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MASTER

Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

December 1966

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

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ANL-7286
Reactor Technology
(TID-4500)
AEC Research and
Development Report

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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

December 1966

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

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Issued January 26, 1967

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:

June 1966	ANL-7230
July 1966	ANL-7245
August 1966	ANL-7249
September 1966	ANL-7255
October 1966	ANL-7267
November 1966	ANL-7279

REACTOR DEVELOPMENT PROGRAM
Highlights of Project Activities for December 1966

EBWR Plutonium Recycle Program

EBWR is operating at 40 MW. At the present level of operation, the average exposure in the plutonium zone will reach approximately 900 MWd/tonne by the end of the present fiscal year.

EBR-II

Operation of the reactor has been very satisfactory with Run No. 23 (690 MWd) completed on December 3 and Run No. 24 (630 MWd) completed on December 31, for a total integrated reactor time of 12,245 MWd. The rotating plug seal troughs continued to operate very smoothly; only $3\frac{3}{4}$ hr were required between Runs No. 23 and 24 to bring the seal alloy to the molten condition. The rotating oscillator was operated this month for the first time to obtain reactor kinetics data. For Run No. 25, scheduled for January, it is planned to increase the core size of the reactor from 81 to 91 subassemblies and, likewise, to increase the number of experimental capsules in the reactor from 235 to 292.

Thirteen fuel subassemblies were fabricated this month, which is in excess of the normal capabilities of the Fuel Cycle Facility.

Plans are being formulated and implemented to expand the output of FCF by augmenting the operating and analytical staff, procuring additional equipment, and by minor modifications to the facility, with the ultimate objective of attaining a higher reactor plant factor. Some of the details of this imminent program are discussed in the current report.

ZPPR

Construction of the facility is on schedule. All foundation posts for the reactor cell are in place, and pouring of concrete has been completed on the service wing foundation and the floor slab, as well as the east and interior walls of the vault workroom area. Seventy-five percent of the north and south walls of this building have been completed. Excavation for the AFSR foundation has begun.

ZPR-3

Results of heterogeneity experiments with Assembly 48, a large, dilute, plutonium-fueled core, are given. A series of experiments with

Assembly 48A was initiated this month for PNL, and is scheduled for 8 weeks in support of the FFTF design group. The experiments consist largely of measurements of control-rod worths, centrally and in the reflector.

AARR

Burns and Roe have completed the final draft of specifications for Construction Package No. 1, the primary pumps, and the primary heat exchanger.

The materials compatibility and corrosion studies being performed by ORNL have been completed and a final report issued. All work in this area has been terminated.

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I. PLUTONIUM UTILIZATION

A. Research and Development

1. EBWR Plutonium Recycle Program

EBWR is operating at 40 MW. At the present level of operation, the average exposure in the plutonium zone will reach approximately 900 MWd/tonne by the end of the present fiscal year.

A proposal to increase the power level to 70 MW has been submitted to the Chicago Operations Office and is being reviewed.

II. LIQUID-METAL FAST BREEDER REACTORS

A. EBR-II

1. Operations

Run No. 23 completed its scheduled 690 MWd of operation on Dec 3. After necessary fuel handling and scheduled minor maintenance, Run No. 24, planned for 630 MWd of operation, was begun on Dec 9. Because of excessive friction in the rotary oscillator, requiring temporary discontinuation of its use, as described below, this run was interrupted on Dec 24 to await approval to install a stainless steel control rod for the purpose of monitoring reactor kinetics by rod-drop techniques. The run was resumed Dec 29 following the receipt of such approval, and completed on Dec 31 for a total integrated reactor time of 12,245 MWd. This delay is not expected to affect the startup of Run No. 25.

Use of the new rotary oscillator in the reactor during power operation was begun at midmonth. A series of kinetics measurements were made at 30 MW, 100% flow, at frequencies from 0.007 to 8 cps. Operation appeared to be satisfactory. Oscillations were next made at 30 MW and 100% flow, 500 kW and 100% flow, and 20 MW and 52% flow. As the outlet temperature of the reactor coolant increased, it was noted that some binding occurred in the oscillator system at rotational frequencies below 0.3 cps. Further testing indicated a definite binding, apparently between the oscillator rod and thimble, occurring at average coolant temperature rise of greater than 90°F and for rotational speeds less than 0.3 cps. Finally, at 45 MW and 100% flow, measurements indicated that the friction was excessive, and the decision was made not to operate the oscillator under these conditions. At this point approval was requested and later received to insert the stainless steel control rod in place of a normal one to obtain kinetics data by the rod-drop technique. These data are important as a reference base in view of the proposed core change from 81 to 91 subassemblies for the next power run.

Upon resumption of Run No. 24, rod-drop data were obtained at 500 kW, 30 MW, and 45 MW.

Plans are being made for the removal and inspection of the oscillator rod and thimble when replacements are available.

Nine outer blanket subassemblies were raised slightly and re-seated to test for any indication of adhesion or binding. This is a continuation of the surveillance previously carried out in Dec 1965 and June 1966. At least one subassembly in each row and sector are raised during the testing period. No adverse results were observed.

Core changes for Run No. 24 involved the exchange of 19 subassemblies. Experimental subassembly XA07 was removed at the request of the experimenter, and XO18 containing structural and materials samples was inserted. Two outer-blanket alpha-measurement subassemblies were removed after long residence in Rows 13 and 15, respectively.

Several maintenance tasks, originally scheduled following Run No. 24, were performed during the reactor down time during the interruption of the run. These included the following: (1) the main turbine emergency governor (overspeed) was removed and sent to the manufacturer for inspection and testing; (2) the turbine-driven condensate pump was disassembled for annual inspection; (3) inspection and modifications were begun on the fuel-unloading-machine gripper-sense-indication system.

During the required balance of Run No. 24 the turbine-generator was not operated and steam was bypassed directly to the condenser.

a. Rotating Plugs. On Dec 3, preparatory to the fuel-handling operations following Run No. 23, the large plug was rotated without difficulty after being in the molten mode of operation for about $3\frac{3}{4}$ hr. The small plug was rotated after about 4 hr. Subsequent performance during fuel handling was normal in all respects. The seals were returned to the half-molten mode of operation on Dec 7, preparatory to Run No. 24.

To check alloy composition, samples for analysis were taken from the seal troughs on Dec 6. The results are shown in Table I. The original average composition of the alloy when first placed in the seal was about 58 w/o Bi and 42 w/o Sn, with a melting point of 281°F. The indicated present deviation from this analysis is not considered significant.

TABLE I. Chemical Analyses of Seal-trough Materials

Sample Description	Constituent		
	Bi (w/o)	Na (ppm)	Fe (ppm)
Alloy from outer annulus of large plug trough, taken near alloy surface (7 in. above bottom of trough).	59.2	2400	22
Alloy from outer annulus of large plug trough, taken 2 in. above bottom.	59.0	2500	25
Alloy from outer annulus of small plug trough, taken near alloy surface (7 in. above bottom of trough).	58.1	860	20
Alloy from outer annulus of small plug trough, taken 2 in. above bottom.	56.5	820	17

b. Experimental Irradiations. The status of all experimental irradiations in the reactor and the approximate exposure following completion of Run No. 24 is as follows:

Subassembly	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	10,300
XG06	9/3/65	UO ₂ -20 w/o PuO ₂ (12) U-15 w/o Pu-10 w/o Zr (1) U-15 w/o Pu-10 w/o Ti (1) Structural (5)	GE ANL ANL ANL	9,317	20,600
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 v/o PuO ₂ (1) 304 SS-20 v/o PuO ₂ (1) 304 SS-20 v/o UO ₂ (1)	ANL GE PNWL PNWL PNWL	5,094	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-1A (Metal) (2)	NuMEC GE ANL ANL	1,320	11,000
XO17	11/15/66	PuO ₂ -UO ₂ (11) (Pu,U)C (3) MK-1A (Metal) (5)	NuMEC UNC ANL	1,320	6,500
XO18	12/6/66	Structural (3) Structural (1) Structural (2) Structural and Heavy Metal Fission Yield Samples (1)	GE PNWL ANL ANL	630	21,300

(a) Maximum attainable before removal is required for experiment of higher priority.

The following changes to the loading of experimental subassemblies were made during the shutdown following Run No. 23.

(i) Subassembly XA07 was removed at the experimenter's request. The subassembly contents and exposures are:

16 capsules, U-15 w/o Pu-11 w/o Zr	~4 a/o burnup
3 capsules, structural	~3 x 10 ²² nvt
	(integrated total flux)

(ii) Subassemblies U-1550X and U-1511X containing special alpha-measurement foils were removed after being exposed for 9,985 MWd of reactor operation in positions 13B7 and 15B8, respectively.

(iii) Subassembly XO18 containing structural samples and about 1.4 g of heavy metal oxides, as shown in the status table above, was loaded into grid position 2B1.

Ten structural samples of the Mk-B-19 design were received from GE and are scheduled to be combined with nine similar types from ANL to form the complement for the Row-4 subassembly XO16.

Disassembly of experimental subassemblies XA07 and XO09 was completed in the Air Cell of the Fuel Cycle Facility. Fourteen of the 16 metal fuel capsules from XA07 and five ceramic capsules from XO09 were transferred to the Laboratory's Metallurgy Division.

