Chemical Engineering Division Semiannual Report, July-December 1965, ANL-7125, p. 60.

⁸ Ibid., pp. 42 and 43.

	Concepts		
	1	2	3
Geometry	Pancake	Cylindrical	Annular
Ratio of height to diameter	0.134	0.34	-
Fuel	Mixed oxides	Mixed oxides	Mixed oxides (and vented)
Moderation	Nil	Nil	Nil
Refueling	Double rotary plugs and shielded cask	Hot cell	Hot cell
Primary system	Pot	Loop	Loop
Reprocessing	Off site	Off site	On site

The comments from Atomics International, Combustion Engineering, General Electric, and Westinghouse in regard to background-patent and technical-data requirements have been received and are being reviewed by the AEC.

F. General Research and Development

1. Fast Reactor Core-parameter Study

Neutronics calculations were made to compare the diffusion and transport theories for cores having low ratios of height to diameter (H/D). For a given set of core dimensions and volume fractions, a composition that would give criticality was determined by use of the MACH-I diffusion code and ANL-224 cross-section set. The same dimensions and resultant compositions were used in the SNARG transport code (S_8 approximation), again with the ANL-224 cross-section set, to determine k . At an average core power density of 300 kW/liter, k_{SNARG} was found to vary with core half-height as follows:

Core half-height (cm)	k_{SNARG}
22.85	1.00825
36.27	1.00386
57.46	1.00147
106.07	1.00028

For a given H/D ratio, k_{SNARG} increased with increasing sodium volume fraction; the effect increased sharply with decreasing H/D .

The fissile-to-fertile ratio was computed by SNARG for $H/D = 0.1$ and average core power densities of 300, 400, 500, 600 and 700 kW/liter.

With a technique similar to that given in ANL-7001,⁹ sodium-void coefficients were computed for uniform removal of 40% of the core sodium. The calculations were made with the MACH-I diffusion code and ANL-224 cross-section set. As expected, for a given average core power density, the void coefficient decreased with decreasing core H/D. The decrease was gradual down to H/D $\approx$ 0.6, and rapid below that. For the reference systems, the coefficient at H/D = 0.2 was computed to be 1.0019 for $\bar{P}$ = 300 kW/liter, 1.0040 for 400 kW/liter, and 1.00455 for 500 kW/liter. It was found that not accounting for increased transverse leakage due to the partial voiding resulted in underprediction of the coefficient. Some void calculations will be repeated using the Russian ABN cross-section set in which there are U²³⁸ sets having different values of the potential scattering cross section.

Before beginning work on radial core zoning, two-dimensional calculations will be required to test the applicability of self-consistent diffusion theory.

The development of a computer program to perform reactor composition (fuel, structure, and coolant) calculations and thermal-hydraulic analyses, including coolant orificing, has been completed. This program was developed as two separate programs, a reactor composition program and a thermal-hydraulic analysis program, because the memory size of the CDC-160A computer was too small to permit compilation of the combined program at one time. Because iteration between the two programs is necessary to converge to the proper maximum coolant velocity-core structural fraction-frictional pressure drop condition for a reactor, the reactor composition program and the thermal-hydraulic analysis program have been assembled as a single program to be run on the CDC-3600 computer. This program is being modified to improve its running time and input-data requirements.

2. Use of U²³⁵ in a Fast-reactor Era

With widening geographical acceptance of water reactors, the price of electrical energy from new stations will become reasonably uniform throughout the United States. This means that, within a few years after the fast breeder reactor (FBR) power plant is commercially accepted, there will be a large demand for such plants. To meet the demand for FBR plants, large quantities of plutonium will be needed for initial core loadings. If light-water reactor (LWR) plants, some of which have a plutonium recycle, form the backbone of the nuclear industry when FBRs are commercially accepted, a plutonium shortage will exist. Because of that possibility an investigation has been made of the use of uranium-235 as the available fissile species.

⁹ Link, L. E., et al., 1000-MWe Metal-fueled Fast Breeder Reactor, ANL-7001 (1966).

U^{235} could be used in three ways: (a) for water-type plants to generate power and to produce plutonium for FBR plants; (b) for the start-up of FBRs, with a gradual change to plutonium as enough is produced; and (c) in specially designed fast converter reactor (FCR) plants that produce plutonium for fueling new FBRs as well as electricity. To obtain background information on U^{235} use, simple neutronic, thermal, and cost calculations were made. These preliminary investigations were mainly centered on FCR plants, although some indicative information was developed pertaining to U^{235} uses in FBR and LWR plants.

As a first approach to the development of FCR systematics, one-dimensional neutronic calculations in a spherical system were made. The starting point was to consider the General Electric Co. "16" pancake concept¹⁰ as a sphere. Oxide fuel, stainless steel cladding and structural material, and tantalum poison material were used. Midcycle equilibrium conditions at a core burnup of 50,000 MWd/tonne of heavy atoms were estimated and formed a basis for neutronic calculations.

The thermal-design aspects of the reactors were considered in setting forth the compositions for the various neutronic calculations. Sufficient sodium volume for heat transport was specified to maintain a reasonable pressure drop across the core and axial blanket. The blanket volumetric composition for neutronic calculations was about the same as the core in each case because the reactor model was the pancake concept; slight changes in blanket composition were specified to account for the control material normally in the blankets.

One measure of possible worth of U^{235} fast converters is the comparison of their fuel-cycle costs to plutonium-fueled systems under various conditions of fissile plutonium price and U_3O_8 cost. Also, U^{235} fast converter designs can be partially compared on an intraclass basis by calculating fuel-cycle costs. Unfortunately, the only practical method of making a number of survey calculations at the present time is through the use of equilibrium fuel conditions, so the important startup effects are omitted.

a. Neutronic Investigations. The MACH-1¹¹ code with cross-section set ANL-224 was used for investigating the neutronic aspects of U^{235} fast converter reactors. Eight cases using U^{235} fuel and seven cases using fissile plutonium fuel were calculated. Summaries of some of the input and output information are given in Table XV for the U^{235} cases.

¹⁰ Cohen, K. P., and O'Neill, G. L., "Safety and Economic Characteristics of a 1000-MWe Fast Sodium-cooled Reactor Design," pp. 185-204, in Proceedings of the Conference on Safety, Fuels, and Core Design in Large Fast Power Reactors held at Argonne National Laboratory, Oct. 11-14, 1965, ANL-7120 (1966).

¹¹ Meneley, D. A., et al., MACH-1, A One-dimensional Diffusion-theory Package, ANL-7223 (June 1966).

TABLE XV. Results of Spherical Calculations for a U^{235} Oxide-fueled Fast Converter Reactor

	Cases							
	A	B	C	D	E	F	G	H
INPUT DATA								
Core vol (liters)	696				1770	2855		4400
Blanket (cm)	30			45	40	42		
Core radius (cm)	55				75	88		102
Core Composition (v/o)								
Fuel	38		45.2	45	41		38	
Clad and structure	18				17		18	
BeO	7.3		None					
Sodium	36	36.2		36.4	41.5	41.6	43.6	
Poison (Ta)	0.7	0.5	0.6		0.5	0.4		
Fissile Plutonium (v/o)								
Core	2.3	1.9	2.7	1.6	1.8	2.4		
Inner blanket	2.3	2.0	2.8	1.6	1.8	2.7		
OUTPUT DATA AT MIDLIFE								
U^{235} (w/o)	21.4	21.3	18.0	20.0	15.8	12.9	13.8	12.5
Core PDF ratio ^a	.337	.346	.393	.371	.491	.587	.550	.611
Reactor PDF ratio	.629	.696	.753	.842	.825	.882	.844	.854
Change in reactivity for voiding core (% Δk)	-1.85	-1.90	-1.53	-1.76	-1.13	-0.18	-0.48	+0.27
Doppler coefficient ($\times 10^{+3}$)				-2.8	-4.0	-4.9	-4.8	-5.4

^aFissile atoms produced/fissile atom destroyed on an instantaneous basis.

There were five main pieces of desired output information: fissile weight fraction in the fuel, core conversion ratio, total conversion ratio, sodium-void effect, and Doppler coefficient. The cases using plutonium as the fuel were calculated to provide a reference basis and only the U^{235} cases are reported.

(i) U^{235} -fueled Cases. The criticality search is made only on U^{235} content: the concentration of fissile plutonium was estimated prior to the computer runs. In these calculations no U^{236} was specified because of the preliminary nature of the survey and because the cross sections are not compiled.

The reactors having small-volume cores (Cases A through D) do not have as good neutronic performance as the larger cores. The ratio of fissile material produced to that destroyed ("PDF ratio") increases with reactor size. Although insufficient number of cases were calculated to express the relationships, Fig. 11 illustrates the general trends. The PDF ratio for the reactor at midlife increases if the fuel-and-fertile-material volume fraction is not significantly decreased with an increase in reactor size. Because the average concentration of plutonium in the core increases with size, the better breeding characteristics of plutonium compared to U^{235} tend to increase the PDF ratio. Normally, in a plutonium-fueled breeder with constant fuel fraction, a small decrease in instantaneous breeding ratio is expected with increase in size.

Fig. 11

Trends in Ratio of Fissile Material
Produced to that Destroyed for
Fast Converter Reactors

Totally voiding the core of sodium causes negative reactivity effects up to approximately the 4000-liter size; at smaller sizes, partial voiding causes a positive reactivity effect. To illustrate the effect of partial voiding, the change in reactivity is plotted as a function of volume-fraction voided in Fig. 12.

Fig. 12

Effect of Partial Sodium Voiding on
Reactivity of Fast Converter Reactors

With β_{eff} of about 0.6%, the worst case of voiding changes reactivity by less than a dollar for core sizes up to ~3000 liters; the cores fueled with fissile plutonium meet this condition under 700 liters.

Another significant characteristic of the U^{235} system relates to the Doppler coefficient. The increase in the U^{238} and decrease in U^{235} concentration in the core as reactor size is increased causes a significant increase in the reactor Doppler coefficient. The coefficient for a 4400-liter core with U^{235} fuel (Case H of Table XV) is about the same as for a 700-liter core with BeO in the core. Thus, somewhat similar safety characteristics may be expected from the Doppler point of view with a large U^{235} -fueled core as with a small fissile-plutonium-plus-BeO core.

Another interesting feature of large U^{235} -fueled cores is the fact that a reactor having an average core conversion ratio of 0.5 will have a reactivity swing about equal to that of a fissile plutonium-fueled breeder reactor with an instantaneous core conversion ratio of ~0.65. Thus, a smaller reactivity swing may be expected in a large U^{235} -fueled system than in a small fissile plutonium-fueled system.

b. Thermal-design Aspects. The thermal-design aspects, that is, the transport of heat from the fuel and the allowable heat generation of the fuel, are entwined with the mechanical and safety aspects of design. To simplify the situation for this study, the heat transport was considered within a frictional pressure-drop limitation and the allowable heat generation was considered within a range of maximum kilowatts per foot of fuel.

(i) Frictional Pressure Drop. The design for frictional pressure drop depends on allowed sodium velocities and temperature rises as well as the availability of equipment to do the pumping. Sodium pumps capable of developing heads equivalent to 150 psi are reasonably expected in the future. Present designs indicate that the average temperature rise in the reactor is $\sim 300^\circ\text{F} \pm 20\%$. Because the volume of sodium in the core serves no inherent purpose under steady-state conditions, the sodium should be pushed through the reactor as rapidly as cost permits. Safety considerations may modify that statement, but this modification will be reflected mainly in allowable Δp and Δt across the reactor. For a sodium velocity of 30 ft/sec, a series of generalized curves were developed to show the frictional pressure drop per foot of fuel tube versus total sodium fraction for various fuel-tube diameters. (Because the type of fuel-element spacer selected significantly influences the results, the curves are only indicative.)

The length of fuel tube, considering core, axial blanket, fission-gas chamber, transition lengths, etc., will probably never be less than 5 ft. With allowances for pressure drops in the distribution plenums, subassembly nose pieces, external loop, etc., the upper limit for frictional drop for the fuel tube will be ~ 20 psi/ft.

(ii) Fuel Heat Generation. For oxide fuel, the maximum allowable linear heat generation is ~20-25 kW/ft. This maximum is slightly influenced by the diameter of the fuel pin (i.e., the unclad fuel). The maximum fuel rating, which is factored by the core maximum to average ratio, is a function of design and geometry. Thus average fuel rating, in terms of specific power, is influenced strongly by the pin outer diameter; the cost implications of this will be covered under fuel-cycle costs.

The amount of cladding, spacer, and subassembly wall material required in a design, as well as the interchannel sodium and control volume, were included in design considerations. For these calculations, it was estimated that 20% of the core volume will be filled with material other than the fuel and the coolant sodium. Because there is an incentive to maintain a high fuel fraction (0.35 to 0.45) for neutronic performance, the coolant sodium fraction will probably be in the range from 0.32 to 0.45.

c. Fuel Cycle Costs. On the basis of these neutronic and thermal design factors, the fuel cycle costs, U_3O_8 requirements, and plutonium production are being estimated for five types of reactors.

III. GENERAL REACTOR TECHNOLOGY

A. Applied and Reactor Physics Development

1. Oscilloscope Display for On-line Computer

A simple oscilloscope display has been connected to the DDP-24 computer to assist in monitoring the progress of on-line experiments.

Oscilloscope display patterns are normally constructed of individual points, with one point generated by each display operation. With a picture tube having a conventional phosphor, it is necessary to redraw the display at frequent intervals to obtain a flicker-free presentation. If the points are generated directly by the central processor, the repeated redrawing of a moderately complicated display could monopolize the central processor and leave no time for it to do the remainder of its job. Therefore, display units for large systems incorporate auxiliary memory units that are loaded by the central processor with display information which then maintain the display without further attention from the central processor. In addition, some oscillator-display units include means for internally generating lines and other elementary figures, as well as alphanumeric characters, in order to reduce the amount of digital information that must be stored.

The recent availability of storage oscilloscopes at a moderate cost creates an attractive method for relieving the central processor of the task of display regeneration. With a storage oscilloscope, a given point in a pattern need be generated only once and will then remain on the screen until the entire pattern is erased. This feature is obtained with minimum complication to the system. Nonstorage operation may still be used where the display is constantly changing, as in the live display for a pulse-height analyzer.

On the other hand, the storage oscilloscope has two disadvantages for this application which must be tolerated. First, it is not possible to delete individual portions of a pattern. If deletions are necessary, the whole pattern must be erased and redrawn. Secondly, it is not possible to use a light pen, since the storage action makes it impossible to establish a coincidence between the light-pen signal and the generation of a particular point.

The storage oscilloscope in use is a Tektronix RM564 with two 2A60 plug-in amplifiers, one for each of the two axes. The only modification that had to be made in the scope was a provision for spot brightening under external control.

The interface circuitry has two tasks. First, it must translate the digital data from the computer into analog voltages for spot positioning. Secondly, it must perform various timing and control functions to be discussed. The display is connected to the computer through the standard unbuffered parallel output channel. Transfer of a single word through this channel displays a single point on the scope.