2. Transfer-function Measurements

An on-line data-reduction system was used to produce the transfer-function data shown in Fig. 1. In the method basic Fourier analysis of data received from the oscillator mechanism and the ion-chamber signal is made. The oscillator drive produces the sine and cosine information and this, along with the ion-chamber current, is sent directly to an analog-to-digital converter in conjunction with an IBM 1620 computer. The computer then uses the digitalized data to perform the numerical integration at a rate of 60 samples per second. Processing of the data produces the Fourier components after a predetermined number of integral cycles of the oscillator.

The results tabulated by the computer are:

- a. frequency of oscillation;
- b. number of sample points used for the calculation;
- c. real component of the Fourier coefficient;
- d. imaginary component of the Fourier coefficient;
- e. the absolute value of the Fourier coefficient;
- f. phase angle.

Fig. 1. EBR-II Transfer Function

The calculated phase angle represents not only the phase characteristic of the reactor but also the phase displacement of the sine, cosine function generator with respect to the actual position of the reactivity generator. This correction, except for phase-error uncertainty due to backlash in the drive, is determined by reverse oscillation or by normalization at the highest frequency.

The zero-power results are in excellent agreement with the calculated zero-power transfer function with respect to both gain and phase. The calculated curve is based on the β fraction used for the original transfer function ($\beta_{\text{eff}} = 0.00736$).

The transfer function data for 30-MW operation are being analyzed.

3. Reactor Physics

To predict the effects of changes in reactor loading on the distribution of fission rate in the reactor, it is necessary to provide computing techniques sophisticated enough to give an accurate, but simple enough, picture for routine use. To this end, considerable effort has been expended in characterizing the reactor using one- and two-dimension diffusion theory and one-dimension transport theory. The results of these calculations can be compared at present with only two sets of experimental data: (a) the flux measurements for the "wet critical" core and the fuel-burnup data obtained during the Mark-I-fuel surveillance program when the core size was about 70 subassemblies; and (b) experimental data obtained with flux wires in Run No. 20 (80-subassembly core). Some data are available from burnup determinations for Mark-IA fuel, but because of the changing core size and changing enrichment, these data are difficult to evaluate except on an average basis.

The results of calculation and experiment have appeared in previous monthly reports. The conclusion that must be drawn on the basis of currently available information is that the present codes are not sufficiently definitive in the regions of perturbations caused by experiments in the reactor and at or near the boundary of the core and reflector. Thus, it will be necessary to measure fission rates experimentally throughout the core using special flux-wire subassemblies. The first measurements are planned for Run No. 25, when the core will comprise 91 subassemblies, with experiments in Rows 2, 4, and 6. The data will be used for calculating a new fission-rate distribution to be compared with results of a two-dimension transport code being prepared for EBR-II.

4. Plant Modifications

One of the current reactor improvements being accomplished includes the installation of a new cold trap and complete revision of the in-cell piping in the purification system for the primary sodium. The

reason for this modification is that the current performance of the existing cold trap indicates it is nearing the end of its useful life and must soon be replaced. Most of the present piping must be removed to facilitate removal of the cold trap from the cell through the upper access opening provided for this purpose. Along with the new cold trap, a new piping system has been designed which will allow semiremote removal of the cold trap from above in the future, without disrupting the piping system. Special couplings, which may be disconnected from above with long-handled tools connect all pipe lines to the cold trap. Simple devices to detect sodium leaks are provided beneath all couplings.

Two identical cold traps, one a spare, are ready for shipment from the manufacturer. An ANL engineer visited the manufacturer's plant this month to witness a pneumatic pressure and leak test on the sodium shells and economizer units for both cold traps.

5. Surveillance

a. Examination of Irradiated EBR-II Fuel Jacketing. In September, two Type 304L stainless steel fuel jackets irradiated at 371 (at bottom) to 517°C (at top) in EBR-II to a fluence of 5×10^{21} to 1.2×10^{22} nvt, fast neutrons, were each cut into five nearly equal sections for tube-burst testing. The sections from one jacket were annealed at 900°C for one hour, to recover some of the ductility lost through atomic displacements, and were air-cooled before being tested.

Tube-burst tests at 500°C then showed the trend for the strengths of the unannealed jacket to be progressively greater from top to bottom, and that irradiation of the annealed jacket had changed the average tangential rupture strength and elongation from 48,000 psi and 9.7% to about 79,000 psi and about 0.4%, respectively. Postirradiation annealing resulted in average values of 53,000 psi and 13.2%.

Since September, similar tube-burst tests at 600, 700, 750, 800, and 900°C indicate that a nonrecoverable loss of ductility, attributed to the presence of helium from (n,α) reactions, persists at temperatures above 700°C. The comparative data are presented in Table II. Between 700 and 750°C the extent to which ductility is recovered on annealing changes definitely. In tests at 700°C and lower, the 900°C anneal has resulted in complete recovery of ductility.

The rupture strengths of specimens annealed after irradiation tend to remain slightly greater at 600 and 700°C than the rupture strengths of unirradiated controls at these temperatures, but to be less at 800 and 900°C than in the case of the controls.

TABLE II. Effect of Postirradiation Annealing on Strength and Ductility of EBR-II Fuel Cladding (Type 304L)

Test Temperature (°C)	Unirradiated		Irradiated		Irradiated and Annealed	
	Rupture Strength (psi)	% $\Delta D/D$	Rupture Strength (psi)	% $\Delta D/D$	Rupture Strength (psi)	% $\Delta D/D$
600	42,500	13.8	59,100	0.3	45,000	13.7
			63,400	0.6	44,800	12.1
			65,700	0.2	44,600	12.7
			67,100	1.2		
			64,300	1.7		
700	33,800	11.1	35,200	8.8	35,800	12.9
			40,700	1.3	37,200	12.3
			42,800	3.6		
			41,600	5.0		
			37,000	1.5		
750					25,700	2.0
					25,700	1.8
800	27,600	13.5	23,000	3.9	18,400	1.0
	29,400	16.7	21,800	1.7	20,000	1.7
			23,400	2.2		
			24,100	3.3		
900	19,300	15.8	14,900	4.6	17,200	5.7
	19,300	19.5	14,700	2.2	15,900	3.1
			13,800	2.9	14,500	0.9
			14,500	3.2	15,200	3.2
			14,500	3.6	15,600	3.3

Helium generation in Type 304 stainless steel is not apt to be a problem under present operating conditions in EBR-II, for the effects of the helium become evident only above 700°C. The ductility losses due to atomic displacements are important. The capability of lower-temperature anneals, say at 500°C, for removal of any appreciable number of atomic displacements will therefore be determined.

6. Fuel Cycle Facility (FCF)

a. Program to Increase Reactor Fuel Production. In order to meet increasing requirements for reactor fuel, needed to attain a higher reactor plant factor, plans are being prosecuted to expand significantly the FCF

operating and analytical staff and to provide corresponding auxiliary personnel, to procure certain additional equipment, and to effect minor modifications in the facility which will facilitate operations. In addition, plans are being developed for the commercial procurement of fuel for the reactor, for its processing off-site, and for the interim fabrication of unirradiated fuel elements at the EBR-II site other than in-cell. Implementation of these latter programs must await the approval of the AEC.

(i) Equipment Procurement. To speed operations and minimize downtime for repair in the FCF and its analytical facilities, procurement of the following typical equipment is planned or under way: additional master-slave and electromechanical manipulators, a skull-oxidation furnace, fuel-element-bonding furnaces, fuel-storage and -transfer units, and other minor equipment. The utility of an emission spectrograph, capable of handling irradiated material for rapid analyses of minor fuel-alloy impurities, is also being studied with a view to such an installation.

(ii) Facility Modifications. Modifications of FCF and its auxiliary facilities which are planned to permit increased in-cell sub-assembly production, and to increase the reliability of facility operation, include the following:

1. a manipulator decontamination and repair area (to be provided on the cell roof);
2. a process equipment decontamination and repair area in the FCF basement, to function in connection with the Transfer Cell Access Door, now being installed;
3. increased storage capacity for radioactively hot subassemblies in transit to or from the reactor;
4. an improved sample-transfer system (pneumatic) to connect the FCF with the analytical hot cells.

(iii) Commercial Procurement of EBR-II Driver Fuel. A specification suitable for the commercial procurement of EBR-II driver fuel elements has been prepared and submitted to the AEC for review along with a procurement plan. Prospective vendors have been visited to ascertain their capabilities and interest, and information on fabrication techniques has been furnished to potential vendors.

Since commercial procurement will increase the FCF and reactor inventory beyond that which can be stored or processed, discussions have been held with the staff of the Idaho Chemical Processing Plant concerning the capability of this facility for storage and reprocessing of EBR-II fuel. To accomplish such storage or reprocessing, transfer

casks will be required. Off-site reprocessing will increase the availability of the FCF for the experimental irradiation program and ultimately permit its utilization for the development and demonstration of processes for other fuels.

(iv) Interim Fabrication of Unirradiated Fuel. As an interim measure, pending the availability of driver fuel from commercial sources, consideration is being given to providing facilities at the EBR-II site for casting, assembly, welding, and testing of fuel elements prepared from unirradiated alloy, to effect the most rapid production increase. In order to facilitate this, existing space in the FCF and in the ITF (Inspection and Test Facility) would be used. The ITF is also being proposed for assembly of commercially procured fuel elements into reactor subassemblies as well as its present use in the EBR-II experimental fuels irradiation program.

b. Hot Fuel Examination Facility. Design studies are under way for a Hot Fuels Examination Facility (HFEF) which it is proposed to attach to the FCF to provide for the safe and expeditious handling of EBR-II irradiation experiments, and principally for nondestructive testing and reinsertion of fuels in the reactor. The main cell will be located as an extension at the northwest corner of the FCF and will provide a radiation-shielded, inert-atmosphere space for subassembly transfer and equipment repair, as well as for the disassembly and reassembly, dimensioning, radiography and eddy-current examination, subassembly testing, sample preparation, and physical testing of experimental fuel and material specimens which have been irradiated in EBR-II. An auxiliary decontamination cell will be provided, along with a hot shop and a shielded mechanical and storage area.

c. FCF Operations

(i) Summary of Production. A summary of production operations is contained in Table III.

(ii) Effect of Sodium-removal Operation. The reinsertion of experimental irradiation assemblies into the reactor will normally involve reinserting stainless steel capsules from which adhering sodium has been removed by treatment with humid air and with water. To assure that no significant deleterious effects on the stainless steel will occur as a result of the sodium-removal operation, a test is being carried out in which fuel elements, from which sodium has been removed by the normal procedure (except for double washing), were irradiated and then removed for detailed examination of the cladding. These elements were contained in subassembly C228, removed from the reactor at the end of Run No. 22. Close examination of six elements using a periscope, revealed sound welds and no abnormal surface conditions. Staining and residue on the lower ends of the elements were normal. Specimens of the cladding were obtained for metallographic examination.

TABLE III. Production Summary for December 1966

Subassemblies received:	23	
Subassemblies dismantled:	16	
Subassemblies fabricated:	13(a)	
Subassemblies transferred to reactor:	4	
Elements decanned:	1623	
Melt refining:	9 irradiated	4 recycle
Pour Yield (av):	92.8	93.0
Injection casting runs:	13	
Elements processed:		
Accepted:	1232	
Rejected:	102	
Elements welded:		
Accepted:	868	
Rejected:	28	
Elements bonded and tested (completed runs):		
Accepted:	1090	
Rejected:	56	
Subassemblies subjected to surveillance:	4	

(a) Plus 6 additional subassemblies containing flux wires, fabricated from unirradiated fuel for reactor flux measurements

B. Physics Development

1. ZPR-3

a. Assembly 48. The results of the heterogeneity experiments conducted on Assembly 48 last month are given here. Assembly 48 is a 400-liter, plutonium-fueled core of simple composition, with a degraded neutron spectrum similar to those in some anticipated large power-reactor cores.

Both fuel-bunching experiments and sodium-void experiments were run. Figures 2 and 3 show the locations of the drawers used in the experiments. The central core measurements were made in the front 4 in. of the drawers marked A and B. Measurements at the axial core edge were made in the outer 4 in. of core material in the drawers marked A and B. Measurements at the radial core edge were made in the front 4 in. of the drawers marked C and D. Figure 4 is an end view of the arrangement of material in the 4-in. section of the core drawers used in the experiments.

The results of the heterogeneity experiments are listed in Table IV. The effect of bunching and unbunching the fuel columns was measured at the core center. The effect of fuel bunching was also measured at the axial core edge.

Fig. 2. Drawers Used in Heterogeneity Experiments in Half No. 1 of Assembly 48 of ZPR-3

Fig. 3. Drawers Used in Heterogeneity Experiments in Half No. 2 of Assembly 48 of ZPR-3

Fig. 4
Material Arrangements in
Core Drawers for Heterogeneity
Experiments with ZPR-3
Assembly 48

TABLE IV. Results of Heterogeneity Experiments in Assembly 48

No. of Drawers	Experiment	Core Location	Drawer Designation	Change in Reactivity (1h)	No. of Drawers	Fuel Configuration	Core Location	Drawer Designation	Sodium Removed (kg)	Reactivity Change (1h/kg)
Fuel Experiments ^(a)					Sodium-void Measurements ^(a)					
9	Bunching	Center	A (Half #1)	16.1 ± 1.0	18	Normal	Center	A	1.32	7.9 ± 0.8
9	Bunching	Center	A (Half #2)	17.1 ± 1.0	24	Normal	Center	B	1.76	3.2 ± 0.6
24	Bunching	Center	B	41.7 ± 1.0	42	Normal	Center	A and B	3.08	5.2 ± 0.3
42	Bunching	Center	A and B	75.2 ± 1.0	42	Bunched	Center	A and B	3.08	3.1 ± 0.3
42	Bunching	Axial Edge	A and B	16.8 ± 1.0	42	Unbunched	Center	A and B	3.08	5.7 ± 0.3
18	Unbunching	Center	A	-8.3 ± 1.0	42	Standard	Center	A and B	3.08	5.6 ± 0.3
24	Unbunching	Center	B	-11.5 ± 1.0	42	Normal	Axial Edge	A and B	3.08	-20.7 ± 0.3
42	Unbunching	Center	A and B	-20.0 ± 1.0	42	Bunched	Axial Edge	A and B	3.08	-23.6 ± 0.3
					42	Standard	Axial Edge	A and B	3.08	-20.5 ± 0.3
					42	Bunched (15 in.)	Axial Edge	A and B	3.08	-23.5 ± 0.3
					12	Standard	Radial Edge	C	0.88	-17.6 ± 1.1
					10	Standard	Radial Edge	D	0.73	-19.6 ± 1.4
					22	Standard	Radial Edge	C and D	1.61	-18.5 ± 0.6

^(a) All measurements made in both halves of reactor, except where stated otherwise.

The change in reactivity due to the removal of sodium was measured at the center, axial edge, and radial edge of the core. This was done in the various fuel-loading configurations shown in Fig. 4 by removing the cans of sodium and replacing them with an amount of stainless steel equal to that in the sodium cans. The "standard" fuel configuration listed in Table IV is the same as the "normal" fuel loading except that the 1/4-in. Pu-U alloy fuel column had not been replaced with the two 1/8-in. columns used in the fuel bunching and unbunching experiments. Sodium-void measurements were made in both the "standard" and "normal" fuel configurations because the substitution of two 1/8-in. columns for one 1/4-in. column of fuel reduced the total Pu^{239} and Pu^{241} loading by ~0.7 kg in the 42-drawer central and axial core-edge experimental region. During the sodium-void measurements at the axial core edge, a change in the worth of sodium between the "normal" and "bunched" fuel configurations was noted. To determine if this change in reactivity was due to neutron streaming from the 11-in. portion of the fuel columns not bunched during the bunching measurements, the experiment was repeated in a "standard" fuel loading and a loading in which the entire 15-in. columns of fuel were bunched. The results indicate that the change in worth of the sodium in this region is due to fuel bunching and not to neutron streaming.

b. Assembly 48A. On December 1, a series of experiments was begun for Pacific Northwest Laboratories (PNL) in connection with the FFTF project. Argonne National Laboratory, through the AEC, is providing the critical assembly experiments in support of the reactor being designed by the FFTF group. This initial set of experiments, mutually agreed to by ANL and PNL, is to run for eight weeks, and will provide data necessary for the development of calculational techniques to be used in the design of the FFTF reactor, particularly in the area of control-rod designs. The core in Assembly 48 is being used since the neutron spectrum is fairly representative of that expected in the FFTF reactor.

These experiments consist primarily of measurements of control-rod worths, both centrally and in the reflector. The FFTF design will utilize a reflector of medium atomic weight (largely nickel), and a quadrant of such a reflector will replace the uranium reflector on Assembly 48. However, initial experiments will be performed with the full uranium reflector to provide comparative data with reflectors of both heavy and medium atomic weights. With both reflectors the emphasis will be on control-rod worths (poison rod experiments), reactivity-traverse measurements with small samples, and reaction-rate traverses.

The poison rod measurements and the axial reactivity coefficient sample traverses have been made with the depleted uranium blanket still in place. The loading is now under way to replace two segments of the depleted uranium blanket with an Inconel reflector of the type that might be used on the FFTF reactor. The remaining experiments will be conducted with the new blanket-reflector configuration.

Figure 5 shows the B_4C arrangement in several poison rod mockups. The worths of these rods, which were 54 in. long, were measured at the axis of the core and blanket by replacing a drawer filled with eight 1/4-in. columns of sodium. The normal rod mockup with enriched B_4C was measured in steps through the blanket and core to its full 54 in. No other poison rod mockups were measured in steps. The effect of bunching the enriched B_4C was measured by replacing the reference loading with the bunched loading. The effect of enrichment was measured by replacing the enriched B_4C with natural B_4C in the normal loading. The results are given in Table V. Subcritical multiplication techniques were used to make the measurements. The error is estimated to be $\pm 2\%$.

Fig. 5

Cross-sectional View of
Drawer Loadings Showing
 B_4C Arrangement in Poison
Rod Mockups for Assem-
bly 48A

TABLE V. Results of Measurements with Central Poison
Rod Mockup in Assembly 48A

Rod Insertion (in.)	Reactivity Change for Rod Mockups (Ih)			
	Normal Enriched B_4C	Normal Natural B_4C	Reference Enriched B_4C	Bunched Enriched B_4C
0 (Blanket Edge)	0	0	0	0
8	-2.9			
12 (Core Blanket)	-26.9			
19	-288			
27 (Core Midplane)	-894			
35	-1487			
42 (Core Blanket)	-1723			
54 (Blanket Edge)	-1742	-747	-1737	-1531

An axial traverse drive mechanism was installed to make traverses with reactivity coefficient samples. The traverses were made through a half-inch guide tube placed in the central core drawers. The materials measured were 2-in.-long cylindrical samples of plutonium, enriched uranium, depleted uranium, boron, and tantalum. Also, stainless steel was measured so that a correction for the sample container could be made. The empty sample carrier was also run as a reference for the material samples. The servo-control rod (autorod) was calibrated and used for measuring the reactivity changes of the samples. The results of the traverse runs are being analyzed.