The x- and y-coordinates of a point are each represented by a ten-bit number read into a ten-stage flip-flop register. The analog deflection voltage is formed by a ladder attenuator connected to the flip-flop outputs. This attenuator feeds an emitter follower that transmits the voltage to the oscilloscope amplifier input. Either or both halves of the screen may be erased under program control or manually with the pushbuttons on the interface chassis.

In some on-line applications, the scope will be used for occasional monitoring of the status of an on-going measurement, such as a noise analysis. For this it is desirable for the human operator to decide when a display should be generated. It is also desirable for the display to be held as long as necessary for leisurely study by the operator while the collection of further data proceeds uninterrupted.

To achieve this type of operation, a "lockout" facility has been incorporated. Such a display is generated in the normal way, except that the last point is generated so that the output channel is left Not Ready. The program for the measurement is written in such a way that, at the time for a display, it tests the readiness of the output channel. If the channel is ready the program generates a display. If the channel is not ready the program bypasses the display operation and directly proceeds to collect more data.

When the operator is through looking at the display and wants to see a new one, he presses the erase button. The erase operation leaves the channel ready. When the program next tests the channel status, it will find the channel ready and so will generate an updated display before continuing with data collection.

A subroutine has been written to provide for generation of displays by FORTRAN-IV programs.

2. Evaluations of Cross Sections

Some modifications have been made in the SAD program which is used in the study of angular distributions of scattering. One modification is the option of using a logarithmic Y-axis in plotting the angular cross section, $d\sigma/d\Omega$, versus the cosine of the scattering angle. This is useful in plotting angular distributions, at high energy, which are strongly peaked

in the forward direction. Another change allows cards to be punched with Legendre coefficients in the ENDF/B normalization, as well as in the MC² normalization available previously.

The DRIVER program, which reads Legendre coefficients punched by SAD at a number of energies and plots coefficients for each l value as a function of energy, has been modified to accept input cards in the ENDF/B normalization as well as in the MC² normalization.

3. The ARC System

a. Module Development. The one-dimensional diffusion theory capability in the ARC system was successfully demonstrated for a small-dimension source calculation with the IBM System/360 Model 50 computer. The modules invoked by the k-calculation Standard Path were:

1. BCDDS--a module that generates BCD data sets from card input;
2. NUI001--Neutronics Specifications;
3. NUI002--Geometry Preparation;
4. NUC002--1D-Diffusion.

The modules concerned with cross-section homogenization were not yet available, so cross sections were supplied to the DATAPOOL manually.

Work is currently in progress to implement further modules of the system:

1. NUI003--Cross Section Homogenization Specifications;
2. NUC001--Cross Section Homogenization;
3. AJI001--Neutron Inventory Specifications;
4. AJC001--Neutron Inventory;
5. NUI0010--Neutronics Output Manipulation Specifications;
6. NUE001--Neutronics Output Manipulation.

The initial Phase One modules of the burnup package (see Progress Report for May 1966, ANL-7219, pp. 34-35) have been inserted into the ARC linkage structure on the Model 50 machine for the purpose of debugging the code logic. The major capabilities are as follows:

1. may be run with zero-, one-, or two-dimensional neutronics modules;
2. arbitrary burnup and decay chains;

3. generalized fuel management;
4. enrichment, control, burnup, and equilibrium-cycle searches;
5. multiple reprocessing plants and priority-ordered fuel fabrication.

The major portions of the work remaining are in the area of input formats and processing, nonequilibrium problem control, interrupt facilities, and output edits. Some further options will be added to the search routines at a later date.

One-dimensional problems will be tested as soon as one neutronics support module is available. One 22-group problem, run with direct flux input, has compared favorably with results of the zero-dimensional test program on the CDC-3600.

b. Development of Resonance Absorption Code. In calculating resonance absorption in a lump it is usually assumed that the temperature distribution in the lump is spatially flat. As the resonance absorption code RABBLE permits five compositions or mixtures, each of which can be at a different temperature, RABBLE has been used to investigate the effect of a nonuniform temperature distribution on effective cross sections and on the Doppler effect for U^{238} . A hypothetical fast reactor cell in slab geometry was considered. The inner mixture was U^{238} with a half-thickness of 0.30 cm, the middle mixture was iron with a thickness of 0.20 cm, and the outer mixture was sodium with a half-thickness of 1.0 cm. The relative change in the absorption rate in U^{238} with a change in the average temperature of uranium from 600 to 1200°K was computed for two energy ranges, 1.4-1.0 keV and 0.6-0.2 keV, for two cases. In one case, the temperature distribution of the uranium slab was taken as uniform; in the other, the temperature distribution was taken as parabolic and the uranium slab was subdivided into three compositions, each at a different average temperature. The parabolic temperature distribution was obtained by use of the average temperature and a surface temperature of 400°K. Resonance parameters for U^{238} were taken from the compilation of Hughes *et al.*,¹² for energies below 1 keV and from the measurements of Garg *et al.*,¹³ for energies above 1 keV.

The results of these calculations are given in Table XVI, from which it is seen that the effective cell-averaged absorption cross section, $\langle \Sigma_a \rangle_{\text{cell}}$, and the relative change in the absorption rate with temperature, $\Delta R_a/R_a$, are weakly dependent on the assumed temperature distribution in the absorbing slab. The difference in $\langle \Sigma_a \rangle_{\text{cell}}$ and $\Delta R_a/R_a$ for the

¹²Hughes, D. J., Magurno, B. A., and Brussel, M. K., BNL-325, Suppl. 1, 2nd ed. (1960).

¹³Garg, J. B., *et al.*, Phys. Rev. **134**, B985 (1964).

two temperature distributions is only between 1 and 2%. It is difficult to predict whether the assumption of a uniform temperature distribution will overestimate the relative change in absorption at these energies because of the effect of interference scattering. At lower energies, where resonances are highly absorbing, it would be expected that the assumption of a uniform temperature would overestimate $\Delta R_a/R_a$ by overestimating the temperature change near the surface of the absorbing slab where neutrons are preferentially absorbed. To illustrate this point, calculations were made of absorption in the 6.7-eV resonance of U^{238} for a water-moderated cell; the results of these calculations are given in Table XVII. From Table XVII, it is seen that $\Delta R_a/R_a$ is overestimated for a uniform temperature distribution in the absorber. But it is also seen that $\Delta R_a/R_a$ is small because the resonance, being at a low energy, is not greatly Doppler broadened.

TABLE XVI. Resonance Absorption in U^{238} -Fe-Na Cell for 1.4-1.0 keV and 0.6-0.2 keV

Material	X_{outer} (cm)	$\langle T \rangle$ (°K)	$\langle \phi \rangle$	$1 - p$	$\langle \Sigma_a \rangle_{cell}$ (cm ⁻¹)	Energies (keV)	Regions	$\Delta R_a/R_a$
U	0.3	600	0.8371	0.22064	0.008400	1.4-1.0	6	0.115320
Fe	0.5	400	0.8554				2	
Na	1.5	400	0.8582				4	
U	0.3	1200	0.8174	0.24609	0.009578	1.4-1.0	6	
Fe	0.5	400	0.8369				2	
Na	1.5	400	0.8401				4	
U	0.1	689	0.8350	0.22143	0.008431	1.4-1.0	2	0.113826
U	0.2	622	0.8382				2	
U	0.3	489	0.8458				2	
Fe	0.5	400	0.8548				2	
Na	1.5	400	0.8576				4	
U	0.1	1556	0.8148	0.24663	0.009597	1.4-1.0	2	
U	0.2	1289	0.8182				2	
U	0.3	755	0.8262				2	
Fe	0.5	400	0.8365				2	
Na	1.5	400	0.8396				4	
U	0.3	600	0.6577	0.57966	0.008468	0.6-0.2	6	0.119056
Fe	0.5	400	0.6808				2	
Na	1.5	400	0.6831				4	
U	0.3	1200	0.6120	0.64867	0.010138	0.6-0.2	6	
Fe	0.5	400	0.6368				2	
Na	1.5	400	0.6392				4	
U	0.1	689	0.6575	0.58796	0.008626	0.6-0.2	2	0.121556
U	0.2	622	0.6600				2	
U	0.3	489	0.6666				2	
Fe	0.5	400	0.6764				2	
Na	1.5	400	0.6784				4	
U	0.1	1556	0.6106	0.65943	0.010367	0.6-0.2	2	
U	0.2	1289	0.6138				2	
U	0.3	755	0.6211				2	
Fe	0.5	400	0.6314				2	
Na	1.5	400	0.63389				4	

TABLE XVII. Resonance Absorption in U^{238} -Fe-H₂O Cell for 12-3 eV

Material	X_{outer} (cm)	$\langle T \rangle$ (°K)	$\langle \phi \rangle$	$1 - p$	$\langle \Sigma a \rangle_{cell}$ (cm ⁻¹)	Regions	$\Delta R_a / R_a$
U	0.3	600	0.8305	0.04084	0.027638	6	0.012700
Fe	0.5	400	0.8810			2	
H ₂ O	1.5	400	0.9490			4	
U	0.3	1200	0.8268	0.04135	0.028031	6	
Fe	0.5	400	0.8795			2	
H ₂ O	1.5	400	0.94833			4	
U	0.1	689	0.8233	0.04083	0.027636	2	0.012389
U	0.2	622	0.8290			2	
U	0.3	489	0.8400			2	
Fe	0.5	400	0.8810			2	
H ₂ O	1.5	400	0.9490			4	
U	0.1	1556	0.8196	0.04134	0.028018	2	
U	0.2	1289	0.8255			2	
U	0.3	755	0.8372			2	
Fe	0.5	400	0.8795			2	
H ₂ O	1.5	400	0.9480			4	

These calculations indicate strongly that absorptions rates and their change with temperature are only weakly dependent upon the assumed temperature distribution in the absorbing lump. Therefore, it appears that the current practice of assuming a uniform temperature distribution is valid.

B. Reactor Fuels and Materials Development

1. Chemistry of Fuel Materials

a. Mass Spectrometric Study of the System ReO₂-ReO₃-Vapor.

Vapor equilibria in the rhenium-oxygen system are being studied in connection with the projected use of rhenium as a container material for high-temperature thermodynamic investigations of plutonia-containing systems. It is anticipated (see Progress Report for September 1966, ANL-7255, pp. 52-53) that rhenium will react with plutonia to some extent at high temperatures, resulting in the formation of rhenium oxide vapor species. The results of a mass-spectrometric determination of the vapor composition over solid ReO₂ at 850°C (see ANL-7255, pp. 52-53) disagree considerably with the mass-spectrometric data reported by Semenov and Ovchinnikov¹⁴ for vaporization of ReO₂ and of ReO₃. The discrepancy prompted us to examine the vaporization behavior of ReO₃, which is discussed below, and to extend the earlier study on ReO₂ vaporization.

¹⁴Semenov, G. A., and Ovchinnikov, K. V., J. Gen. Chem. USSR (Eng. Transl.) 35(9), 1517 (1965).

On heating solid ReO_3 at 411°C in a platinum effusion cell, the following ions were observed mass spectrometrically at ionizing electron energies of 25 eV: ReO_2^+ , ReO_3^+ , Re_2O_5^+ , Re_2O_6^+ , and Re_2O_7^+ . (Additional ions were observed at higher ionizing energies.) The appearance potentials obtained from two sets of measurements are listed in Table XVIII. An analysis of the appearance potentials and of the variation of the ion-intensity ratios with ionizing electron energy showed that Re_2O_7 was the predominant parent molecule. ReO_3 may have existed as a minor parent molecule comprising, at most, 3% of the total vapor. However, most of the ReO_3^+ that was observed, as well as the Re_2O_6^+ , Re_2O_5^+ , and ReO_2^+ , originated from fragmentation of parent molecules. In contrast to these results, Semenov and Ovchinnikov concluded that the vapor over solid ReO_3 at 380°C is composed predominantly of almost equal amounts of ReO_3 and Re_2O_7 .¹⁵

TABLE XVIII. Approximate Appearance Potentials of Species Vaporizing from Solid ReO_3 at 411°C

Appearance Potential (eV)		Appearance Potential (eV)	
Re_2O_7^+	12.9, 13.1	Re_2O_5^+	~20
ReO_3^+	15.7, 16.2	ReO_2^+	~20
Re_2O_6^+	17.6		

X-ray powder diffraction patterns of the solid residues from similar experiments showed that, on heating at 411°C , $\text{ReO}_3(\text{s})$ is converted from a single phase to the two-phase mixture $\text{ReO}_3(\text{s}) + \text{ReO}_2(\text{s})$. (Prolonged heating at 411°C or higher produced a residue of ReO_2 only.) This result, together with the observation that the vapor phase was predominantly Re_2O_7 , indicates that the vaporization proceeds mainly according to the equation

The system, therefore, is univariant, consisting of two solid phases and a vapor phase.

The vapor pressure of the system ReO_2 - ReO_3 -vapor, assuming the vapor to be Re_2O_7 entirely, was obtained as a function of temperature by combining the results of two types of measurements: Re_2O_7^+ ion intensity versus temperature, and rate of Re_2O_7 effusion (by weight loss) at a single temperature.

¹⁵A significant amount of HReO_4 was also reported, but no explanation was offered for its presence.

The temperature dependence (339 to 437°C) of the Re_2O_7^+ ion intensity was obtained in three series of measurements. The ReO_3 samples (~250 mg) were first preheated at 411°C until the ion intensities reached constant values, and then the temperature-dependence measurements were begun. (During the pretreatment period of ~90 min, the $I_{\text{Re}_2\text{O}_7^+}/I_{\text{ReO}_3^+}$ ratio remained constant within experimental error.) The ion intensity data ($\log IT$ versus $1/T$) was least squared, and the three series were combined by normalizing the data at 388°C, the midrange temperature.

The rate of effusion [$\text{g}/(\text{cm}^2)(\text{sec})$] was measured at 411°C in three runs. The size of the ReO_3 samples, the pretreatments, and the duration of the runs were the same as for the temperature-dependence measurements. Also, X-ray powder diffraction patterns of the residues from both types of measurements showed ReO_3 as the major phase and ReO_2 as a minor phase. The average vapor pressure, calculated from the three effusion measurements, was 1.30×10^{-5} atm at 411°C.

The variation of vapor pressure with temperature, calculated from the relationship $IT = kP$, can be represented by the equation

$$\log P (\text{atm}) = 12.529 - \frac{11,913}{T} \quad (339-437^\circ\text{C}). \quad (2)$$

The combined uncertainty (95% confidence level) is $\pm 14\%$. The vapor pressure in our study is lower by about one order of magnitude than the vapor pressure obtained by Deev and Smirnov¹⁶ from Knudsen effusion measurements on the vaporization of ReO_3 . Furthermore, the enthalpy of vaporization [i.e., reaction according to Eq. (1)] obtained in this study is 54.5 ± 0.8 kcal/mole of Re_2O_7 , which differs from the values reported by Deev and Smirnov, 49.8 kcal/mole, and by Semenov and Ovchinnikov, 49 ± 3 kcal/mole. The explanation(s) for the various discrepancies is not readily apparent.

2. Fabrication and Evaluation

a. Properties of Actinide Compounds with Nonmetals of Groups IV to VI. Work on actinide compounds included the preparation by arc-melting of equiatomic compounds of uranium and of plutonium with the germanium, antimony, selenium, and tellurium (see Progress Report for November 1966, ANL-7279, p. 53). X-ray diffraction patterns demonstrated the stoichiometry of the compounds.

Plutonium arsenide has since been prepared by reaction of decomposed PuH_{2-x} with arsine gas. A procedure of alternate decomposition and gas reaction at 400, 500, and 600°C similar to the technique used to

¹⁶Deev, V. I., and Smirnov, V. I., Dokl. Akad. Nauk. SSSR 140, 822 (1961).

prepare the sulfides and phosphide of plutonium was tried. This same procedure could not be used for preparation of plutonium arsenide because the arsine gas decomposed at temperatures above 300°C. Therefore, the hydride was decomposed at the higher temperatures (400 to 700°C) and the reaction with arsine was carried out at 250 to 300°C. This reaction was extremely exothermic because the liberated hydrogen reacted with the plutonium powder at this temperature. Many cycles of alternate decomposition and gas reaction were needed to produce PuAs, which appears to be the only compound in the Pu-As system. To prevent undue temperature rise the gas was admitted to the system slowly.

Chemical analysis of the PuAs powder gave values of 0.08 w/o oxygen and 0.006 w/o nitrogen. This impurity level is considered good for the gas-metal reaction. Preliminary measurements indicate that the melting point of this compound is near 2450°C.

An alternative method of solid-phase reaction was used for the preparation of uranium and plutonium arsenides since the vapor pressure of arsenic is less than one mm below 370°C. This technique consisted of mixing arsenic powder with either the hydride of uranium or plutonium in near-stoichiometric proportions and slowly heating this mixture in a vacuum of 0.020 Torr. Decomposition of the hydride and reaction with the arsenic was observed at temperatures below about 400°C. The mixtures were heated to approximately 700°C in a vacuum to complete the reaction. This treatment was followed by homogenization at higher temperatures in a vacuum of 10^{-6} Torr. The temperature was increased very slowly at this stage of the process to allow any free metal present to equilibrate with the arsenic-rich phases. Chemical analysis showed that this material contained more than 0.2 w/o oxygen; therefore, this procedure was discontinued.

b. Thermal Stability of Plutonium Ceramics. Of the refractory metals, iridium appears to be the most satisfactory material for holding a pellet of PuO₂ in a vacuum at 2116°K (see Progress Report for January 1967, ANL-7302, p. 65). A coil of rhenium wire, however, was satisfactory for holding PuO₂ pellets during evaporation runs at 1930 to 2115°K. Despite continuation of evaporation runs until the weight-loss rate of the PuO₂ appeared constant, the O/Pu ratio continued to change with time of heating.

Characterization of specimens to be used in experiments on mixed uranium-plutonium oxides requires accurate determination of oxygen-to-metal (O/M) ratios in nonstoichiometric compositions. Work is therefore in progress to develop an analysis technique based on equilibration of specimens in a flowing 10/1 (by volume) CO/CO₂ gas mixture at 850°C as proposed by Markin et al.¹⁷

¹⁷Markin, T. L., Walter, A. J., and Bones, R. J., The Determination of Oxygen/Metal Ratios for Uranium, Plutonium and (U·Pu) Oxides, AERE-R 4608 (1964).

Separate equilibration runs with U_3O_8 and PuO_{2-x} showed that the CO/CO_2 mixture reduced the former to $UO_{2.00}$, and oxidized the latter to a composition that was assumed to be $PuO_{2.00}$. In a further experiment, a PuO_{2-x} specimen was oxidized to constant weight in a helium-1 v/o oxygen mixture at $1100^\circ C$. On subsequent equilibration of the specimen in CO/CO_2 at $850^\circ C$, lack of weight change indicated that the final composition was $PuO_{2.00}$. The evidence that the equilibration method can produce the stoichiometric composition in both end members indicates that it should be a suitable method for the determination of O/M ratios in $(U,Pu)_2$ solid solutions.

Experiments were begun to determine O/M ratios on $(U,Pu)O_{2-x}$ powder and pellets. It was discovered that it was necessary to preoxidize the mixed-oxide specimens to insure that the CO/CO_2 equilibration could be done in a reasonable length of time (ca. 4 hr). Preoxidation of the specimens to O/M ratios greater than 2.00 was done by heating at $1100^\circ C$ in helium-1% oxygen.

A previous report (see Progress Report for October 1966, ANL-7267, p. 57) indicated that a mixed-oxide pellet was destroyed by oxidation. The pellet used in the present experiments remained intact after oxidation. X-ray results showed that the former pellet contained two phases, whereas the latter was a single-phase solid solution. Houston observed similar behavior and attributed the destruction of the pellet to oxidation of a uranium-rich oxide phase to U_3O_8 .¹⁸ The oxidation behavior of the pellet apparently is a sensitive criterion for determining whether complete solid solution exists.

PuO_2 powder has been supplied to the Chemistry Division for development work on a technique for the absolute determination of O/M ratios in PuO_2 and mixed oxides. A check by such an independent method would remove any doubts concerning the accuracy of the CO/CO_2 equilibration analysis.

c. U-S-O System. Both U_2S_3 and U_3S_5 are orthorhombic and US_2 is the only uranium sulfide that loses significant weight on vacuum sintering (see Progress Report for November 1966, ANL-7279, pp. 53-55). The electrical resistivities were reported for US , U_2S_3 , U_3S_5 , and US_2 , and also the order of reactivity of these sulfides and UOS in warm air.

Melting-point determinations for compounds on the U-S join and UOS were made by use of the tungsten wedge technique in helium at one atmosphere. Values determined were: US , $2440^\circ C$; U_2S_3 , $1775^\circ C$; U_3S_5 , $1695^\circ C$; and UOS , $1925^\circ C$. US , U_2S_3 , and UOS seemed to melt congruently, as evidenced by the sharpness and consistency of melting, whereas the congruency of U_3S_5 was questionable and α - US_2 decomposed to U_2S_3 .

¹⁸Houston, M. D., Effect of PuO_2 Content on Sintering Behavior of Mechanically Mixed UO_2 - PuO_2 , NUMEC-2389-8 (1965).

Vacuum firings of pellets in effusion-type crucibles added supporting data. The α - $\text{US}_{2.00}$ polymorph retained the α structure but decomposed at 1280°C to a $\text{US}_{1.88}$ composition which slowly transformed to U_3S_5 at 1450°C . In turn, U_3S_5 decomposed completely to U_2S_3 at 1605°C , but the U_2S_3 showed no tendency to decompose at 1655°C .

The relative resistance of tungsten and tantalum to sulfur corrosion was observed by decomposing US_2 pellets at 1600°C in closed crucibles. The tantalum crucible showed considerable weight gain due to the formation of a moderate reaction layer of tantalum sulfide (probably $\text{TaS}_{1.1}$). The tungsten crucible, however, showed negligible weight change or reaction since the sulfur potential in the crucible apparently favored the WS_2 decomposition reaction.

Metallographic analysis of various compositions in the $\text{US-UOS-U}_2\text{S}_3$ ternary subsystem showed it contains no ternary eutectic or peritectic reactions but rather a class-II, four-phase equilibrium of the type $\text{L} + \alpha \rightleftharpoons \beta + \gamma$, which is intermediate between the two. In this case, $\text{L} + \text{US} \rightleftharpoons \text{U}_2\text{S}_3 + \text{UOS}$. Thus a composition near US containing small amounts of oxygen and hyperstoichiometric sulfur when cooled from the melt will show large primary US grains with a secondary precipitate of U_2S_3 between the grains, and a final eutectic-like precipitate of U_2S_3 and UOS at the multigrain junctions. The solidification temperature for this final U_2S_3 - UOS precipitate has been bracketed at between 1480 and 1605°C .

The $\text{US-UOS-}\text{UO}_2$ ternary subsystem also appears to be somewhat complex. Compositions of UOS-20 m/o UO_2 and UOS-40 m/o UO_2 remained as a two-phase mixture with negligible solubility of one phase in the other when fired at 1200°C . At 1650°C , however, X-ray analysis indicated the UOS area had expanded to take all the UO_2 into solid solution. About 5 to 10 v/o US was present as a secondary phase. Systematic d-spacing changes indicated that, with increasing oxygen content, the tetragonal UOS unit cell was contracting in the c-direction while remaining constant in the a-direction. This would be expected, to a point, if oxygen were replacing sulfur atoms in the sulfur layers, which lie perpendicular to the c-axis of this completely layered PbFC_2 -type structure.

d. Thermal-diffusivity of Ceramic Materials. The thermal diffusivity of pressed and sintered 93% dense plutonium monocarbide was measured in order to resolve the scatter of data obtained from earlier reports. The present data were combined with the previously determined heat capacity to yield the thermal conductivity between room temperature and 800°C as shown in Fig. 13. The thermal conductivity determined by us is considerably lower than that reported by Leary *et al.*¹⁹ between 200 and 400°C . The slope of the curve is also less than that of Leary *et al.*; this

¹⁹Leary, J. A., Thomas, R. L., Ogard, A. E., and Wonn, G. C., Symposium on Carbides in Nuclear Energy at Harwell, Vol. 1, p. 365 (MacMillan & Co., London, 1964).

Fig. 13. Thermal Conductivity of Plutonium Monocarbide

would be expected on the basis of our electrical-conductivity results. The results on the monocarbide of plutonium are being analyzed further and we intend to correlate these data with the work on plutonium monophosphide and monosulfide now in progress.

e. Irradiation of Prototype Paste Fuel Element. Postirradiation

examination of the prototype paste fuel element developed by United Nuclear Corporation and irradiated by Argonne National Laboratory is essentially complete. The specimen jacket consisted of a Type 304 stainless steel tube 18 in. long, of 0.375-in. OD, and with a 0.020-in. wall thickness. The tube contained a 9-in.-long fuel column of a 50-50 mixture by volume of sodium and 20% enriched UO_2 powder. The particle size of the urania powder was approximately $10\ \mu$. The fuel element was irradiated in the MTR to one a/o burnup at a maximum jacket temperature near 600°C .

As reported previously (see Progress Report for November 1966, ANL-7279, pp. 55-56), neutron radiographs revealed that the fuel had redistributed within the specimen, that the Type 304 stainless steel tube was bowed and distorted, and that the specimen was probably breached in one area. In addition, temperature records indicated that no upward movement of the fuel column had occurred during irradiation. Additional temperature records for irradiation between 0.5 and 1 a/o burnup revealed that the fuel column behaved as predicted during the later cycles of irradiation.

The inner Type 304 stainless steel specimen container capsule was warped and severely oxidized over an area approximately 18 in. long. This capsule was punctured with a laser to collect the fission gas in the event the fuel element was ruptured. Determination of the amount of fission gas collected is not yet complete. The fuel element was ruptured in two areas, approximately 2-3 and 4-5 in. from the bottom of the element. Maximum diameter increases of 0.030 in. occurred in regions immediately adjacent to the ruptures and near the top of the original fuel column.

A gamma scan of the fuel element showed high levels of activity below the lower rupture and at the top end plug. Subsequent sectioning substantiated the results of the gamma scan. No fuel remained in the fuel element above the lower rupture except in an area, below the top end plug, that was 0.5 in. long. Portions of fuel fell out of the lower rupture during sectioning, and it is possible that the fuel in the regions indicated by the neutron radiographs was lost during disassembly.

The remaining phases of the evaluation will include burnup analysis and metallographic examination of the jacket, pieces of loose fuel, and fuel in the lower and upper regions of the fuel element.

f. Irradiation of High-temperature Materials. A series of irradiations in instrumented capsules in the MTR are in progress on ceramic fuel materials being developed under the high-temperature materials program. A summary of the irradiations is shown in Table XIX.

TABLE XIX. Status of Irradiations of Ceramic Fuel, Clad in Nb-1 w/o Zr, in MTR

Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions			
		Fuel Composition (w/o)	Effective Density (%)	Cladding OD (in.)	Cladding Thickness (in.)	Power Density (kW/cc) ^a	Max Cladding Temp (°C)	Burnup to Date	
								a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ ^a
56-11	MV-2	UC-20 PuC	79	0.281	0.012	1.2	470	7.0	18.0
56-8	MV-3	UC-20 PuC	81	0.281	0.012	1.2	715	7.5	19.8
56-8	MV-5	UC-20 PuC	80	0.281	0.012	1.2	705	7.2	18.8
56-11	MV-6	UC-20 PuC	80	0.281	0.012	1.2	480	7.5	19.6
56-13	Z-4	UC-20 PuC	79	0.174	0.015	1.2	665	2.4	6.2
56-13	Z-5	UC-20 PuC	79	0.174	0.015	1.2	585	2.4	6.2
56-13	Z-7	UC-20 PuC	79	0.174	0.015	1.2	570	2.4	6.2
56-13	C-45	PuC	84	0.174	0.009	1.4	700	2.4	6.6
56-8	S-7	US	80	0.281	0.012	1.0	535	5.8	11.2
56-8	S-8	US	89	0.281	0.012	1.0	725	7.8	16.8
56-8	S-9	US	76	0.281	0.012	1.0	750	7.8	14.4
56-8	S-10	US	91	0.281	0.012	1.0	690	7.8	17.2
56-11	S-15	US	82	0.281	0.012	1.0	380	4.9	9.8
56-11	S-16	US	90	0.281	0.012	1.0	510	6.8	14.8
56-11	S-17	US	88	0.281	0.012	1.0	500	5.5	11.7
56-11	S-18	US	77	0.281	0.012	1.0	610	7.3	13.6

^aBased on effective density.

As reported (see Progress Report for January 1967, ANL-7302, pp. 61-61), heat treatment of vibratorily compacted unirradiated PuC_{1.02} particles at 1300°C caused but little sintering, whereas irradiation of PuC_{1.33} caused extensive swelling and sintering, with PuC forming a network around coarse UC particles. Irradiation appears to cause depletion of UC₂ platelets at the surface of UC_{1+x} particles.

Electron-microscopical studies of finer microstructural changes in high-burnup specimens of vibratorily compacted UC-20 w/o PuC powders have been completed. Greater fission-gas release in the PuC_{1-x} was related to the fact that fission-gas cavities in the PuC_{1-x} are generally larger and more interconnected than in the PuC_{1+x}. The PuC_{1+x} exhibited more of spherically shaped second-phase particles than did PuC_{1-x}. It was tentatively proposed (see ANL-7302, pp. 62-63) that this difference in the distribution of the second-phase particles accounts for the difference in the size and distribution of fission-gas bubbles in PuC_{1+x} and in PuC_{1-x}. However, identification of the second-phase particles and study of other irradiation effects (such as solid-solution formation and reactions between fuel and jacket material) require a shielded electron-beam microprobe X-ray analyzer.

Technical specifications for a modified microprobe that can be used to analyze highly radioactive materials (for example, approximately one gram of fuel specimens containing uranium and plutonium irradiated to 10 a/o burnup and allowed to cool for 60 days) were prepared and sent out

for bidding by vendors. A vendor has been selected on the basis of his response. Procurement of the analyzer has begun, as well as necessary modifications to the laboratory space to be used.

g. Dissolution Kinetics in Liquid Metal Systems. A proposed method for analysis of oxygen concentrations in sodium is being developed in conjunction with the study of corrosion mechanisms in refractory metal-sodium systems. The method involves the equilibration, with respect to oxygen, of tantalum wires in liquid sodium and the subsequent measurement of the oxygen concentration in the tantalum by internal friction techniques.