2. ZPR-6

Reaction-rate traverses of U^{235} and U^{238} were done in the radial and axial directions in ZPR-6 Assembly 5. These results are shown in Fig. 6. The extrapolation to zero fluxes results in an equivalent bare radius of 92 cm. The radial traverses do not fit a zero-order Bessel function.

Experiments were performed with an accelerator-type pulsed neutron source in Assembly 5. Tritium target for the $T(d,n)He$ reaction was placed in the center of the core.

A pulse width of about $1.5 \mu\text{sec}$ was used to study the decay of neutron bursts. U^{235} and U^{238} fission chambers were used as detectors at different radial locations in the midplane of the reactor. At two locations (near the core center and near the core-blanket boundary) the transients immediately following the pulse were studied in detail with $0.15\text{-}\mu\text{sec}$ -width pulses using a U^{235} chamber. All decay measurements were made with the reactor subcritical between 0.1 and 2% $\Delta k/k$.

A proton-recoil counter was placed in the core to measure the neutron spectrum at different time intervals after the pulse, using a pulse width of $1.5 \mu\text{sec}$. The proton-recoil counter was gated on for $10 \mu\text{sec}$ or $3 \mu\text{sec}$ at different times after the pulse.

The neutron intensity emerging from a beam hole which was created by removing four central drawers from the core was measured with a U^{235} fission chamber.

A detailed evaluation of all the pulsed experiments will be made.

3. ZPR-9

The results of the central worth measurements made in ZPR-9 Assembly 12, obtained from period measurements, are given in Table VI. Assembly 12 is a small zoned assembly designed to have a central neutron-energy spectrum similar to those of Assemblies 4Z and 5 of ZPR-6. The constancy of the ratios of worth in Assembly 12 to worth in Assembly 5 is an indication that the spectra are well matched. The deviation in this ratio for the light scattering materials, such as carbon and beryllium, indicates that a perfect match has not been achieved. Because of the general shape of the adjoint function in this type of core, a deviation of less than 1% in this function can cause changes as great as 100% in the worth of a pure scattering sample. The scattering materials are therefore a very sensitive test of the spectral match. In the case of sodium, however, the small signal caused an uncertainty in the ratio of ~30%. It therefore gives only limited information on the spectral match; additional sodium measurements are underway to improve the precision of the measurement of this important quantity.

Fig. 6a. Reaction-rate Traverse of U^{235} in the Radial Direction of ZPR-6 Assembly 5

Fig. 6b. Reaction-rate Traverse of U^{238} in the Radial Direction of ZPR-6 Assembly 5

Fig. 6c. Reaction-rate Traverse of U^{235} in the Axial Direction of ZPR-6 Assembly 5

Fig. 6d. Reaction-rate Traverse of U^{238} in the Axial Direction of ZPR-6 Assembly 5

TABLE VI. Central Worth Measurements in ZPR-9 Assembly 12
(1% $\delta k/k = 451.2$ Ih)

Sample	Sample Weight (g)	Can Weight (g)	Gross Worth (Ih)	Net Worth (Ih)	Net Worth (Ih/kg)	Ratio of Worth in Assembly 12 Worth in Assembly 5
Polyethylene	23.973	Bare	+9.539	+9.539 $\pm$ 0.010	+397.9 $\pm$ 0.5	-
Polyethylene	58.98	Bare	+26.155	+26.155 $\pm$ 0.015	+443.5 $\pm$ 0.3	1.368
Be	114.308	Bare	+1.395	+1.395 $\pm$ 0.010	+12.208 $\pm$ 0.087	1.235
B ¹⁰	29.29	55.73 (SS)	-32.731	-32.589 $\pm$ 0.015	-1112.6 $\pm$ 0.6	1.393
C	102.01	Bare	+0.161	+0.161 $\pm$ 0.011	+1.578 $\pm$ 0.11	0.888
Na	51.38	54.52 (SS)	-0.210	-0.0707 $\pm$ 0.0103	-1.376 $\pm$ 0.20	1.713
Al	166.9	Bare	-0.428	-0.428 $\pm$ 0.010	-2.564 $\pm$ 0.060	1.427
Fe	488.0	Bare	-1.022	-1.022 $\pm$ 0.010	-2.095 $\pm$ 0.020	-
SS	480.98	Bare	-1.137	-1.137 $\pm$ 0.010	-2.365 $\pm$ 0.021	1.389
SS can	72.378	Bare	-0.184	-0.184 $\pm$ 0.010	-2.542 $\pm$ 0.138	-
Ni	546.0	Bare	-1.634	-1.634 $\pm$ 0.010	-2.993 $\pm$ 0.018	1.441
Nb	481.2	30.10 (SS)	-5.406	-5.329 $\pm$ 0.015	-11.074 $\pm$ 0.031	1.389
Mo	599.0	Bare	-4.826	-4.826 $\pm$ 0.010	-8.057 $\pm$ 0.017	1.413
Ta	924.7	27.54 (SS)	-16.698	-16.628 $\pm$ 0.015	-17.982 $\pm$ 0.016	1.386
W	1108.0	Bare	-8.133	-8.133 $\pm$ 0.010	-7.340 $\pm$ 0.010	1.404
U ²³⁵	18.11 (Total) 16.87 (U ²³⁵)	72.378 (SS)	+0.993	+1.183 $\pm$ 0.015	+70.12 $\pm$ 0.88	1.399
U ²³⁵	59.40 (Total) 55.32 (U ²³⁵)	55.44 (SS)	+3.526	+3.691 $\pm$ 0.015	+66.72 $\pm$ 0.27	-
U ²³⁸	1153.8	Bare	-5.398	-5.398 $\pm$ 0.010	-4.679 $\pm$ 0.009	1.398
Pu	148.36 (Total) 113.34 (Pu ²³⁹) 5.34 (Pu ²⁴⁰) 0.54 (Pu ²⁴¹)	22.27 (SS) 6.85 (Al)	+11.019	+11.094 $\pm$ 0.20	+93.05 $\pm$ 1.7 ^(a)	1.336

(a) Value is for plutonium mixture.

4. ZPPR

The overall construction of the ZPPR facility is on schedule. All foundation posts for the reactor cell have been grouted in place. The foundation of the service wing has been poured except for the AFSR wing. The floor slab, the east wall, and the interior separation wall of the vault-workroom area have been poured. Seventy-five percent of the north and south walls has been completed. The counting-room concrete including the roof has been poured. The floor slab of the control room has been formed and poured. The excavation for the foundation of the AFSR has commenced.

The present status of the reactor components is as follows:

a. Items Completed. Completed items that have been inspected, accepted, and presently stored at the Idaho Division are: bed and tables, fuel control and safety rod drives, matrix tubes, louvers for the reactor cooling system, tube plug clamping assemblies, reactor frame, pit bumpers, ramp assembly, platform support bracket, seismic switch, beta-gamma air particulate monitors, area gamma monitors, nuclear instrumentation, and reactor control consoles and cabinets.

b. Matrix Drawers. The vendor, Mechanical Products Manufacturing Company, Seattle, Wash., has supplied the following acceptable matrix drawers: 100 special tab front drawers, 1677 regular front drawers, and 2146 back drawers. Approximately 400 regular front drawers remain to be finished as well as 48 special narrow drawers.

c. Poison Safety Rod Drives. All parts for the first rod drive have been received or fabricated. Assembly of this first rod for check out has begun.

d. Reactor Knees. An ANL representative inspected the knees at the vendor's plant, McCauley Foundry, Berkeley, California. The surfaces and dimensions were satisfactory. Relocation of the plenum mounting holes on one set of knees was required.

e. Personnel Shields. The bids for the reactor personnel shields have been evaluated and have been sent to AEC-CH for approval.

f. Matrix Alignment. The bid packages on the matrix alignment plates have been sent out. The due date on bid packages is January 17.

g. Source Drives. Work is continuing.

h. Rod-drive Mounting Plates. One rod-drive mounting plate was rejected because of excessive unrepairable cracks around the poison rod drive mounting holes and the center experimental hole. The vendor was instructed to recast a new plate. The second plate was also conditionally rejected due to some cracks and voids. This plate could be repaired. The vendor was given the choice of repairing it satisfactorily or recasting the plate. An extension of six weeks for delivery was given the vendor.

i. Drawer Springs. The resubmitted samples were found acceptable. The vendor was instructed to proceed with the fabrication of the drawer springs.

j. Matrix Tie-down. Fabrication of the reactor matrix tie-down is continuing.

k. Loading Platform. An ANL representative inspected the completed loading platform at the vendor's plant. The platform was acceptable and has been painted. It is now being crated for shipment.

l. Reactor Plenum Assemblies. The fabrication of the plenum assemblies is continuing. The samples of Neoprene plugs were reviewed and accepted. The vendor was instructed to begin fabrication of the plugs.

m. Pit Conveyor. The conveyor was sent back to the vendor to correct the location of holes.

n. Fuel Control and Safety Rod Drawers and Ball Joint Assemblies. The control drawers have been Scottsonized. The monoballs for the joints have been received. The units are being assembled.

o. Poison Safety Blades and Sheaths. A blade and sheath have been Scottsonized and are now being inspected for dimensional tolerances. Evaluation of these samples will be made by the vendor prior to processing the remaining units.

p. Boron Fabrication. The bid packages for the fabrication of boron into B_4C are being processed in the Laboratory.

q. Stack Gamma Monitor. A service request for the mound area stack gamma air monitor has been issued.

r. Reactor Control Cable Stand. Purchase items for the Reactor Control Cable Stand have been initiated.