The relationship between internal friction peak height and oxygen concentration in tantalum was determined by comparing measured internal friction peak heights with vacuum fusion data. The results obtained with 0.51-mm-dia tantalum wire specimens gave the following relationship:

$$C_{O_{Ta}} = 10,850 Q^{-1}.$$

The primary advantage of the internal friction method is believed to be the higher sensitivity at low oxygen levels. Well-defined internal friction peaks corresponding to less than 2 ppm by weight of oxygen in tantalum have been measured.

The oxygen partitioning between tantalum and sodium has been calculated previously from the best available solubility and free-energy data:

$$K_w = \left(\frac{\text{ppm } \underline{O} \text{ in Ta}}{\text{ppm } \underline{O} \text{ in Na}} \right) = 7.829 \exp \left(\frac{1098}{T} \right);$$

this corresponds to a value of 25.8 at 650°C.

Several tantalum wires have been exposed in sodium systems in an attempt to verify this distribution coefficient experimentally. Significant changes (both increases and decreases, depending upon the initial conditions) in the oxygen peak heights of the tantalum wires have resulted from the exposure to sodium. The oxygen analyses in sodium based on the calculated distribution coefficient have generally been lower than oxygen analyses of the sodium by other methods. One tantalum specimen exposed to sodium cold-trapped at 167°C indicated an oxygen level of 1.73 ppm from the friction peak, 3.70 ppm from cold-trap solubility data, and 13.3 ppm by mercury amalgamation. The amalgamation method is believed to give high values since it does not distinguish between oxygen and some of the other interstitial elements in sodium.

Tantalum wires exposed in a low-oxygen, noncirculating sodium pot have indicated lower oxygen levels than obtained by vacuum distillation techniques. The oxygen level of the sodium in this test was near the lower limit for accurate analysis by vacuum distillation.

It appears that the calculated distribution coefficient is high; however, more tests are needed to establish the value. Present equipment is being modified to permit vacuum distillation analysis on a cold-trapped circulating system.

A significant result of the experimental tests has been the pickup of nitrogen by the tantalum wires exposed to sodium. Since nitrogen does not diffuse as readily as oxygen in tantalum, the nitrogen was not equilibrated in these tests. However, a large increase in the nitrogen peak height has been detected in each sample. The nitrogen content of the sodium is yet to be determined because of lack of basic thermodynamic data; however, the detection of the nitrogen pickup by the tantalum indicates an important feature of the internal friction technique, that of distinguishing between the various interstitial elements.

The detection of nitrogen has led to an expanded investigation of this technique for interstitial-element analysis. The long equilibration time for nitrogen in tantalum has led to the use of smaller (0.25-mm-dia) wires. The possible use of niobium or vanadium instead of tantalum is also being investigated, since the diffusivities of nitrogen are considerably higher in these latter metals.

Although the actual amount of nitrogen in the sodium systems has not been determined, the large pickup of nitrogen by the tantalum wires has led to considerable interest in the possible effects on corrosion of refractory metals in sodium caused by interstitial elements other than oxygen. Other methods of oxygen analysis are also being more closely examined to determine what impurities they actually measure.

3. Radiation Damage to Structural Materials

a. Fast-neutron Spectra and Radiation-damage Rates in EBR-II.
A characterization of the EBR-II neutron spectra has been initiated in order to make irradiation experiments in the reactor more meaningful (see Progress Report for December 1966, ANL-7286, p. 46). Fast-neutron spectra are now being determined as a function of position in the core of the EBR-II. This information can be used to report flux rates and fluences in a fast reactor for radiation-damage studies. Experimenters can use the information to predict exposure levels for the design and planning of irradiations.

Spectra are calculated by using the SNARG and MACH-1 codes, and the absolute magnitude of the flux is obtained by irradiation and analysis of foil wires in several locations in the EBR-II core. The wires give vertical flux profiles and the calculations supply radial flux variations. The total information makes possible the construction of a complete flux map.

Wires of iron, nickel, titanium, copper, and uranium were irradiated in special subassemblies (six locations in Rows 1 through 5) during 30 kWhr runs at low power. The induced activity was low enough that the wires and the stainless steel tubes containing them could be handled without special shielding. A high-power run is planned to obtain better statistics on the $\text{Cu}^{63}(\text{n}, \alpha)\text{Co}^{60}$ reaction.

Table XX gives activation ratios and values of flux above arbitrary energies that help to characterize the spectrum. Fluxes are given

TABLE XX. Flux and Reaction-rate Data; EBR-II 4-C-1 Core Midplane (per MW of reactor power)

	Calculated (act/sec/ 10^{24} atoms)	Measured (act/sec/ 10^{24} atoms)
<u>Reactions</u>		
$\text{Fe}^{54}(\text{n}, \text{p})\text{Mn}^{54}$	$7.84 \times 10^{11*}$	7.84×10^{11}
$\text{Ni}^{58}(\text{n}, \text{p})\text{Co}^{58}$	8.86×10^{11}	9.81×10^{11}
$\text{Ti}^{46}(\text{n}, \text{p})\text{Sc}^{46}$		6.5×10^{11}
$\text{Cu}^{63}(\text{n}, \alpha)\text{Co}^{60}$		
$\text{Fe}^{54}(\text{n}, \alpha)\text{Cr}^{51}$	1.0×10^{10}	
U^{235} fission		6.98×10^{13}
U^{238} fission	3.02×10^{12}	2.46×10^{12}
<u>Ratios</u>		
$\text{Ni}^{58}(\text{n}, \text{p})/\text{Fe}^{54}(\text{n}, \text{p})$	1.13	1.25
$\text{U}_f^{238}/\text{Fe}^{54}(\text{n}, \text{p})$	3.86	3.14
<u>Damage Rates</u>		
RDU, radiation dose units ²⁰	2.195×10^{13}	
Dt, displacements/unit time ²¹	2.432×10^{16}	
SPIKE ²²	3.324×10^{13}	
<u>Fluxes</u>		
$\phi > 1 \text{ MeV}$	7.79×10^{12}	
$\phi > 0.5 \text{ MeV}$	1.5×10^{13}	
$\phi > 0.183 \text{ MeV}$	2.48×10^{13}	

*Used for normalization of calculated fluxes.

²⁰Rossin, A. D., Dosimetry for Radiation Damage Studies, ANL-6826 (March 1964).

²¹Shure, K., Radiation Damage Exposure and Embrittlement of Reactor Vessels, Nucl. Applications 2, 106 (April 1966).

²²Beeler, J. R., and McGurn, J. L., Computing Primary Knock-on Spectra and Populations in Polyatomic Material Irradiated by Neutrons, GEMP-101 (1961).

per megawatt of reactor operating power. Thus they may be normalized to a particular power level or multiplied by total megawatt-seconds of operation to obtain fluence. This table is an example of data being prepared for a typical experimental location in EBR-II. It is for subassembly 4-C-1 in Row 4 at the reactor midplane; 4-C-1 has been used for irradiations of prospective fast reactor jacket materials.

Knowing the spectrum, one may calculate damage rates on the basis of any model. Here we show the RDU^{20} and Dt^{21} (based on energy available to produce displacements), a "spike" model, based on the work of Beeler,²² and "flux > 1 MeV." These can be compared throughout the reactor core using tables, profiles, and flux maps.

At EBR-II temperatures much of the embrittlement observed in cladding materials may be caused by lattice displacements. Since loss of ductility at elevated temperatures may be attributed to helium formation, the rate of the $Fe^{54}(n,\alpha)Cr^{51}$ reaction is also reported. However, this must be taken as an estimate since the cross section is not well known.

b. Effect of Fast-neutron Irradiation on Jacket Materials. A total of 103 tube-burst and 420 tensile-type specimens of V-20 w/o Ti, V-15 w/o Ti-7.5 w/o Cr, AISI Type 304 stainless steel, Hastelloy-X, and Inconel 625 are being irradiated in EBR-II in order to determine the effects of fast-neutron irradiation at elevated temperature on their mechanical properties. The irradiations are being conducted at temperatures between 500 and 650°C in EBR-II subassemblies XA08, XO10, XO16, XO18, and XO20. The status of the capsules did not change in this report period. The identification of the capsules is given in Table XXI.

Seventeen capsules, AS-35 through 49 and AS-54 and 55, from subassembly XO13, which were irradiated in sodium at 650°C to a maximum total neutron exposure of 7.5×10^{21} n/cm², have been disassembled and their contents removed. The capsules contained a total of 54 tensile specimens of V-20 w/o Ti that were given one-hour final vacuum anneals at three different temperatures: 900, 1000, and 1100°C; 54 tensile specimens of V-15 w/o Ti-7.5 w/o Cr, which were given equivalent treatments at 850, 1100, and 1250°C; and 27 and 18 tensile specimens of commercially heat treated AISI Type 304 stainless steel and Hastelloy-X, respectively. A complete description of these capsules was given in the Progress Report for September 1966, ANL-7255, pp. 46-47.

Preparations are being made to analyze the temperature monitors and the flux monitor wires of iron, nickel, and titanium. Since the activity of the Type 304 stainless steel and the Hastelloy-X was between 2.5 to 3 R/hr at one foot, these specimens will be tested in a hot cell. The vanadium-base alloy specimens will be tested in an unshielded area since their activities ranged from only 25 to 100 mR/hr at one foot. Two 20,000-lb-capacity Satec Model D creep-rupture machines are being set up

in a shielded cave for testing the Type 304 stainless steel and Hastelloy-X specimens. Five similar creep-rupture machines and one 5000-kg-capacity Instron tensile tester are being set up on a low-activity area for testing the vanadium-base alloy specimens.

TABLE XXI. Status of Cladding Materials Irradiations in EBR-II

S/A No.	Capsule Number	Design Parameters				Operating Conditions	
		Cladding Composition	Type of Specimen	No. of Specimens	Specimen Environment	Max Specimen Temp (°C)	Exposure to Date (n/cm ²)
XA08	AS-1	V-20 w/o Ti	Tube-burst	12	Argon-helium	540	3.4 x 10 ²²
XA08	AS-2	V-20 w/o Ti	Tube-burst	12	Argon-helium	540	3.4 x 10 ²²
XA08	AS-3	Hastelloy-X	Tube-burst	12	Argon-helium	540	3.6 x 10 ²²
XA08	AS-4	Hastelloy-X	Tube-burst	12	Argon-helium	540	3.6 x 10 ²²
XA08	AS-5	304 SS	Tube-burst	12	Argon-helium	540	3.6 x 10 ²²
XA08	AS-6	V-20 w/o Ti	Tensile	16	Argon-helium	580	3.7 x 10 ²²
XA08	AS-7	Hastelloy-X	Tensile	16	Argon-helium	580	3.4 x 10 ²²
XA08	AS-8	304 SS	Tensile	16	Argon-helium	580	3.4 x 10 ²²
XA08	AS-12	V-20 w/o Ti	Tensile	16	Argon-helium	580	3.3 x 10 ²²
XO10	AS-16	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.2 x 10 ²²
XO10	AS-17	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.3 x 10 ²²
XO10	AS-18	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.2 x 10 ²²
XO10	AS-19	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.2 x 10 ²²
XO10	AS-20	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.4 x 10 ²²
XO10	AS-21	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.6 x 10 ²²
XO10	AS-22	Hastelloy-X	Tensile	13	Argon-helium	500	1.4 x 10 ²²
XO10	AS-23	304 SS	Tensile	13	Argon-helium	500	1.6 x 10 ²²
XO10	AS-24	304 SS	Tensile	13	Argon-helium	500	1.4 x 10 ²²
XO10	AS-25	304 SS	Tensile	13	Argon-helium	500	1.3 x 10 ²²
XO10	AS-26	304 SS	Tensile	13	Argon-helium	500	1.3 x 10 ²²
XO16	AS-29	304 SS	Tensile	9	Sodium	650	9.1 x 10 ²⁰
XO16	AS-30	V-15 w/o Ti-7.5 w/o Cr	Tensile	9	Sodium	650	8.6 x 10 ²⁰
XO16	AS-31	V-20 w/o Ti	Tensile	9	Sodium	650	8.2 x 10 ²⁰
XO16	AS-32	Hastelloy-X	Tensile	9	Sodium	650	9.3 x 10 ²⁰
XO16	AS-33	V-15 w/o Ti-7.5 w/o Cr	Tensile	9	Sodium	650	8.9 x 10 ²⁰
XO16	AS-50	V-15 w/o Ti-7.5 w/o Cr	Tensile	9	Sodium	650	8.5 x 10 ²⁰
XO16	AS-51	V-20 w/o Ti	Tensile	9	Sodium	650	8.1 x 10 ²⁰
XO16	AS-52	Hastelloy-X	Tensile	9	Sodium	650	9.1 x 10 ²⁰
XO16	AS-53	V-20 w/o Ti	Tensile	9	Sodium	650	8.3 x 10 ²⁰
XO18	AS-56	V-20 w/o Ti, V-15 w/o Ti-7.5 w/o Cr	Tensile, Tube-burst	75	NaK	650	5.3 x 10 ²¹
XO18	AS-57	304 SS, Hastelloy-X	Tensile, Tube-burst	75	NaK	650	5.3 x 10 ²¹
XO18	AS-58	V-20 w/o Ti, V-15 w/o Ti-7.5 w/o Cr	Tensile, Tube-burst	25	NaK	650	5.3 x 10 ²¹
XO20	AS-59	V-20 w/o Ti	Tensile	9	Sodium	650	5.3 x 10 ²⁰
XO20	AS-60	V-20 w/o Ti	Tensile	9	Sodium	650	5.3 x 10 ²⁰

Seven additional capsules are to be shipped to Lemont for disassembly and postirradiation testing. The capsules are AS-34 from subassembly XO13, which contained nine tensile specimens of Hastelloy-X, capsules AS-9, AS-10, and AS-11 from subassembly XA07, which contained 16 tensile specimens each of V-20 w/o Ti, Hastelloy-X, and Type 304 stainless steel, and capsules AS-14, AS-15, and AS-27, which contained a total of 26 tensile specimens of V-20 w/o Ti, and three and ten tensile specimens, respectively, of Hastelloy-X and Type 304 stainless steel. The description and irradiation conditions of subassemblies XA07 and XO09 were given in the Progress Report for November 1966, ANL-7279, pp. 56-57.

Inner capsule tubes (33.5 in. long) from irradiation of structural materials in EBR-II will be used for tube-burst tests over a range of temperatures. Flux and temperature variations along their lengths existed during the irradiation exposure because of distribution patterns within the reactor.

Seventeen inner capsules from subassembly XO13 have been returned to ANL, Illinois, and have been removed from the capsules for subsequent sectioning into 4-in. lengths. Four of the tubes are Inconel 625, three are Type 304 stainless steel, two are Hastelloy-X, and eight are V-15 w/o Ti-7.5 w/o Cr.

Other irradiated structural materials capsules from subassemblies XA07, XO09, and XO13 are out of the reactor and are to be shipped to ANL, Illinois. Inner capsule tubes in these capsules are as follows: XA07, three Hastelloy-X tubes; XO09, three Hastelloy-X tubes; and XO13, one Hastelloy-X tube.

4. Techniques of Fabrication and Testing

a. Ultrasonic Instrument and Transducer Development. Porous tungsten samples made with 100 to 400 μ particles were reported (see Progress Report for January 1967; ANL-7302, p. 67) to require a density of less than 70% to match the acoustic impedance of lead zirconate-titanate transducers. Argonne has made a 70.8% dense sample. Also, samples 60, 70, and 80% dense, received from Sintered Specialties, were being evaluated.

Evaluation of the porous tungsten samples has continued. Longitudinal velocities in these -325 mesh samples have been measured, and the resultant acoustic-impedance values have been calculated. Theoretically, an acoustic-impedance match with Clevite's PZT-4 type of lead zirconate-titanate is attained at a theoretical density of about 63%.

Further evaluation of sintered porous tungsten as a backing material for ultrasonic transducer probes will be delayed until lower-density samples (below 60%) and PZT-4 elements are received. Lower-density samples appear to be necessary for Gulton's HDT-31 type of lead zirconate-titanate. The acoustic impedance of HDT-31 will be obtained from the manufacturer or determined by measurements on typical elements. Lower-density samples will also be useful as backing media for lead metaniobate.

A 60% dense tungsten sample has been used as a backing medium for both PZT-4 and HDT-31 piezoelectric elements. Best results

were observed (short initial pulse and minimum background noise) when the sample was applied to the PZT-4 element. Unfortunately, the quality of the PZT-4 element used in the test is unknown.

b. Development of a Neutron-Image Intensification System. The track-etch neutron image detection method has been considered (see Progress Report for January 1967, ANL-7302, p. 69) as a means of improving the n- γ response and of storing image information. This detection method appears to have many properties that make it attractive for use with low-level neutron intensities. The technique depends upon charged particles causing radiation damage in insulating materials.²³ The damaged areas can then be selectively etched.

For neutron detection one can consider conversion materials that convert neutrons into alpha particles or into fission fragments for use with this technique. We have evaluated the latter technique in which enriched uranium foils, 0.025 mm thick, are used with cellulose nitrate films of similar thicknesses. Good-quality neutron images have been obtained for total thermal-neutron exposures of 2×10^9 n/cm². Deviations from this exposure by an order of magnitude in either direction yield poorer results. Underexposures yield a mottled image after the plastic is etched (in 6.5 N solution of sodium hydroxide at 55°C); overexposures yield less contrast. Etch time is also an important variable. A typical etch time is 5 min. Longer etching periods yield less contrast; shorter periods yield a mottled image.

Best results thus far have produced images which display a high contrast resolution of better than 0.025 mm and a contrast sensitivity of about 10%. The contrast result is poorer than one would like. Methods of enhancing contrast are now under study.

The attractive features of this method of low-intensity-neutron detection are insensitivity to gamma radiation and amenability to indefinite integration of signal information. The method is capable of good resolution and offers the conveniences of light insensitivity and immediate image development. The method will be pursued to determine how it compares with the neutron-image intensifier for detection of low-intensity neutrons.

Measurements of the thermal conductivity of irradiated fuel were being held up by repair operations on a hot-cell hoist in the cave area. The high-temperature set-up has now been reassembled in the cave area. Operation of the system is being checked out before measurements on radioactive materials are started.

²³Fleischer, R. L., Price, P. B., and Walker, R. M., Tracks of Charged Particles in Solids, Science 149, 383-393 (July 23, 1965).

C. Engineering Development

1. Electric Master-Slave Manipulator, Mark E4A

The new synchro gear system (see Progress Report for December, 1966, ANL-7286, p. 48) was released for fabrication, and parts for two manipulator servo drive units have been built. Tests will be performed on these two units to determine if the problem of inadvertent "jumping" of positional nulls has been eliminated. Additional gain will probably be required to get sufficient signal amplitude; this could be easily obtained by adding inexpensive operational amplifiers.

The complete brake system has been released for fabrication with a requested delivery schedule of 17 April 1967. Bench testing of one unit of the brake system will be started as soon as components are received. If preliminary tests are satisfactory, the tests on this unit will be extended into a life test. We expect to have brakes assembled on all seven servo drive units in June.

Modifications to the slave servo drive units are now being made to accommodate the new synchro and brake components.

2. Heat Transfer, Fluid Flow, and Mechanics of Materials

a. Niobium-1% Zirconium Loop. Recalibration of the absolute-pressure transducers is complete. Flowmeter calibrations are under way, pending resolution of some erratic thermocouple readings. Some of the tantalum condenser shutters have been interfering with the condenser tubing and have caused the shutter drive to shear; a repair is being completed. The shutters will be operated from 0 to 75% open to avoid interference.

b. Heater Experiments

(i) Electron-bombardment Heater (EBH) Experiment. Further EBH tests will be carried out in the new EBH test facility that is being constructed (see Progress Report for January 1967, ANL-7302, p. 71). It will enable tests of heaters to their maximum heat flux capabilities before they are installed in the Nb-1% Zr loop.

c. Heat Transfer in Double-pipe Heat Exchangers

(i) Countercurrent Turbulent Liquid-metal Flow. Twenty-one runs were completed with the second heat-exchanger test section. The objective was to investigate whether the experimentally determined heat-exchanger efficiencies (effectiveness) and fully developed overall heat-transfer coefficients depended on the fluid inlet-temperature difference.

This anomalous dependence on inlet-temperature difference was found with operation of the first test section, as well as with previous cocurrent-flow experiments (see Progress Report for September 1966, ANL-7255, p. 55).

Fig. 14. Comparison of Data from First and Second Mercury-Mercury Counterflow Heat-exchanger Test Sections

The data obtained show that the dependence of heat-exchanger efficiencies on inlet-temperature difference has been eliminated with the second test section. This test section is longer than the first, has "improved" inlet fittings, and does not have the air-space-insulated flow-calming section. A comparison of data from the first and second test sections at similar operating conditions is shown in Fig. 14. However, a completely satisfactory explanation of the inlet-temperature effect with the first test section, and its elimination with the second test section, is still not available (see Progress Report for January 1967, ANL-7302, pp. 71-72).

Although the inlet-temperature effect has been eliminated with the second test section, the experimentally determined fully developed overall heat-transfer coefficients are generally too low and reproducibility is poor (data scatter as much as 30% in some runs). The cause is being studied. One possibility is that the added heat-exchanger length, together with the low Reynolds numbers in the annular space, resulted in nearly laminar or transition-region flow. Operation at higher flowrates is in progress to test this possibility.

Extensive computations for the mathematically "degenerate" case of equal-heat-capacity flowrates on both sides of the exchanger have been completed. Figure 15 shows typical results on these computations as related to local heat-transfer coefficients and heat fluxes. The dimensionless distance parameter z , shown as the abscissa in Fig. 15, is defined by

$$z = (4/Pe_1)(l/D_1),$$

where Pe_1 is the tube-side Peclet Number and (l/D_1) is distance from the tube inlet measured in tube diameters. The dimensionless total length of the exchanger is represented by Z . Note that there are both thermal entrance and exit regions. For the equal-heat-capacity flowrate case, the existence of these regions results in actual overall heat-transfer rates larger than would be predicted by customary engineering methods using fully developed heat-transfer coefficients. With liquid metals, the difference

between actual heat-transfer rates and those predicted by customary methods becomes important at efficiencies less than about 0.7; the differences become less important as the thermal resistance through the wall increases.

Fig. 15. Local Heat-transfer Coefficient and Heat Flux Normalized with Respect to Fully Developed Uniform Flux Values for a Counterflow Concentric-tube Heat Exchanger with $H = 1$, $K = 0.1$, and $K_w = 0$

d. Core Structural Dynamics. A survey report on vibration of reactor core components was drafted and is being reviewed.

A method of approach for our study has been identified and considered preliminarily. It involves treating the cylinder flow-induced-vibration response problem as a problem in random vibration. The random excitation mechanism is postulated to consist of the fluid pressure fluctuations caused by normal nonuniformities in the flow passage, flow separation, and turbulence. For the initial investigation, a linear system is assumed such that the response of the cylinder can be represented by the linear differential equation

$$M\ddot{y} + C\dot{y} + L(y) = p(\bar{x}, t),$$

where M is the line density including the virtual mass of the fluid, C is the equivalent viscous damping coefficient, $L(\cdot)$ is the appropriate linear differential operator including fluid as well as elastic effects, and $p(\bar{x}, t)$

represents a space- and time-varying random excitation. As an example, if Paidoussis' equation of motion²⁴ for a cylinder in axial flow were adopted, the differential operator would be of the form

$$L(\cdot) = \left\{ EI \frac{\partial^4}{\partial x^4} + [(m_f U^2 - T_0) - \frac{1}{2} \rho D U^2 C_f (\frac{1}{2} \ell - x)] \frac{\partial^2}{\partial x^2} + 2m_f U \frac{\partial^2}{\partial x \partial t} + \rho D U^2 C_f \frac{\partial}{\partial x} \right\},$$

where EI is the flexural rigidity, m_f is the virtual mass of fluid per unit length, U is the mean axial flow velocity, T_0 is the longitudinal tension applied at supports, D is the diameter of the cylinder, ρ is the fluid density, C_f is the frictional drag coefficient, and ℓ is the cylinder length.

The use of correlation functions in space, in time, and in space-time is being studied to describe statistically the random pressure fluctuations in the flow. As a method of solution to determine the response, a normal mode method and a Green's function approach are being considered.

In general, the random-vibration approach looks promising and seems to be the most realistic method for describing the flow-induced, random response of the structure.

The new flow loop has been completed and, after minor modifications, satisfactorily operated. A 235-gal accumulator has been fabricated and installed.

Various capacitive and inductive displacement-sensing probes and transducer circuitry were studied.

e. Anisotropy Studies. To provide refined analytical methods for determining thermally induced displacements and stresses in reactor core components, study has begun of the significance of effects introduced by (a) intrinsic anisotropic material properties of fuel elements and (b) regular substructure patterns, including how they affect the gross anisotropy of larger structural elements. Particular emphasis will be placed on the latter cause of anisotropic behavior and its possible utility in developing models to describe the overall thermal deflection of fuel-element subassemblies and the entire core structure.

In the first phase of this study, consideration has been restricted to rotationally symmetric, steady-state thermoelasticity problems for transversely isotropic materials. The problems analyzed have been primarily

²⁴Paidoussis, M. P., "Vibration of Flexible Cylinders with Supported Ends, Induced by Axial Flow" (to be published in 1965-1966 Proceedings of the Institution of Mechanical Engineers, Volume 180, Part 3J).

ones chosen to reveal analytical and numerical difficulties associated with the introduction of anisotropic effects and to demonstrate the degree to which anisotropy can qualitatively and quantitatively alter thermal-stress distributions.

A typical example of a sample problem solution designed to compare the stress fields in isotropic, near isotropic, and strongly anisotropic materials is shown in Fig. 16. Illustrated in dimensionless form is the axial variation of the axial stress component at the center of a right circular cylinder whose surface has a segment of its length maintained at unit temperature while the remainder of the surface is at zero temperature.

Fig. 16. Variation of Axial Stress Component along Cylinder Centerline

In addition to the isotropic solution, we have plotted the equivalent stress component for two transversely isotropic materials whose axes of symmetry lie along the cylinder axis. In the case of a single crystal of magnesium (a nearly isotropic material), we see that the stress distribution is almost identical with the isotropic results. The third curve refers to a hypothetical com-

posite material whose properties in the axial direction differ significantly from those in the direction transverse to the cylinder axis. It is anticipated that such wide variations in material properties with direction will be typical in cases where anisotropic models are used to describe entire core structures (e.g., loosely interconnected hexagonal packings of fuel-element subassemblies). Thus the composite-material graphically illustrates how radically stress distributions can differ from those obtained by isotropic analysis.

In addition to the solution of sample problems, an extensive study is underway of recent developments in the field of anisotropic thermoelasticity. This study emphasizes recent analytical results likely to provide tractable methods for numerically evaluating specific problem solutions.

D. Chemistry and Chemical Separations

1. Fluoride Volatility Processes

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

(i) Fluorination of UO_2 - PuO_2 -Fission Product Pellets. Development studies are being performed in a 2-in.-dia fluid-bed reactor to

establish optimum conditions for the recovery of uranium and plutonium from UO_2 - PuO_2 pellets containing fission products. In the process, the UO_2 - PuO_2 -fission product pellets are first oxidized to U_3O_8 - PuO_2 fines by reaction with oxygen. Next, the uranium is selectively converted to volatile UF_6 by reaction of the oxide fines with BrF_5 . In this step, plutonium is converted to nonvolatile PuF_4 . Recovery of plutonium as volatile PuF_6 is effected in a subsequent step by reaction of PuF_4 with fluorine. An experiment (Purse-15) was performed, as one of a series of experiments, to determine further the effect of the temperature of the fluorination with BrF_5 upon plutonium retention in the alumina bed. Previous experiments in this series employed temperatures of fluorination with BrF_5 of 200, 250, 300, and 400°C (see Progress Report for December 1966, ANL-7286, p. 51), and the results of these experiments indicated that the best temperature for conducting the fluorination with BrF_5 is within the range from 250 to 300°C. In run Purse-15, the fluorination was carried out at 350°C. The charge to the fluid-bed reactor consisted of 647 g pellets and 1100 g alumina containing 0.6 g CsF, 0.16 g RbF, and 0.5 g NpO_2 . The processing sequence

consisted of: oxidation of the pellets for 4 hr at 450°C with 20 v/o oxygen, fluorination with 8 to 10 v/o BrF_5 at 350°C for 2 hr, and fluorination with 90 v/o fluorine for 3 hr at 300°C, 5 hr while the reactor temperature was increased from 300 to 550°C, and 3 hr at 550°C:

Fig. 17. Concentration of Uranium, Neptunium, and Plutonium in Alumina Fluid Bed during Fluorination with BrF_5 Followed by Fluorine

The course of uranium, plutonium, and neptunium volatilization during both fluorination steps, as determined by periodic analysis of bed samples, is shown in Fig. 17. During the BrF_5 fluorination step, 99.5% of the uranium and 60% of the neptunium in the charge volatilized. An average UF_6 production rate of 39 lb UF_6 /(hr)(sq ft) and an average efficiency of BrF_5 utilization of 32% were observed. A peak UF_6 production rate of 95 lb UF_6 /(hr)(sq ft) was achieved during the initial 0.3 hr of the fluorination step.

The final alumina bed contained 0.008 w/o plutonium, and 0.002 w/o neptunium (see Fig. 17). These values correspond to the removal of more than 99.9% of the uranium, 97% of the plutonium, and 95% of the neptunium present in the charge. (Removal of 99% of the plutonium can be achieved by reuse of the alumina bed; see Progress Report for October 1966, ANL-7267, p. 66.) The residual uranium and plutonium concentrations are nearly identical to those obtained using a BrF_5 fluorination temperature of 250°C. Therefore, it appears that, within the range from 250 to 350°C, the

removal of plutonium from the alumina bed is not affected by the temperature of the BrF_5 fluorination step, and that the final choice of the reaction temperature between these limits may be governed by other parameters such as UF_6 production rates and BrF_5 utilization efficiencies. Experiments to define these reaction parameters are being carried out in engineering-scale equipment.