C. Component Development

1. Sodium Technology and Development

a. Sodium Analytical Laboratory. All components of a 1024-channel system for analyzing radioactive materials in sodium have been received except the phototube socket and preamplifier. A preliminary checkout of the equipment by the supplier has been completed.

Difficulties with the purification system for the glovebox atmosphere were found to be due to an inoperative solenoid valve on the discharge side of the oxygen-getter bed, consisting of hot copper turnings. During the regeneration cycle a hydrogen-helium gas mixture sweeps across the heated bed, reduces the copper oxide, and then flows out through the discharge valve. The jamming of the inoperative valve was due to an accumulation of iron oxide and possibly some copper oxide in sufficient quantity to interfere with its operation. The valve has been cleaned and reinstalled. The glovebox and purification system are again in operation. A filter will be installed in the line to prevent the reoccurrence of this problem.

b. Measurement of Sodium Quality. Installation of a new Sodium Analytical Loop is approximately 65% complete. All line heaters and line thermocouples have been installed, and the cold-trap cooling system has been completed. The major work remaining to be done includes insulation, wiring, and vacuum-gas line hookup.

A modified flat-coiled resistance heater for the EBR-II Primary Sodium System sample cup has been tested. Three sodium distillations made with this type of heater gave favorable low-current and high-temperature operation. The fabrication of this improved heater is much simpler, and the design will be incorporated in a new modified sample cup.

2. Materials Evaluation

a. Effects of Sodium on Stainless Steels. Two coupons each of 304 stainless steel and 304L stainless steel were soaked in sodium containing ~20 ppm oxygen for $9\frac{1}{2}$ days at 1200°F. Resistivity measurements made with the original material, after sodium treatment, and after soaking in Strauss reagent for two successive 30-min periods gave values shown in Table VII.

Strauss reagent vigorously attacks chromium carbide precipitates located in the grain boundaries of the steel. These tests indicate that the embrittlement or extent of carbide precipitation in stainless steel resulting from its exposure to sodium at 1200°F is considerably greater for Type 304 stainless steel (with 0.06-0.08% carbon) than for Type 304L (with 0.03% max carbon), the embrittlement being determined by increase in resistivity after treating the specimens in Strauss reagent.

TABLE VII. Electrical Resistivity Values (ohm-cm) of Stainless Steels

Sample No.	Steel Type	Carbon Content (%) (nominal)	ρ Original	ρ after Sodium Treatment	ρ after 30 min in Strauss Solution ^a	ρ after 60 min in Strauss Solution ^a
1	304L	0.03 max	70.7	71.3	71.3	71.3
2	304L	0.03 max	73.4	74.3	75.1	75.6
3	304	0.06 (by analysis)	69.8	70.5	137.2	(Sample Disintegrated)
4	304	0.07 (by analysis)	74.9	74.6	98.4	97.6

^aStrauss Solution: Cupric Sulfate 30 g
 Conc. H₂SO₄ 55 ml
 Distilled Water 870 ml.

b. Wear Testing. In the area of wear testing to be performed in the CAMEL loop, a complete Falex wear test unit was assembled and mounted on a sodium-filled pot. A pair of Stellite 19B vee blocks was mounted in this unit in contact with a Carboloy 98B shaft. The unit was checked out with the data-acquisition system which will be used to collect the data on rpm, motor torque, wear and wear rate, and temperature.

A test of the data-acquisition system demonstrated satisfactory collection of the significant parameters.

A program is being written so that the collected data can be translated into frictional coefficients. A comparison will be made of the frictional coefficients under sodium with those measured in inert gas. Data points will be examined over the temperature range from 300 to 1000°F. A number of alloys have been received which will be used in future tests. The materials are being fabricated into vee blocks and shafts.

D. Fuel Development

1. Irradiation of Uranium-Plutonium Alloys

Jacket collapse above the U-Pu-Fs pin in each of three experimental EBR-II fuel elements* (see Progress Report for November 1966, ANL-7279, p. 23) was found on examination to have restrained the elongation of the fuel pins that was observed in 16 companion fuel elements with uncollapsed jackets.

Eighteen full-length fuel elements of U-Pu-Zr jacketed severally in tubing of V-20 w/o Ti, of Types 304 and 316 stainless steel, and of Hastelloy-X and -X-280 had last month attained a calculated maximum burnup of 4.7 a/o at a maximum jacket temperature of 630°C. The burnup of two additional rods operating at a maximum jacket temperature of 540°C has increased to about 5.2 a/o near the end of December.

*Hitherto referred to as "fuel rods."

Sixteen of the eighteen U-Pu-Zr fuel elements were removed from the reactor in December and are awaiting transfer to Argonne, Illinois, for a postirradiation examination after a maximum burnup of 5.1 a/o.

The fuel pins of the four elements remaining in the reactor are jacketed in V-20 w/o Ti tubing. Two of the elements contain U-Pu-Zr fuel and two U-Pu-Ti. Irradiation is proceeding at maximum jacket temperatures of 540°C.

Neutron radiographs through V-20 w/o Ti jackets showed last month that two Th-20 w/o U fuel pins irradiated at 560 and 590°C maximum jacket temperature in CP-5 to 11.9 a/o U burnup had not elongated, but that Th-10 w/o Pu-10 w/o U pins irradiated, with jackets at 590°C, to 13.3 a/o burnup and Th-10 w/o Pu-20 w/o U pins irradiated, with jackets at 560°C, to 7.8 a/o burnup had elongated an average of 8%. The burnups of the Th-U pins have now increased to 13.6 a/o, and the burnups of the two sets of Th-Pu-U pins have now increased to 15.8 a/o (U + Pu) and 9.2 a/o (U + Pu), respectively.

Table VIII summarizes design details and irradiation variables for all current experiments with metal fuels as well as for the specimens recently removed from the reactor.

TABLE VIII. Status of Metal Fuel Irradiations in Progress--Fast Reactor Development

Test Reactor	Capsule or S/A No.	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)	Burnup to Date	
										a/o (U + Pu)	fiss/cc x 10 ⁻²⁰
EBR-II	XG06	ND23	U-15 Pu-10 Zr	66	V-20 w/o Ti	0.209	0.016	9.1	540	5.0	12.1
EBR-II	XG05	ND24	U-15 Pu-10 Zr	66	V-20 w/o Ti	0.209	0.016	9.1	535	4.8	11.6
EBR-II	XA07	ND28	U-15 Pu-9 Zr	75	304 SS	0.208	0.021	9.5	630	5.1	14.2
EBR-II	XA07	ND41	U-15 Pu-9 Zr	75	304 SS	0.208	0.021	9.5	625	4.9	13.9
EBR-II	XA07	ND32	U-15 Pu-9 Zr	75	316 SS	0.196	0.015	9.5	605	4.7	13.3
EBR-II	XA07	ND43	U-15 Pu-9 Zr	75	Hastelloy-X	0.196	0.015	9.5	615	4.9	13.7
EBR-II	XA07	ND25	U-14 Pu-12 Zr	76	304 SS	0.208	0.021	9.1	600	4.4	11.5
EBR-II	XA07	ND27	U-14 Pu-12 Zr	76	304 SS	0.208	0.021	9.1	605	4.5	11.7
EBR-II	XA07	ND26	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	8.6	590	4.3	11.1
EBR-II	XA07	ND29	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	8.6	595	4.4	11.6
EBR-II	XA07	ND30	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	9.1	615	4.9	12.7
EBR-II	XA07	ND31	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	9.1	610	4.7	12.3
EBR-II	XA07	ND33	U-14 Pu-12 Zr	76	Hastelloy-X	0.196	0.015	9.1	605	4.6	12.0
EBR-II	XA07	ND34	U-14 Pu-12 Zr	76	Hastelloy-X	0.196	0.015	9.1	610	4.7	12.2
EBR-II	XA07	ND35	U-14 Pu-12 Zr	76	Hastelloy-X	0.196	0.015	9.1	615	4.9	12.7
EBR-II	XA07	ND37	U-14 Pu-12 Zr	66	Hastelloy-X-280	0.208	0.015	8.6	610	4.9	12.2
EBR-II	XA07	ND39	U-14 Pu-12 Zr	66	Hastelloy-X-280	0.208	0.015	8.6	610	4.8	12.1
EBR-II	XA07	ND44	U-14 Pu-12 Zr	66	Hastelloy-X-280	0.208	0.015	8.6	600	4.7	11.7
EBR-II	XG05	NC17	U-15 Pu-10 Ti	63	V-20 w/o Ti	0.209	0.016	9.1	540	4.9	10.9
EBR-II	XG06	NC23	U-15 Pu-10 Ti	63	V-20 w/o Ti	0.209	0.016	9.1	540	5.2	11.5
CP-5	CP-50	1N16	Th-20 U	75	V-20 w/o Ti	0.196	0.015	8.7	590	13.6	6.5
CP-5	CP-50	4N19	Th-20 U	75	V-20 w/o Ti	0.196	0.015	8.7	560	13.6	6.5
CP-5	CP-50	2N17	Th-10 Pu-10 U	75	V-20 w/o Ti	0.196	0.015	9.2	590	15.8	7.4
CP-5	CP-50	5N20	Th-10 Pu-10 U	75	V-20 w/o Ti	0.196	0.015	9.2	590	15.8	7.4
CP-5	CP-50	3N18	Th-10 Pu-20 U	75	V-20 w/o Ti	0.196	0.015	8.7	560	9.2	6.7
CP-5	CP-50	6N21	Th-10 Pu-20 U	75	V-20 w/o Ti	0.196	0.015	8.7	560	9.2	6.7

^aBased on effective density.

The U-Pu-Zr and U-Pu-Ti specimens that were recently removed from CP-5 and that were reported (see Progress Report for October 1966, ANL-7267, p. 35) as having calculated burnups of 10.2 and 9.0 a/o,

respectively, have been subjected to radiochemical analyses to determine the actual burnup. The U-Pu-Zr specimen had reached a burnup of 12.5 a/o; the U-Pu-Ti specimen achieved a burnup of 10.7a/o.

Postirradiation metallographic examinations of the two specimens are being conducted. Transverse as well as longitudinal sections on these rods showed the fuel to be in excellent condition. The specimens were irradiated in V-20 w/o Ti tubing at maximum jacket temperatures of 640°C. There is no evidence of interaction of the fuels with this cladding material.

Microscopic examination of the U-Pu-Zr alloy shows interconnected fission gas bubbles, which apparently form a continuous path to the surface of the fuel. The supposition is supported by the large volume of fission gas that was recovered from the plenum above the fuel element. Although the U-Pu-Zr alloy showed radial cracks, there was no evidence of cracks in the U-Pu-Ti alloy. Also, the pore distribution was much finer in the U-Pu-Ti alloy than in the U-Pu-Zr alloy. The transverse sections of both fuel elements showed distinct bands that could be correlated to the transformation temperatures of the respective alloy phase.

Utilization of the volume of the gas released in the U-Pu-Zr element and that of the plenum, as determined by the actual sodium level in the specimen, made it possible to calculate an operating internal pressure of ~2200 psi, considerably lower than that calculated to produce jacket failure. It is apparent that the element was capable of attaining burnup higher than 12.5 a/o.

2. Oxide and Carbide Fuels

a. Irradiations of Ceramic and Cermet Fuels in EBR-II. In November, four elements fueled with UC-20 w/o PuC were charged into EBR-II to yield performance information at linear heat ratings of 16 to 25 kW/ft and a nominal burnup originally planned to be 5 a/o, but now scheduled for 6 a/o burnup, while similar rods, intended for burnup to 10 a/o, had achieved burnups of 2.7 to 3.9 a/o. Three similar fuel elements, intended for 5 a/o burnup had attained burnup levels of 4.5 to 4.6 a/o. The jacket materials for these fuel elements include Type 304 stainless steel, Hastelloy-X, and Nb-1 w/o Zr.

The elements intended for 6 a/o and 10 a/o burnup have now attained burnups of 0.50 to 0.80 a/o and of 3.0 to 4.3 a/o, respectively. Two are jacketed with Nb-1 w/o Zr, one with Hastelloy-X, and one with V-20 w/o Ti. Discharge of the other three, which were intended for 5 a/o burnup, is imminent, as their present burnup levels are now 4.8 to 5.0 a/o.

Nine fuel elements containing UO_2 -20 w/o PuO_2 , also under irradiation in November and still in EBR-II, are as follows: one in V-20 w/o Ti at 575°C , burnup now 3.0 a/o; five in Type 304 stainless steel at 555 to 630°C , burnup now 3.0 to 3.1 a/o; three in Hastelloy-X at 590 to 615°C , burnup now 2.9 to 3.1 a/o. The mixed oxides are scheduled for discharge at 5 a/o burnup.

The burnup levels for the stainless steel-jacketed cermet SS-40 w/o PuO_2 , SS-27 w/o PuO_2 , and SS-27 w/o UO_2 have increased during the month as follows: from 3.6 a/o in November to a current 4.2 a/o for the plutonia cermets, and from 2.8 a/o to 3.2 a/o for the urania cermets. The discharge level scheduled for the cermets is 7 a/o burnup.

Tables IX, X, and XI list the mixed oxide, mixed carbide, and stainless steel- PuO_2 or - UO_2 cermet fuel elements undergoing irradiation in EBR-II.

Three UC-20 w/o PuC elements, two with Type 316 stainless steel jackets and one with a vanadium jacket, and two UO_2 -20 w/o PuO_2 elements with Type 304 stainless steel jackets, all at burnup levels between 2 and 3 a/o, have been removed from the reactor for shipment to the Metallurgy alpha-gamma hot cell.

TABLE IX. Status of Irradiations of UC-20 w/o PuC Fuel 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 ($^\circ\text{C}$)	Burnup to Date	
								a/o (U + Pu)	fiss/cc $\times 10^{-20}$ (a)
XG05	SMV-2	84	304 SS	0.297	0.020	19.4	645	4.8	13.4
XG05	HMV-5	80	Hastelloy-X	0.297	0.015	22.0	670	4.0	13.1
XG05	NMV-11	84	Nb-1 w/o Zr	0.281	0.012	20.0	645	5.0	14.0
X008	NMP-2	82	Nb-1 w/o Zr	0.281	0.012	16.7	545	3.0	8.1
X008	NMV-4	80	Nb-1 w/o Zr	0.281	0.012	25.0	635	4.3	11.4
X008	NMV-7	80	Nb-1 w/o Zr	0.281	0.012	22.2	605	3.9	10.4
X008	NMV-12	86	Nb-1 w/o Zr	0.281	0.012	24.9	635	4.3	12.1
X008	HMV-1	80	Hastelloy-X	0.297	0.015	22.7	640	3.8	10.2
X008	HMV-4	80	Hastelloy-X	0.297	0.015	25.0	670	4.2	11.2
X008	HWMP-1	82	Hastelloy-X + W	0.297	0.020	16.1	555	2.9	7.8
X008	HWMP-1	83	Hastelloy-X + W	0.297	0.020	24.6	685	4.3	11.7
X015	NMP-1	80	Nb-1 w/o Zr	0.281	0.012	15.8	535	0.50	1.3
X015	NMV-3	80	Nb-1 w/o Zr	0.281	0.012	24.3	625	0.74	2.0
X015	HMV-2	79	Hastelloy-X	0.297	0.015	16.6	670	0.81	2.1
X015	TVMV-1	82	V-20 w/o Ti	0.297	0.020	21.4	620	0.65	1.8

^aBased on effective density.

TABLE X. Status of Irradiations of UO_2 -20 w/o PuO_2 Fuel 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 ($^\circ\text{C}$)	Burnup to Date	
							a/o (U + Pu)	fiss/cc $\times 10^{-20}$ ^a
SOV-3	83	304 SS	0.296	0.019	21.7	610	3.1	6.4
HOV-4	80	Hastelloy-X	0.295	0.014	22.5	600	3.1	6.0
TVOV-1	77	V-20 w/o Ti	0.297	0.020	20.8	575	3.0	5.7
SOV-7	85	304 SS	0.296	0.019	23.4	630	3.0	6.3
SOV-1	80	304 SS	0.296	0.019	19.9	590	3.0	5.9
HOV-10	86	Hastelloy-X	0.295	0.014	23.7	615	2.9	6.2
HOV-15	80	Hastelloy-X	0.295	0.014	21.1	590	2.9	5.8

^aBased on effective density.

TABLE XI. 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 ⁻²⁰ ^a
5P-9	SS-40PuO ₂	98	0.301	0.015	10.8	495	4.2	3.2
5P-12	SS-27PuO ₂	99	0.294	0.015	7.1	450	4.2	2.1
5U-14	SS-27UO ₂	98	0.298	0.013	5.6	435	3.2	1.6

^aBased on effective density.

b. Fluidized-bed Process for Preparation of (U, Pu)C. A series of twelve runs has been conducted to demonstrate the preparation of high-purity (U-15 w/o Pu)C by the fluidized-bed technique (see Progress Reports for June, July, and October 1966, ANL-7230, p. 45; ANL-7245, p. 10; and ANL-7267, p. 41). In each run, 300 g of uranium-plutonium alloy (3/8-in.-dia rods) were hydrided at 250°C to form a powder; this, in turn, was fluidized and reacted at 550 to 800°C with methane-hydrogen gas mixtures to form the monocarbide. These runs were carried out using once-through fluidizing gas flow, in a 1 $\frac{5}{8}$ -in.-ID reactor which could be manipulated (e.g., for product removal) in a high-purity helium-atmosphere glovebox. This reactor is capable of producing 600-g batches but the batch size was limited to conserve material. The run conditions and product analyses are shown in Table XII.