(ii) Neptunium Fluoride Chemistry. Laboratory-scale studies are being carried out to determine the behavior of neptunium when oxidized nuclear fuel is reacted with BrF_5 . Current emphasis in these studies is on evaluating the selective reduction of NpF_6 to NpF_4 by elemental bromine as a method for separating uranium from neptunium in UF_6 - NpF_6 mixtures. Previous studies (see Progress Report for January 1967, ANL-7302, p. 76) have indicated that the reaction between NpF_6 and bromine proceeds slowly at 30°C . Recent experiments were performed at 75°C with the reactants partially in the vapor and partially in the condensed phases. In these experiments, approximately equal amounts (120 mg) of UF_6 and NpF_6 were contacted with excess bromine for one hour. The results indicated that neptunium is rapidly and completely reduced to solid NpF_4 under these conditions. Since small amounts of uranium (up to 7 mg) were found in the NpF_4 product, additional treatment of the solid product may be required to obtain neptunium of high purity.

(iii) Instruments for Plutonium Analysis. An experimental program is under way to develop techniques for plutonium analysis based on the counting of neutrons derived from the α, n reaction of plutonium alpha particles with light-element nuclei such as fluorine. Two types of neutron detectors are being investigated. One type is a portable detector for locating and estimating plutonium deposits in process lines and equipment. The other is being used for quantitative determination of samples derived from fluoride volatility process operations.

Interference from extraneous neutron sources must be avoided or minimized if portable detectors are to find wide application in plutonium analysis. Several experiments were performed to examine the effect of shielding on the directional response of the instrument. The angular responses of two detectors were measured. These instruments incorporated a moderated, 1 in. in dia by 2 in. long proportional detector tube filled with 10 atm of helium-3. The helium-3 tube and the paraffin moderator were eccentrically placed in cylindrical geometry to obtain a balance between maximum and minimum responses in opposite radial directions. The small shielded detector, $6\frac{3}{8}$ in. in dia by 6 in. long, weighed 9.3 lb, and the large one, $11\frac{1}{4}$ in. in dia by 6 in. long, weighed 27 lb. The shield for the small detector contained a 0.062-in.-thick cadmium sheet and a layer of paraffin. The shield for the large detector was a 0.062-in.-thick sheet of cadmium and water. Since the cylindrical ends of these

detectors were not shielded, the angular response was determined with the detectors at least twice the distance from any other moderating material than from the source. Angular response, obtained by rotating each shielded detector on its axis at each of two distances from a 48-g source of PuF_4 , was corrected by application of the inverse-square law to obtain response at a constant distance between the source and the helium-3 tube at all angles.

The response of both detectors was observed to decrease with rotation, as greater thicknesses of shield were positioned between the source and the helium-3 tube. In the large detector, an increase by a factor of two or three in the thickness of cadmium in the shield did not significantly alter the angular response. Ratios of responses in one direction to those in the reverse (180°) direction were 3.4 for the small detector and 5.2 or 6.3 for the large detector, depending upon the distance between the source and the helium-3 tube. Although both detectors have similar responses at detection angles in the reverse direction, the large detector appears to offer better angular discrimination at forward angles of less than 90° . Both detectors, particularly the small detector owing to its light weight, appear promising for use in locating plutonium deposits in process equipment during laboratory and pilot-plant operations.

The instrument that is being developed for quantitative determination of the plutonium content of a sample consists of four 1-in.-dia by 12-in.-long proportional detectors, spaced around a cylindrical sample well. The counters are submerged in water which acts as a moderator for the fast neutrons from the sample. The detector geometry is approximately 4π , and about 10% of the neutrons from the plutonium fluoride are counted when 2 in. of moderator (water) separates the detector tubes and the sample well.

It was observed in the evaluation tests that background counts were highly dependent on the presence of plutonium fluoride in the same room with the detector. Modest gamma-ray sources from any of the samples tested did not interfere with the detection of neutrons. Also, the analyses were not influenced by the nature of the sample container, whether glass, metal, or plastic.

Although the counting rate of a sample was observed to vary with the position of the sample in the cylindrical well, a relative standard error of 0.26% in counting could be achieved by carefully positioning the sample in the well. Several materials, representing samples of interest in fluoride volatility processing of nuclear fuel materials, have been analyzed by this instrument with a relative standard error from 1 to about 6%. Samples of alumina-bed material containing as little as 5 mg PuF_4 were analyzed for plutonium content within the standard error of 6%. It was observed that the presence of americium-241 can affect the absolute

counting rate and therefore the apparent plutonium content of the sample. Also, the absolute counting rate was found to be influenced by the specific alpha activity of the plutonium isotopes present in the sample. Further development work on the use of this instrument for the analysis of laboratory samples is continuing.

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

(i) Engineering-scale Alpha Facility. Studies are being performed in the engineering-scale alpha facility to demonstrate handling of PuF_6 on an engineering scale and to determine the feasibility of fluoride volatility flowsheets. Current work involves the erection and preliminary testing of equipment which will be used in experiments in which nuclear reactor fuels will be fluorinated with BrF_5 . The equipment includes a series of 4-in.-dia traps filled with activated alumina and soda lime for the disposal of UF_6 and BrF_5 . These traps are being erected in a mockup area and, upon completion of performance tests, will be installed within the alpha enclosure.

The feed system for BrF_5 has been installed in an enclosure adjacent to the alpha box. This facility includes a heating system for the BrF_5 supply vessel, a flow control valve, and a system of temperature and pressure safety controls.

(ii) Fluid-bed Studies with Irradiated Fuels. A series of experiments are under way in a $1\frac{1}{2}$ -in.-dia fluid-bed reactor to determine the distribution of fission products during the processing of highly irradiated UO_2 fuel by the oxygen-bromine pentafluoride-fluorine process. Two experiments have been described previously (see Progress Reports for September 1966, ANL-7255, p. 60, and November 1966, ANL-7279, pp. 70-71). Two additional experiments (BRF-3 and BRF-4), which employed essentially the same processing conditions as those used in the previous experiments, have been performed to evaluate techniques for purifying the plutonium product. The processing sequence consisted of: oxidation of the irradiated oxide fuel with 20 v/o oxygen for 2 hr at 400°C , fluorination of the oxidized fuel with 10 v/o BrF_5 for 1.5 hr at 300°C (run BRF-3) and for 3.0 hr at 350°C (run BRF-4), and fluorination with 90 v/o fluorine for 10 hr as the reactor temperature was increased from 300°C (run BRF-3) or 350°C (run BRF-4) to 550°C .

Preliminary observations on the distribution of fission products during run BRF-3 indicated that, except for ruthenium, the fission product distribution compared closely with those observed in the previous runs. About 3% of the ruthenium volatilized during the oxidation step, as compared with previous values of 0.0002 and 0.17%. Also, about 14% of

the ruthenium volatilized with the PuF_6 , while previously only about 3.5% of the ruthenium volatilized. Attempts to evaluate a plutonium-purification scheme involving sublimation of PuF_6 at 0°C were thwarted in run BRF-3 by operational difficulties.

The plutonium-purification operation was successfully completed in run BRF-4. This operation involved condensing the PuF_6 stream leaving the reactor at -78°C , subliming the PuF_6 at 0°C in a nitrogen carrier gas stream, and then thermally decomposing the PuF_6 to solid PuF_4 on a bed of refractory alumina. Preliminary analysis of the PuF_4 -alumina bed indicated that high decontamination was achieved from the gamma-active fission products, cerium, cesium, zirconium, niobium, and antimony. Decontamination of the plutonium product from ruthenium was slight, and a further purification step involving contact of the PuF_4 -alumina bed with BrF_5 at 300°C is planned in order to remove the residual ruthenium.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. General

The specification for primary pumps has been completed, approved, and sent to vendors for bids. Revisions to the heat-exchanger specifications are being completed.

Burns and Roe, the project architect-engineer, has been directed to proceed with detailed design of all of the facility within its scope of work for the balance of Title II.

The construction schedule and price of the reactor pressure vessel are being negotiated with Combustion Engineering.

The drawings and specifications for the control-rod guide assembly have been completed. This is the first procurement package on the reactor-vessel internals.

2. Corrosion in the Secondary Coolant Loop

Coupons of ten different materials considered for the AARR water-cooled heat exchangers were exposed to the ZGS (Zero Gradient Synchrotron) coolant stream for a total of 270 days in a "customers service" test rendered by International Nickel Co.

The ZGS coolant loop uses water of the same origin and treated condition as are specified for the AARR secondary coolant loop (see Table XXII). Three test spools, each containing 20 coupons of the ten candidate materials, were installed in the ZGS coolant loop in April 1966. The average water temperature was 75°F, varying between 60 and 85°F, and the flow velocity past the spools was 4 to 5 ft/sec. The test spools were retrieved after 90, 183, and 270 days. Test results, as assessed by International Nickel Co., are shown in Table XXIII.

Investigations of the latent risk of chloride-actuated stress-corrosion cracking of susceptible candidate materials were beyond the scope of these tests.

The pitting found in the aluminum and aluminum-clad test specimens is probably attributable to the presence of copper ions in the water (the ZGS coolant loop contains heat-exchanger tubes of admiralty and other high-copper alloys). The measured corrosion rates of all materials, especially the carbon steel, were remarkably low, which seems to indicate good

corrosion-inhibiting properties of the low chromate-zinc water-treatment formula. However, the higher temperatures (85-105°F) specified for the AARR cooling water might result in appreciably faster corrosion.

TABLE XXII. ZGS Coolant-loop Water Analysis for 1966.

Characteristic	Avg (mg/liter)	Range (mg/liter)	
		From	To
Hardness, total as (CaCO ₃)	1060	710	1320
Calcium (Ca)	315	192	392
Magnesium (Mg)	78	56	93
Alkalinity to "P" as (CaCO ₃)	0	0	0
Alkalinity, total, "M" as (CaCO ₃)	28	11	64
Chloride, (NaCl)	463	326	618
Phosphate (PO ₄)	2.1	0.8	5.1
Sulphate (Na ₂ SO ₄)	1509	992	1946
Silica (SiO ₂)	10.6	8.4	12.5
Nitrate (NaNO ₃)	130	71	208
Sodium (Na)	227	126	310
Iron (Fe)	-	-	-
Aluminum (Al)	-	-	-
Turbidity	11.3	4.5	17.0
Potassium (K)	27	19	38
Zinc (Zn)	10.3	5.6	18.3
Chromate (Na ₂ CrO ₄)	8.6	5.0	16.3
Filterable Residue 180°C	1062	1354	2740
Nonfilterable Residue	26	0	65
Total Residue	1994	1387	2427
pH Value	6.9	6.3	7.5

18 psia, heat fluxes to 0.5×10^5 Btu/hr/ft², liquid superheats required to initiate boiling ranging up to $\sim 200^\circ\text{F}$ (ranging up to 400°F for pressures below 1 psia), steady-state boiling superheats from 5 to 30°F , sodium heads ranging from 4 to 16 in. and liquid-sodium temperatures up to 1800°F . Pressure pulses of up to 200 psia have occurred both during the initiation of boiling and during steady-state boiling.

Additional heaters are being installed to extend the range of heat flux.

3. Critical Flow

Calculations indicate that critical flow may occur in the core of a sodium-cooled reactor during an accidental power surge. Critical flow could lead to detrimental effects such as voidage of the coolant channel, shock phenomena, and pressure buildup.

The sodium loop has operated successfully up to 1550°F and critical-flow data have been obtained, including exit critical pressure ranging from 0.5 to 5 psia, flowrates up to 1200 lb/sec/ft², and stagnation temperatures up to 1500°F . Subatmospheric steam-water critical-flow data have also been collected with test sections and procedures, identical with those used for the sodium loop. Both sets of data are being analyzed.

4. Electron-bombardment Heater Tests

The operation of various electron-bombardment heaters (EBH), which will be used to simulate in-pile experiments pertaining to reactor safety, is being examined in a large evacuated vessel. Both single- and multi-pin heaters will be built and tested.

Working drawings of the test facility and an EBH section for studying tube flow have been completed. Specifications for the test vessel and ion pump are complete; the EBH section is being fabricated.

5. Convective Instability

Experiment construction is progressing on schedule and, depending on delivery of final components, should be completed within six weeks.

Analytically, the reduced two-dimensional boundary-layer equations have been solved by the method of lines; this solution compares excellently with that obtained by the method of integral relations. Only the Prandtl number was varied widely (variation of Grashof number was limited) because the method of lines was used merely as a check, not for solutions per se, and it is time-consuming on the computer.

As a final step in the solution of the transient free-convection problem with a step increase in wall temperature, more terms will be used in an effort to approach the exact solution. Then the same methods will be used to solve for the case with a step increase in wall heat flux. Finally, if time permits, an effort will be made to obtain a closed-form solution to the initial approximation of the equations for both cases; this would be valuable for studying the effect of pertinent parameters and for use by designers.

6. Thermophysical Properties of Sodium

The literature on the thermophysical properties of sodium is being evaluated to form a basis for recommending "best values" of these properties. Comparisons of safety studies of sodium-cooled fast breeder reactors will then be more meaningful if the various groups doing such studies use standardized data. Recent related experimental work serves as the impetus for this evaluation.

Liquid-density and vapor-pressure data and correlations have been reviewed. For computing the vapor pressure, the Ditchburn equation is recommended for temperatures below 1600°F; and the Stone equation for temperatures above 1600°F. After study of the literature on P-V-T and thermodynamic properties, a computer subroutine was prepared to compute such properties. This subroutine can be used either in any compatible computer program calling for these properties or as the core of a table-making program. Given the temperature and pressure, the program determines the state and outputs the pertinent properties. For saturation conditions, these properties include the density, enthalpy and entropy of the liquid and vapor, the mean molecular weight, composition and compressibility of the vapor, and the heat of vaporization.

The transport properties of both the saturated liquid and vapor phases are also being analyzed. These properties include the thermal conductivity, viscosity, electrical resistivity, and surface tension when appropriate to each phase. Graphical comparisons are being made between reported literature sources for any particular transport property. From stated experimental deviations and the techniques used, a recommended set of values will be given in tabular and graphical form.

7. Component Dynamics

The study of the mathematical model for a parallel-plate coolant-structure system subjected to rapid insertion of energy has been completed, that is, a computer simulation has been achieved and the influence of structural flexibility on the resulting pressure transients has been demonstrated. The results of this preliminary investigation can be summarized as follows:

V. NUCLEAR SAFETY

A. Accident Analysis

1. Linking of the Heat Transfer and Two-phase Coolant-flow Modules

There are two versions of coolant-flow modules, namely, one in which boiling of sodium occurs at the exit of the channel and the other in which boiling is allowed to occur anywhere along the channel. The development of the latter version has been accomplished only recently. Consequently, the linking of the heat transfer and the coolant-flow modules began with the first version of the coolant-flow module. This linking has been accomplished (see Progress Report for January 1967, ANL-7302, pp. 85-86), and some sample problems run. The linking of the second version of the flow module is in progress. Several important investigations of the conditions in a fuel cell, such as, the voiding of sodium during transients are also in progress.

B. Coolant Dynamics

1. Sodium Expulsion

This experiment, in which it is planned to investigate the mechanism of coolant expulsion in a simulated reactor environment, includes measurement of the void distribution, expulsion velocities, pressure transients, and liquid superheats during coolant expulsion.

Design of the eddy-current coils to be used to detect the liquid-vapor interface and void fraction during expulsion is continuing. The linearity of the device has been found to depend on input voltage frequency; at higher frequencies, the system becomes more linear than at lower frequencies. In addition, several problems with the effect of void configuration on output are being investigated.

The hot-trap loop has been fabricated, but assembly must await delivery of the vacuum chamber and electron-beam heater.

2. Superheat

In an experiment to determine the amount of liquid superheat required to initiate nucleate boiling in sodium under various conditions simulating a reactor environment, the independent and combined effects of pressure, dissolved gas, heat flux, surface characteristics, and the pressure-temperature history of the system will be measured.

Approximately 350 test runs have been conducted in a vessel with a 10-micron surface finish. These test runs have involved pressures up to

blades are only ~0.15 in. thick, but the reference absorber matrix is 0.2 in. thick, and (c) the design-reference blades are wider. Therefore, it is adequately confirmed that the design-reference control blades would control at least 15% in reactivity in the fresh reactor; this would be acceptable.

At the phase of design when work was stopped on the SS-UO₂ reactor, it had not been decided whether to attempt to load and unload the core assembly in one piece. The question of safety of fuel handling was at issue. It was intended that, if the entire core assembly were to be moved in one piece, additional shutdown margin would be provided in the form of a solid-absorber insert in the ITC, which would be locked in position during movement of the core assembly. An experiment was performed in the Criticality Facility to determine the control worth of one such absorber insert, namely, a hafnium sleeve at the outer edge of the ITC. A thin hafnium sleeve, only 0.05 in. thick, controlled ~4% reactivity. A more heavily absorbing sleeve, therefore, would control well above 5% reactivity in the fresh reactor.

Thermoluminescent-dosimetry (TLD) measurements of gamma-ray heating rates have been made in radial and tangential "beam" tubes in the beryllium reflector. The phosphor selected was CaF₂(Mn). The experiments were performed using 4 x 4-in. and 6 x 6-in. voided tubes, with monitor detectors in the beryllium reflector at an adequate distance from the tubes. Absolute heating rates of the monitors were inferred which were in good agreement (within ~25%) with calculated heating rates in the unperturbed reflector. At a position corresponding to the inner surface of the reactor pressure vessel, the heating rate in the wall of the beam tube was found to be somewhat greater than twice the heating rate measured at a corresponding position but without the effect of streaming of gamma-rays within the tube. The measured heating rate at this position, 4 ft from the ITC axis, is ~0.06 W/g at the wall of the 4 x 4-in. tangential beam tube; it is ~0.10 W/g for the 6 x 6-in. tangential tube.

Additional measurements were made of the perimetrical distribution of heating rate at the tip (inner end) of the 6 x 6-in. tangential tube. The maximum heating rate at the tip was approximately twice the minimum heating rate there.

These heating rates do not apply directly to the new reference AARR design, with the more lightly loaded, HFIR-type core. At 100 MWt, a larger fraction of the gamma rays will leak from the HFIR-type core with the less-dense aluminum matrix, cladding, and structural material. This is compensated in part by the absence of higher energy gamma rays, namely, capture gammas from the steel. There are other differences as well. Nevertheless, it will be feasible to extrapolate from these measurements to arrive at an adequate description of the corresponding heating rates in the AARR with the HFIR-type core.

A very detailed review has been completed of the radial distribution of region compositions; volume-averaged compositions have been established for 27 regions. Beyond the temporary beryllium, the region compositions depend on the angular direction. Three sets of permanent-beryllium compositions have been selected, one set representative of the beryllium sector containing the bulk of the vertical irradiation facilities, one set representative of the sector containing portions of the through-tubes, and the third set representative of each of the two symmetric sectors containing the blind beam tubes.

With these region compositions established, it was feasible to perform scoping calculations for the design of the lateral biological shield. The digital-computer program MAC was used. Preliminary results indicate that it will be feasible to increase the thickness of pool water between the reactor pressure vessel and the biological concrete zone by 1 ft, so as to provide more access space to beam-tube fittings. It is indicated that the radiation-dose-rate criteria limiting the gamma-ray and neutron-flux intensities at the outer surface of the unperturbed shield will be met without adding steel shielding inside the reactor pressure vessel specifically for this purpose. Also, the heating rates at the pressure vessel and at the inner surface of the lateral biological concrete shield appear to be sufficiently small that such layers of steel shielding are not needed for general thermal shielding. The architect-engineer will be directed to revise the drawings to reflect the decision to increase the radius of the inside surface of the lateral concrete shield from $7\frac{1}{2}$ to $8\frac{1}{2}$ ft. The MAC computations indicate that a 7-ft-thick shield of heavy concrete (density 225 lb/ft³) should be sufficient shielding, for the gross unperturbed shield, at 100 MWt. The architect engineer will perform an independent analysis to determine the shielding adequacy of this system.

b. Experiments. Additional data have been processed from the termination critical experiments with the SS-UO₂ loadings. In a subsequent monthly report, details will be supplied of power-distribution measurements in a simulation of the reference graded-fuel loading. Other measurements included: reactivity-control worths of the 12 control blades, additional shutdown margin supplied by an absorbing sleeve at the outer edge of the internal thermal column (ITC), and TLD (thermoluminescent-dosimetry) evaluation of gamma-ray heating rates along the walls of simulated beam tubes.

In comparison to their control worths in the ungraded fuel loading with the enlarged ITC: (a) there is no clearly discernible change in the average control worth of the six in-core control blades and (b) the six peripheral control blades were approximately 20% less effective. The Criticality Facility control blades are less effective than the design control blades in three respects: (a) hafnium is used in the criticals, but europium, which is more effective, is the reference control absorber, (b) the hafnium

TABLE XXIII. Measured Corrosion Rates of Candidate Materials

Material	Corrosion Rate (m/dm) for Various Exposures			Remarks
	90 days	183 days	270 days	
AISI 304	0.3	0.3	<0.3	
AISI 316	0.6	0.3	0.3	
Inconel 600	1.2	0.3	0.9	
Incoloy 800	3.0	0.6	1.2	
Incoloy 825	3.0	0.9	1.5	
Carbon steel	102.0	49.8	60.0	
Al 6061-T6	3.9	3.0	1.8	Few but broad pits
Al-clad 6061-T6	6.0	2.7	1.8	Single pits; all attack in crevice area
Croloy 16-1	0.3	0.0	0.6	14.82 Cr- 1.23 Ni
USS experimental steel*	1.8	0.3	0.6	

*Material analysis not available; a high Si- or Ni-steel with claimed excellent properties to withstand stress-corrosion cracking.

3. Reactor Physics

a. Analysis. Exploratory studies have been made with the present one-dimensional version of the CANDID program of multigroup diffusion theory on the CDC-3600 computer to determine operating limits on the number of regions and the number of mesh points. Problems could be run successfully for 29 regions or fewer, but with fewer than 150 mesh points. A 27-region spatial partition of the reactor has been selected in the radial direction, leaving the slight flexibility of adding two more regions where needed. Because the fuel is graded within each of the two fuel elements, a fine spatial subdivision is necessary within the fuel zone to obtain radial power distributions. For this reason, a limitation to 29 regions is somewhat restrictive. Nevertheless, it will be possible to perform all multigroup diffusion theory statics calculations presently planned within these limitations of 1-D CANDID. Other digital-computer programs are under review with the intent of making still further refinements in the model of spatial partitioning of the reactor.

1. The simplified pressure-pulse model (heat transfer-thermodynamic relations) satisfactorily predicts pressure transients for saturated or nearly saturated water; it will be used in future studies.
2. The flexural rigidity of the channel walls can markedly affect the system response to a power burst (see Progress Report for January 1967, ANL-7302, Fig. 27, p. 90).
3. For certain input and structural vibration periods, structural oscillations result and are reflected in the pressure transient.

This initial phase of the component dynamics study is being described in a technical memorandum.

C. Fuel Meltdown Studies with TREAT

1. Transient In-pile Loop Experiments with EBR-II Mark-I Fuel

An experimental program, centering on the TREAT reactor, is under way to study the behavior of fast reactor core components under transient heating to fuel failure and beyond. The phenomena under study include pre-failure material movements, fuel-failure thresholds and the mechanisms operative in failure, fuel and coolant movements accompanying failure, and post-failure secondary movements of fuel and coolant.

Experimentation with sodium-bonded, metallic EBR-II Mark-I pins has progressed to the study of fuel meltdown in integral sodium loops designed to hold small clusters of seven rods. Initially, test samples consisted of single fuel pins (see Sect. V.B.2), each surrounded by a ring of six dummy rods. Emphasis was placed on study of prefailure phenomena, failure threshold, and movements associated with failure. More recently, two clusters of seven fuel pins each have been run to begin the scale-up of experimentation into cluster effects, postfailure secondary motions, and survey of coherency of movements. In three experiments, no pump power was on; however, transient-induced flow was possible, representing an increase in realism over stagnant sodium capsules. Both clusters were run with tantalum inserts to shape the sample power axially into a "chopped cosine" with maximum power at the fuel center.

2. Summary and Analysis of Single-pin Results

A total of six EBR-II Mark-I pins were run in the single-fuel-pin loop experiments. Analyses of the results from these experiments on fuel rods surrounded by rings of dummy pins have been made and combined with the earlier data from TREAT experiments on single EBR-II pins in stagnant sodium capsules.²⁵ In combination, these results present a summary of the

²⁵Dickerman, C. E., Sowa, E. S., Monaweck, J. H., and Barsell, A., In-pile Experiments on Meltdown of EBR-II Mark-I Fuel Elements in Stagnant Sodium, Nucl. Sci. Eng. 18, 319 (1964).

knowledge of the behavior of single EBR-II Mark-I pins when subjected to severe transient heating in a sodium environment.

Calculations were made with the ARGUS transient heat transfer code, written in cylindrical geometry.²⁶ Two reference points were obtained radiochemically for the ratio of sample power to TREAT power. Calculations of neutron-flux distributions for a loaded loop inside the reactor were made as necessary. Direct comparison between experimental and calculated sample temperatures was possible only for the first pin, which contained an internal fast-response refractory-metal thermocouple. Agreement was satisfactory. None of the five following sample pins contained internal thermocouples, which could interfere with fuel motion.

Once transient temperatures had been calculated, estimates were made of pressure stresses in the cladding and of cladding penetration by molten fuel, using rate data obtained for the safety program.²⁷ The analytical results are summarized in Table XXIV (in none of the cases was failure due to cladding penetration alone calculated).

TABLE XXIV. Summary of Loop Analyses

Sample	TREAT Transient No.	Sodium Flow (m/sec)	Sample Fuel Energy during Power Peak (J/g)	Experimental Failure Time (sec)	Calculated Failure Time (sec)	Maximum Calculated Fuel-cladding Interface Temperature (°C)
1	743	0	510	-	-	950
2	854	3	710	-	-	1260
3	855	0	930	1.86	1.87	1300 ^a
4	925	1.5	995	-	2.15	1580
5	985	4	720 ^b	-	-	1150
6	1007	4	1240 ^b	1.36	1.42	1640 ^a

^aValue at calculated time of failure.

^bIn central axial section; power was shaped axially.

On the basis of the earlier stagnant sodium experiments and the detailed analyses of the single pin loop tests, the following conclusions can be made:

1. Maximum fuel-cladding interface temperatures $\gtrsim 950^{\circ}\text{C}$ produce extensive alloying of fuel and steel cladding.

2. For the thermal conditions typical of stagnant sodium, an interface temperature of 1000°C marks the approximate failure threshold with cladding penetration by eutectic formation presumably playing the dominant role; cladding rupture due to internal pressure can be predicted using the ARGUS calculations for intact geometry.

²⁶Heestand, J., Schoeberle, D. F., and Miller, L. B., A Method of Calculating Transient Temperatures in a Multi-region, Axisymmetric, Cylindrical Configuration, The ARGUS Program, 1089/RE-248, Written in FORTRAN II, ANL-6654 (1963).

²⁷Walter, C. M., and Kelman, L., Penetration Rate Studies of Stainless Steel by Molten Uranium and Uranium-Fissium Alloy, J. Nuc. Mat. 6, 281 (1962).

3. In the case of flowing sodium,

a. An interface temperature of 1000°C can be greatly exceeded without failure if failure is predicted neither by cladding penetration nor internal pressures.

b. If bond-sodium pressures ≥ 10 atm are calculated, molten fuel will be driven upwards into the expansion space inside the cladding.

c. Upward movement of fuel within the cladding can relieve pressure so that a pressure rupture does not occur. Failure did not occur

when the interface temperature was rising at approximately $4000^{\circ}\text{C}/\text{sec}$ at the time of calculated pressure rupture (Sample 4), but it did occur when the calculated rate was approximately $7000^{\circ}\text{C}/\text{sec}$ (Sample 6).

3. Seven-pin-cluster Sodium Loop Experiments

Transients 1049 and 1050 were run with a cluster identical with that used in Transients 1047 and 1048 (see Progress Report for December 1966, ANL-7286, pp. 70-73), but with the loop pump power off. Transient 1049 was a low-energy input check transient; Transient 1050 was a meltdown experiment, run to provide a no-flow comparison with Transient 1048. Data from the transient instrumentation for Transient 1050 are shown in Fig. 18.

Initially, there was zero flow through the test section. At 1.6 sec, significant transient-induced flow started registering on the flowmeter at the test-section inlet, and average flow downward out of the test region was about 1 m/sec during the time from 1.6 to 1.73 sec. From this average flowrate and for the distance from the fuel center to the inlet thermocouple, it was estimated that 0.20 sec would be required for high-temperature coolant from the sample

Fig. 18. Data for a Seven-pin Cluster during Transient 1050 in TREAT

in the highest power region to reach the thermocouple. This delay, plus a calculated time response of 0.020 sec, indicates that the expulsion was begun by sodium in the temperature range registered by the inlet thermocouple at 1.820 sec, i.e., at about 1000°C.

Cladding failure at about 1.730 sec is indicated by the first pressure pulse and coolant-expulsion spike, which peak at about 1.745 and 1.755 sec, respectively. The rate of coolant expulsion remains at about 4.5 m/sec for approximately 0.080 sec, at which time a second pressure spike and coolant expulsion spike occur. A similar time delay between pressure spikes occurred in Transient 1048. The delay may be associated with failure spread within the cluster or with partial coolant-channel voiding and coolant re-entry; it is still under study.

Following the second spike, coolant flow and pressure decay to unperturbed levels (the negative "draft" shown in the pressure transducer output results from the transducer sensitivity to integrated TREAT flux and is in satisfactory agreement with that measured previously), with a final high pressure spike at about 1.985 sec.

These data are consistent with the semiquantitative model which described previous high-energy tests: sample failure, a period of expulsion(s) and test-section sodium voiding, and a final massive re-entry of sodium resulting in a vigorous pressure spike and sodium expulsion. For this stagnant sodium experiment, the re-entry of sodium seems to be basically sodium falling back down on hot fuel under the influence of gravity.