TABLE XII. Preparation of (U-15 w/o Pu)C in a Fluidized-bed Reactor
(300-g scale; 3/8-in.-dia alloy rods hydrided at 250°C prior to carbiding)

Run No.	Carbiding Conditions					Product Analysis (w/o)				X-ray Analysis ^c	
	Temperature (°C)	Pressure (atm)	CH ₄ Conc ^a (v/o)	Carbiding Time (hr)	Final H ₂ Treatment (hr)	Carbon	Oxygen	Nitrogen	Equivalent Carbon ^b	Major	Minor
1	750	4	10 ^d	6	0	4.11	0.04	-f	4.16	MC	BMH ₃
2	750	4	10 ^d	12	0	4.96	0.016	-f	4.99	MC	none
3	550-750	4	10	11.6	0	4.89	0.049	-f	4.95	MC	none
4	750	4	10 ^d	11.8	0	4.92	0.077	-f	5.00	MC	M ₂ C ₃
5	800	4	10	10.3	2	4.78	0.073	-f	4.85	MC	none
6	800	1.33 } 4 }	10	10.5	2	4.89	0.18	-f	5.05	MC	M ₂ C ₃
7	800	4	10	6.8	1.5	4.80	0.058	-f	4.86	MC	none
8	800	2	8	6.8	0.5	4.92	0.057	-f	4.98	none	none
9	800	4	-g	11.4	2.5	4.76	0.02	0.008	4.79	none	none
10	800	2	8	7.9	5.5	4.83	0.086	0.013	4.90	none	none
11	800	2	8	7.0	0.6	4.64	0.12	0.016	4.74	none	none
12	800	2	8	14.3	6.0	4.70	0.036	0.017	4.74	none	none

^aBalance hydrogen.^bOxygen and nitrogen are soluble in (U, Pu)C and replace carbon in the lattice; therefore, the equivalent carbon concentration in weight percent,C_{eq} = w/o C + (12/16) w/o O + (12/14) w/o N.^cM = (U-15 w/o Pu).^dNear the end of the run, CH₄ concentration was decreased to 2.5 v/o.^eThe higher values were assumed to be due to sample contamination. These higher values were not used in the equivalent carbon calculation.^fSpurious results; a value of 0.02 was assumed for calculating the equivalent carbon concentration.^gVariable as follows: 0 to 0.8 hr, 1.0 w/o; 0.8 to 3.8 hr, 2.6 v/o; 3.8 to 6.5 hr, 5 v/o; 6.5 hr to end of carbiding, 10.0 v/o.

In these runs the control of the carbon content and purity of the product were emphasized. Uranium-plutonium monocarbide that is stoichiometric or slightly hyperstoichiometric with respect to carbon is the preferred carbide fuel for fast reactors because (i) it is more compatible with cladding materials and has lower fission gas release during irradiation than carbide fuels with lower carbon-to-metal atom ratios, and (ii) it has higher thermal conductivity than fuels containing a large excess of carbon.

The methods for controlling the carbon content of the product in this series of runs were: (i) adjusting the methane concentration in the methane-hydrogen fluidizing gas to prevent formation of free carbon; (ii) stopping the reaction after the appropriate extent of carburization had been reached; and (iii) treating with hydrogen at the end of the carburization period to remove excess carbon.

The methane concentrations of the inlet and outlet gas streams were determined by infrared analysis. The rate of carburization was determined from the difference between the inlet and outlet methane concentrations, and from the gas flow rate. The extent of carbiding during the course of the run was established with an accuracy of ± 5 to 10% by integrating the rate of carburization with respect to time. However, chemical analysis of the product yielded a more accurate value ($\pm 1\%$) for the final extent of carbiding, and the values obtained during the run were adjusted accordingly. It was thus possible to plot the extent of carbiding versus time and to determine the effect of various changes in the reaction conditions on the rate of carbiding.

In one of the early experiments, run 3, it was found that the rate of carbiding was increased by raising the temperature from 550 to 750°C (see ANL-7230, p. 45). Consequently, beginning with run 5, the temperature was maintained at 800°C, the upper operating temperature for this reactor.

The effect of methane concentration on the rate of reaction was investigated in run 9. The reaction rates at 1.0 and 2.5 v/o methane during the initial portion of run 9 were much lower than the rates at 5 and 10 v/o methane during later portions of the run. The effect of an increase in pressure, observed by comparing the carbiding rates for runs made at 2 atm with those made at 4 atm pressure, was a slight increase in reaction rate. The effect of the pressure change observed in run 6 is a special case that is discussed below.

In runs 2 through 9, carbiding was interrupted after a period of several hours to rehydride the residual metal in the particles and break up the carbide reaction layer which appeared to be inhibiting further carbiding (see ANL-7267, p. 41). After a carbon content of about 4.0 to 4.2 w/o was reached (stoichiometric monocarbide contains 4.80 w/o carbon), the rate of reaction decreased markedly. Lowering the temperature to 25-400°C at this point in runs 2 through 9 to rehydride the residual metal permitted the completion of monocarbide formation in a few hours. This effect can be seen in the data from run 7 in Fig. 7.

In all runs the methane concentration in the methane-hydrogen fluidizing gas was such that $(U, Pu)_2C_3$ would be expected to form if the reaction were carried to completion; however, except for run 6 (discussed below), free carbon would not be expected to form. Any carbon in excess

Fig. 7. Effects of Process Variables on the Preparation of (U,Pu)C in a Fluidized Bed

of the stoichiometric carbon level (4.80 w/o) can be removed at the end of the carbiding period by reaction with hydrogen to form methane (see ANL-7267, p. 41). In run 11, it was found that a hydrogen treatment at the carbiding temperature prior to reaching the stoichiometric carbon concentration also resulted in the release of carbon in the form of methane, as shown in Fig. 7. This result suggests the presence of a sesquicarbide layer on the outside of the particle, an intermediate layer of monocarbide, and a core of unreacted alloy. (In actuality, the layers are not sharply defined since the particles are very irregular in shape and possess interconnected porosity.) The conversion of the sesquicarbide layer to the monocarbide apparently resulted in an increase in the rate of carbiding, but by a different mechanism than that which took place with the low-temperature hydriding of run 7.

Another approach to increasing the rate of carburization was to use a double hydriding cycle prior to carbiding; this was tried in run 5. It is well known that repeated hydriding and dehydriding results in a finer particle size than that from a single cycle. A smaller particle reduces the length of the diffusion path for carbon and, therefore, might be expected to be beneficial. However, the desired result was not obtained by use of this technique; the reaction rate was lower for run 5 than for the other runs, which had only one hydriding cycle prior to carbiding. A particle-size analysis of the product from run 5 showed a substantial portion (14.2 w/o) of large particle agglomerates. These agglomerates were easily broken into fine particles. It appears, therefore, that very fine particles were formed during the two hydriding cycles but that agglomeration of the particles resulted in a lower carbiding rate than in runs in which a single hydriding cycle was used.

In two of the runs, no attempt was made to increase the rate of carbiding. In run 1, in which the period of uninterrupted carbiding was only 6 hr, the product was only 85% converted to monocarbide. In run 12, the 12.5 hr required to reach the stoichiometric monocarbide carbon content (total carbiding time, 14.3 hr) was a longer time than that used in any of the other runs. Therefore, a hydrogen treatment during carbiding appears to be beneficial, but the best sequence of operation has not been established as yet.

As noted above, the operating conditions during carbiding in all runs except run 6 were such that free carbon would not be expected to form. During the first $2\frac{3}{4}$ hr of run 6, the methane concentration in the methane-hydrogen fluidizing gas was maintained at 10 v/o at a total pressure of 1.33 atm, conditions which would cause deposition of free carbon by the reaction

where the equilibrium constant k is $p_{\text{H}_2}^2/p_{\text{CH}_4} = 21.3$ atm at 800°C . A comparison of the rate of carbiding of run 6 with that of run 7 in Fig. 7 indicates the undesirable effect of carrying out the reaction under conditions that produce free carbon. After $2\frac{3}{4}$ hr, the rate of carbiding in run 6 was considerably lower than that in run 7. Raising the pressure to 4 atm resulted in reaction of the excess carbon to form methane and, ultimately, in an increase in the reaction rate.

An analysis of the data on rates of reaction indicated that it is possible to prepare 100 kg of (U, Pu)C by this process in a period of 10 to 12 hr if the carbiding is carried out at a pressure of 10 atm and a reactor of suitable size (8 in. in dia) is employed. This rate of production is sufficient to resynthesize fuel from three 1000-MW(e) nuclear power reactors.

The oxygen impurity levels for these products have been low, as shown in Table XII; frequently the two analyses of the product averaged about 0.05 w/o or lower. The two individual oxygen analyses for runs 2, 4, and 5 varied markedly and, therefore, they are listed separately. (Only the lower oxygen values for these runs were used in determining the equivalent carbon contents of Table XII.) The increase in oxygen content above about 0.05 w/o is almost certainly caused by the introduction of oxygen during sampling prior to analysis. The very low oxygen levels of some runs approach the impurity levels of the alloy feed material, about 0.01 to 0.02 w/o.

Difficulties were encountered in the earlier analyses of the products for nitrogen, and those reported previously for the first eight runs (ANL-7267, p. 41) are believed to be erroneously high. It is now believed that the actual nitrogen content of the first eight runs was similar to that of the last four runs (see Table XII).

The low impurity levels obtained in these runs indicate that the reactant gases, methane and hydrogen, introduce very small amounts of impurities, a conclusion that is in agreement with calculations based on the flow rates and the impurity contents of the gases.

The equivalent carbon contents of the last eight runs, in which a final hydrogen treatment was employed, ranged from 4.70 to 5.05 w/o. It is anticipated that this range will be narrowed when process conditions are optimized.

Sieve analyses of the carbide products showed an average particle size of about 40 to 60 μ . This relatively fine material is suitable for fabrication into pellets by pressing and sintering, with no intermediate processing. Preliminary work on pressing and sintering of one batch of this material has shown that densities in excess of 90% of theoretical can easily be obtained (see Progress Reports for October and November 1966, ANL-7267, p. 59, and ANL-7279, p. 27).

In summary, the following conclusions have been drawn from these studies on the preparation of (U, Pu)C by the fluidized-bed technique:

(i) The operating conditions of pressure, temperature, and methane concentration should be such that free carbon does not form on the particles.