4. Vacuum-Inert Gas Glovebox

An extensible vacuum/pressure glovebox has been developed to meet the needs of the fast reactor safety experimental program.²⁸ The facility is large enough (107 x 102 x 91 cm) to handle all foreseeable TREAT capsules and operations with high-purity sodium and advanced experimental loops, under conditions of high-vacuum or high-purity inert gas atmosphere.

Vacuum is attained with a turbomolecular pump having a rated pumping speed of 260 liters/sec to 1×10^{-8} Torr and an ultimate capability of $\sim 1 \times 10^{-9}$ Torr. The pump has been connected to the box through a liquid-nitrogen cold trap and a sliding-gate vacuum valve; the vacuum line contains a vibration isolation bellows.

Inert gas supplied to the box is dried and cleaned by a gas-purification system capable of reducing impurities to a few ppm. The gas-system instrumentation includes an oxygen analyzer, hygrometer, and hydrocarbon analyzer, all of high sensitivity.

²⁸ Robinson, L. E., and August, C., Extensible, Multi-purpose Vacuum Glovebox, Paper IV-10, Reactor Physics Annual Report, ANL-7210 (Dec 1966).

The completed system has been thoroughly checked for leaks and briefly tested in operation. First application of the facility will involve the use of a special sodium extruder, now being designed, to be used in the preparation of sodium-bonded fast-reactor fuel samples for meltdown experiments.

D. TREAT Operations

Two Zircaloy-clad, vibratory compacted UO_2 samples (CEN-228-T and 229-T) were irradiated in room-temperature water in transparent capsules. Extensive melting was not obtained during either test, but the high-speed motion pictures of sample CEN-229-T showed localized hot spots; posttransient examination showed that the cladding had experienced gross melting and reaction in these areas. The camera failed to operate during the test on sample CEN-228-T. Samples of the blanket gas from these capsules have been submitted for mass-spectrometer analysis to determine the extent of metal-water reaction. Several steady-state irradiations were made to determine the influence of water-column size on the energy input to vibratory compacted fuel samples in transparent capsules.

Good-quality neutron radiographs of three EBR-II blanket rods were obtained using TREAT as a source of neutrons. Installation of permanent neutron radiograph facilities in front of an unused camera access hole is being evaluated. This facility would be quite useful for inspection of TREAT experiments before and after transient irradiations, as well as for inspection of EBR-II fuel, blanket, and experiments.

Sodium was loaded into the large TREAT sodium loop and testing of the out-of-pile portion of the loop is in progress.

A sample of the boron carbide-epoxy resin mixture used in the TREAT control rods was heated in a furnace to determine the dissociation temperature of the epoxy resin. Extensive gas evolution was detected at 340°C and the test capsule failed at 480°C . Since temperatures above 340°C could be generated in the TREAT control rods during a maximum accident, accompanied by a loss of the air cooling system, transient operation of the reactor was temporarily suspended for further evaluation of this matter. A solution was achieved by replacing the control rods required to assure reactor shutdown with compacted boron carbide-filled rods without epoxy-resin binder. These rods were previously used in the reactor during the first 18 months of transient operation. They had been replaced with the rods filled with the boron carbide-epoxy resin mixture because the former had a small unpredictable variation in their worth. This had been due to insufficient compaction of the boron carbide, allowing movement of the carbide within the rod. Prior to reinstalling the powder-filled rods, the density of the boron carbide powder was increased from about 1.6 to about 1.8 g/cc. Preliminary

tests indicate that local void regions large enough to cause significant variations of control-rod worth will not occur in the control rods containing boron carbide powder compacted to a density of 1.8 g/cc.

E. Chemical and Associated Energy Problems (Thermal)

1. Metal-Water Reactions

a. Isothermal Studies of the Stainless Steel-Steam Reaction. Experiments to determine the isothermal reaction rate of stainless steel with steam have been performed over the temperature range from 1300 to 1600°C in a high-pressure furnace apparatus. Studies of the reactions between molten metals and steam are needed to provide kinetic rate data for predicting, by means of mathematical models, the chemical behavior of core materials during hypothetical reactor incidents. The reaction of molten stainless steel with steam is of particular interest because of the large number of power reactors which employ stainless steel core components.

The reaction of stainless steel with steam has been studied previously by an induction heating method²⁹ over the temperature range from 1100 to 1400°C. However, samples heated to 1400°C underwent rapid swelling and foaming, and, as the sample disintegrated, all control of temperature was lost because of changes in inductive coupling between the induction coil and the sample. It was concluded that the reaction-rate data for temperatures above 1300°C were questionable and that another method of heating the samples was needed.

The high-pressure furnace, which is designed to operate at a maximum pressure of 1000 psi and at temperatures up to 1700°C, allows heating of a sample by a method unaffected by sample disintegration. The furnace (see Progress Report for November 1964, ANL-6977, p. 82) consists of two zones: an internal steam-filled reaction zone that is surrounded by an alumina tube, and an external argon-filled heating zone. The pressures in the two zones are automatically matched to avoid stresses on the alumina tube. During sample exposure to steam in the reaction zone, the extent and rate of metal-steam reaction are determined by continuously monitoring the amount of hydrogen evolved.

Isothermal experiments at 1400°C and stoichiometric experiments at 1500°C have been reported previously (see Progress Report for November 1965, ANL-7122, p. 86). The effect of pressure on the stainless steel-steam reaction was examined in those studies. Experiments performed at 1, 2, and 12 atm indicated that reaction rate was independent of steam pressure. Stoichiometric experiments at 1500°C in flowing steam gave an average yield of hydrogen of 0.51 ± 0.01 liter (STP)/g, which is somewhat higher than calculated equilibrium values. Additional isothermal experiments have now been performed at 1300, 1500, and 1600°C.

²⁹Chemical Engineering Division Semiannual Report, January-June 1964, ANL-6900, p. 239.

The samples were Type 304 stainless steel (16.8 w/o Cr, 8.3 w/o Ni, 1.7 w/o Mn, 73.2 w/o Fe) discs, 3/4 in. in diameter and of variable thickness. In the experiments, the samples were contained in alumina crucibles. Steam flowrate through the furnace varied from a minimum of 5 g steam/min (sufficient steam to oxidize a 13-g sample to completion in one min) to a maximum of 12 g steam/min.

Two experiments at 1300°C and one atm pressure confirmed a previous conclusion that the reaction rate decreased as the effective heat-up time of the sample was increased. The initial reaction rate, which was approximately linear, decreased from about 3.1 to 1.1 ml H₂ (STP)/(cm²)(min) as the heatup time increased from 3 to 9 min.

Two experiments were performed at 1500°C and one atm pressure. One sample of 5.3 g (3/4-in.-dia disc, 0.09 in. thick) was completely reacted in 40 min; the other sample of 20 g (3/4-in.-dia disc, 0.35 in. thick) was about 85% reacted after 90 min.

Two experiments were performed at 1600°C and one atm pressure. Both samples weighed 14 g (3/4-in.-dia disc, 0.24 in. thick). Although the two experiments were nominally identical, the observed reaction rates differed markedly. The complexity of the swelling and foaming phenomena is probably responsible for the lack of reproducibility.

Examination of the oxides from the experiments performed at 1500 and 1600°C indicates a similarity in grain structure between samples that reacted to completion. Although there was a 100°C difference in their reaction temperatures, both had a smooth, foamy texture. The oxides from the 1500 and 1600°C samples that did not react to completion were more nodular in appearance. This indicates that the reaction rate is probably associated with the texture of the oxide at temperatures above the melting point of stainless steel.

b. Isothermal Studies of the Nickel-Steam Reaction. Nickel has been proposed as a cladding material in some water-cooled test reactors. Consequently, the reaction between nickel and steam at temperatures around the melting point of nickel was investigated in order to determine the magnitude of the metal-water reaction. Thermodynamic calculations have shown that, at low hydrogen concentrations (below ~8%) in the steam, the reaction between nickel and steam will proceed at least up to the melting point of the oxide (2000°C).

Several investigators have studied nickel oxidation at temperatures below its melting point (1455°C). Farber³⁰ states that the extent of the reaction below the melting point of nickel is not appreciable. Kubaschewski and Hopkins³¹ report that the oxidation of nickel in steam over a temperature

³⁰ Farber, M., J. Electrochem. Soc. 106, 751 (1959).

³¹ Kubaschewski, O., and Hopkins, B. E., Oxidation of Metals and Alloys, 2nd Edition, Academic Press, Inc., New York (1962).

range from 650 to 1050°C follows a parabolic rate law. Recent experiments by Berry and Paidassi,³² investigating the kinetics of the nickel-oxygen reaction at 600 to 1400°C, also show that the reaction follows a parabolic rate law.

The experiments reported herein were carried out in the high-pressure furnace described in Sect. V.E.1.a. The steam flow past the sample was held at 5 g/min for all of the experiments. The samples were cut from Grade A nickel rod in the form of right cylinders (of 3/4-in. dia and 3/4 in. long). They were contained in impervious alumina crucibles during the experiments.

The area of the sample exposed to steam during the experiment was assumed to be the original sample area for experiments at 1400°C, where the nickel is solid. For experiments at 1500 and 1600°C, where nickel is molten, the area was assumed to be that of the inside diameter of the crucible.

The reaction rate at 1400°C was found to be very low. An accurate assessment of the kinetics of the reaction at 1400°C could not be made because of the "background" gas evolution from the furnace, which is due to the thermal decomposition of steam into hydrogen and oxygen. However, the average reaction rate for a 100-min experiment was 0.11 ml H₂ (STP)/(cm²)(min) for the nickel-steam reaction.

The results of experiments at 1500 and 1600°C (see Fig. 19) indicate that molten nickel reacts in accordance with an approximately parabolic rate law.

The low points, apparent at short reaction times, were probably due to the relatively long time (10 min) required for the samples to reach the reaction temperature in the furnace.

Fig. 19. Hydrogen Evolution as a Function of Time for the Reaction between Nickel and Steam at Various Temperatures and Pressures

Experiments at 1500°C were performed with steam pressures at 1, 8, and 16 atm, and the results in Fig. 19 indicate that there is no significant effect of pressure on the reaction rate.

Figure 20 shows the cross section of a sample exposed to 1600°C steam. Large voids were observed. All surfaces of the internal voids were shiny, indicating that they had not been exposed to steam and that the voids probably formed during freezing of the sample at the end of the experiment. X-ray analysis of scrapings from the sides and

³²Berry, L., and Paidassi, J., *Compt. rend., Ser. C*, **262**, 1353 (1966).

tops of the samples after exposure to steam indicated NiO on all surfaces exposed to steam at 1400°C and on the top surface of samples exposed to steam at 1500 and 1600°C. Only nickel and a small trace of NiAlO_4 were found on the sides and bottoms of the molten samples (1500 and 1600°C), indicating that these surfaces had been effectively shielded from the steam by the crucible.

Fig. 20. Cross Section of Nickel Sample after Exposure to Steam at 1600°C for 90 min (Exposed surface at top)

During the course of these experiments, it was noted that NiO appears to be somewhat volatile in a steam atmosphere. Large (3-mm) crystals of NiO were formed on the alumina reaction tube in areas removed from the sample-containing crucible.

F. Plutonium Volatility Safety

1. Environmental Contamination Control for Volatility Processing

a. Cleanup of Cell Exhaust Contaminated with Plutonium Hexafluoride. As part of the study of the containment of plutonium hexafluoride within ventilated enclosures, a computer program is being developed in order to obtain numerical solutions of differential equations describing the mechanisms occurring in filtration. These solutions will be used to study the effects of filter-system variables on the filtration of aerosols using high-efficiency (AEC) filters. The first parametric study involved a

calculation of the fiber efficiency (related directly to the filter penetration) as a function of the following independent variables: fiber properties-- diameter, spacing, charge, dielectric constant; gas properties--viscosity, density, mean free path, velocity, dielectric constant; particle properties-- diameter, density, temperature, charge, dielectric constant.

The calculation involves the following procedure. The Navier-Stokes equations for gas flow are solved at a finite number of points in a two-dimensional periodic array of cylindrical fibers. The boundary value of the gas velocity at the surface of the fiber is obtained from Cunningham's equation for slip flow at the surface. The two components of velocity at any point in the flow field are obtained by linear interpolation from the nearest point to the surrounding points. The equation of particle motion (based on Newton's law of inertial force, Stokes' law for drag force, Cunningham's correction for Stokes' law, and Coulomb's law for electrostatic forces) are integrated to obtain a conformal array of particle paths. Using the conformal array of particle paths along with the particle velocity at each point on the array, solutions to the diffusion equations (including convection) are obtained. From these solutions, values of the collection efficiency of particles by the fiber are calculated.

Fiber efficiency was computed for average conditions present in a typical high efficiency filter. The filter characteristics³³ and the values used in the present computation were: fiber diameter (D_F), 0.3 micron (geometric mean); fiber spacing (S_F), 1.8 microns; fiber surface area per unit area of filter media, 77. The fiber surface area was used only for converting fiber efficiency to filter penetration.

Typical results of fiber efficiency and filter penetration are plotted in Fig. 21 at two levels of gas velocity (V_G), 2.5 cm/sec

Fig. 21. Results of Fiber-efficiency Computation

³³ Kessie, R. W., and Ramaswami, D., Removal of Plutonium Hexafluoride from Cell Exhaust Air by Hydrolysis and Filtration, ANL-7066, pp. 11-12 (Dec 1965).

(100% of rated flow) and 0.25 cm/sec (10% of rated flow). The two fiber geometries: $F_Y = 0$ for a rectangular array and $F_Y = 1/2$ for a triangular array, gave very different efficiencies, particularly at the higher gas velocity. It is very significant that the particle diameter for minimum efficiency or maximum penetration in all cases was greater than 0.2 micron. The experimentally measured di(2-ethylhexyl)phthalate (DOP) penetrations of 1×10^{-5} to 5×10^{-4} for 0.3-micron-dia DOP particles at V_G of 2.5 cm/sec are between the two curves for $F_Y = 0$ and $F_Y = 1/2$. The calculated results are essentially the same for particles with densities of 1 and 6.5 g/ml, which represent the densities of DOP and plutonyl fluoride, respectively. These results are preliminary in that they do not include the effects of fiber-size or fiber-spacing distributions.

This technique has been developed to the point at which satisfactory solutions can be obtained for the majority of conditions present in high-efficiency filters. However, satisfactory solutions have not been achieved under the following conditions:

1. gas mean free path $\gg$ fiber diameter;
2. Reynolds number > 100 ;
3. strong electrostatic repulsive or attractive forces;
4. sieving, i.e., when the particle diameter approaches the fiber spacing (see broken-line portion of the curves in Fig. 21).

An area of vital interest to the experimental program (see Progress Report for August 1966, ANL-7249, p. 88) is to understand and explain the high penetration of plutonium particles through the second and third filters in a series of high-efficiency filters. This behavior may be caused by electrostatic effects. In an attempt to explain the high penetrations in terms of electrostatic forces, a series of computations were made. Preliminary results of these computations indicated that the filtration behavior of small particles, less than 0.1 micron in diameter, is markedly affected by a single electronic charge. It is planned to improve the computer program to describe accurately the electrostatic effects on fiber efficiency and filter penetration.

VI. PUBLICATIONS

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