(ii) The rate of carbiding increases with increasing pressure, methane concentration (within the above restriction), and temperature (at least up to 800°C).

(iii) After a carbon content of about 4.2 w/o is reached, the rate of carbiding can be increased by treating the bed with pure hydrogen either at lower temperatures (below 400°C) to cause rehydriding and, apparently, to break up the outer reaction layer, or at the carbiding temperature, to remove a higher carbide reaction layer.

(iv) A final hydrogen treatment is effective in removing any excess carbon from the surface of the carbide particles to yield the monocarbide.

(v) It has been demonstrated that a powdered product can be produced which contains less than 1000 ppm oxygen and less than 200 ppm nitrogen; this material is suitable directly for fabrication into pellets by pressing and sintering.

3. Fuel Cladding and Structure

a. Jacket Development. In the production of 0.290-in.-OD (0.015-in.-wall) V-15 w/o Ti-7.5 w/o Cr tubing from extruded and swaged 0.80-in.-OD (0.20-in.-wall) tube blanks, the application of tube-reducers was reported (see Progress Report for September 1966, ANL-7255, p. 29) to improve the uniformity of deformation, and to circumvent problems of lubrication and losses that occur on pointing tubing for drawing.

Although the development of this alloy represents an important advance over V-20 w/o Ti for the jacketing of higher-burnup fast-reactor fuels, V-15 w/o Cr-5 w/o Ti specimens fabricated from arc-melted buttons have since shown even better creep properties and compatibility with

carbide fuels. Test pieces of V-5 w/o Cr that were similarly fabricated have shown notable resistance to corrosion in sodium. In view of such results, a fabricability re-evaluation has prompted a scale-up in the fabrication of both the V-15 w/o Cr-5 w/o Ti and a binary vanadium-chromium alloy¹ to provide the material needed for a more definitive evaluation of fabrication parameters for rod, sheet, and tubing, and for a comprehensive testing program on finished (wrought) products.

Semiproduction (15-lb) ingots of V-15 w/o Cr-5 w/o Ti and V-10 w/o Cr have been consolidated by methods developed for the V-15 w/o Ti-7.5 w/o Cr alloy (see Progress Report for February 1966, ANL-7176, p. 26). Ingot chemistry is shown in Table XIII. Both 3.8-in.-dia ingots were machined, canned, and simultaneously extruded to bar stock. Re-extrusion of rod and sheet-bar billets has been completed, and the secondary fabrication of the rod and sheet-bar to finished products is pending. A tube-blank billet of each alloy also is available for re-extrusion.

TABLE XIII. Analytical Results for Arc-cast V-15 w/o Cr-5 w/o Ti and V-10 w/o Cr Ingots

Alloy Element (w/o) ^a	Ingot Number	
	AM-97 (V-15Cr-5Ti)	AM-99 (V-10Cr)
Chromium	14.4	10.2
Titanium	5.5	-
<u>Interstitial Element (ppm)^b</u>		
Carbon	280	310
Oxygen	900	705
Nitrogen	250	280
Hydrogen	13	22
Total Interstitials	1445	1320

^aAverage of three values from top, center, and bottom of ingot.

^bAverage of two values from top and bottom of ingot.

b. Corrosion of Vanadium-base Alloys in Sodium. After 57 days of a continuing test of resistance to corrosion in flowing sodium at 650°C (oxygen <10 wt ppm) the resistance of V-5 w/o Cr is still better by an order of magnitude than that of V-20 w/o Ti.

The progress report for November (ANL-7279, p. 30-32) contained a preliminary discussion of some of the first data developed in this study, and also two misstatements that inadvertently occurred:

¹The V-10 w/o Cr alloy is favored for scale-up rather than the V-5 w/o Cr alloy because of greater strength believed to result from the higher chromium content. The fabrication development of V-10 w/o Cr is, however, expected to be more difficult.

(1) The sodium velocity in the test at an oxygen concentration of 20 wt ppm was 15 cm/sec, not 15 ft/sec.

(2) A logarithmic plot of the depth of the hardened layer versus time of exposure results in straight lines for oxygen concentrations of 20 wt ppm and <10 wt ppm. (Since there are only two data points at 20 wt ppm, the significance is uncertain.) The slope of the 20 wt ppm line is 0.50; that of the <10 wt ppm line is 0.37. It is therefore not accurate to speak of parabolic relationships.

The reported considerable degradation of creep properties of the sample exposed in sodium containing 20 wt ppm oxygen (even after allowing for the decrease in load-bearing area) is of great importance. Of equal importance is the necessity to determine if degradation of creep properties is, in fact, simply related to the thickness of the hardened layer. Accordingly, samples of V-20 w/o Ti are now being exposed at 650°C in sodium, containing <10 wt ppm oxygen, for the same time that is required to produce the same thickness of ($\sim 30 \mu$) of hardened layer that is produced after 7 days in sodium containing 20 wt ppm oxygen.

4. Fuel Reprocessing

a. Processes for Fast Reactor Fuels. The 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, September 1966, ANL-7255, pp. 30-31). The process utilizes liquid metal-molten salt extractions and salt-transport separations² for the separation of fissile and fertile constituents of the fuel from the fission products.

Chemical studies are being conducted to provide basic data of process importance in the development of the salt-transport concept. Laboratory studies in support of this step have included a determination of the temperature dependence of the solubility of liquid magnesium metal in molten MgCl_2 . The solubilities were found to increase about twofold over the temperature range from 750 to 850°C; the concentrations of metallic magnesium were about 0.13 and 0.25 w/o, respectively, at 750 and 850°C. These studies are of importance for the prediction of the quantity of magnesium which may be transferred in the salt-transport separation step between alloys of different magnesium concentrations.

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

²Salt-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.

These studies include the investigation of process concepts on a pilot-plant scale, development of pilot-scale equipment, and evaluation of equipment components. This work requires a variety of experimental and supporting facilities, some of which are described below.

(i) Salt-transport Separations. The engineering feasibility of the salt-transport separation step has been demonstrated with uranium (see Progress Report, July 1966, ANL-7245, p. 14). Similar experiments with plutonium are planned. Until suitable glovebox facilities for pilot-plant studies are completed, experiments will be conducted in a walk-in-type hood which is capable of safely handling up to 100 g of plutonium. Installation of the experimental equipment in the hood has been completed and shakedown runs without plutonium are being continued (see Progress Report for November 1966, ANL-7279, p. 37). Information obtained in these experiments will be useful for designing process equipment to be installed later in the glovebox facility. The gloveboxes and components for an inert atmosphere control system have been ordered.

(ii) Pumped Salt Loop. A stainless steel loop (see Progress Report for April 1966, ANL-7204, p. 27) through which molten 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl was circulated was successfully operated for 1000 hr at various temperatures between 515 and 600°C before being voluntarily shut down. During this period, the loop was utilized for testing equipment items and instruments, which included a centrifugal pump (obtained from Oak Ridge National Laboratory), an induction pressure sensor to measure the pump-discharge pressure, a freeze valve, a bellows-sealed throttle valve, and an orifice flow meter. The loop and its components have performed satisfactorily. Design features of the pump are being incorporated into experimental pumps being developed for pilot-scale pyrochemical process equipment. The salt in the loop was found to contain satisfactorily low levels of corrosion products (iron, nickel, and chromium) and contaminants (oxygen and carbon). Uranium trichloride in molten salt solutions may be involved in pyrochemical processes. Therefore, plans are being made to circulate a molten chloride salt containing UCl_3 through the same loop and to use the same components to test the solution stability of UCl_3 in stainless steel equipment.

(iii) Liquid Metal-Molten Salt Continuous Contacting Equipment. Batch-processing methods are being developed for current pyrochemical processes. A small experimental effort, however, is being initiated to evaluate the technical feasibility of continuous processing.

(iv) Uranium Removal from Metal Crucible. In the current reference flowsheet, about 16 kg of uranium mixed with a small quantity of magnesium are accumulated in one of the process crucibles. Removal and recovery of this uranium by dissolution in a Zn-Mg solution would require

retorting large quantities of magnesium-zinc. Previous experiments³ showed that it is possible to disintegrate precipitated uranium enveloped in a Zn-Mg matrix by a hydriding procedure. However, in the limited number of experiments conducted at that time, it appeared that conversion of the precipitate to a free-flowing powder was not fully successful if the uranium was in low concentration or segregated within the Zn-Mg matrix. Since the concentration of uranium in the magnesium matrix in the current reference flowsheet will be considerably higher, it is anticipated that the hydriding procedure will be more successful. Equipment is being assembled to conduct this investigation.

(v) Salt Purification. The pilot-plant facility for makeup and purification of molten chloride salt mixtures (see Progress Report for July 1965, ANL-7082, p. 28) in 100-lb batches has been installed in a new location. The purification procedure that was previously used is being followed. This procedure consists of contacting the impure molten salt with liquid Cd-30 w/o Mg alloy so as to oxidize the impurities and remove the reaction products by settling and filtering of the supernatant salt. The purified salt is now being discharged to an inert-atmosphere glovebox and cast into a thin slab to permit easy breaking into small pieces. The facility is currently being operated to provide purified salt (42.5 m/o NaCl-42.5 m/o CaCl₂-15 m/o MgCl₂) for pilot-plant testing of the oxide reduction-partition step in the current reference flowsheet.

E. Design Concept Analysis and Advanced Systems Evaluation

1. 1000-MWe Study

At the request of the Division of Reactor Development and Technology, AEC, a meeting was held in Washington on November 30 and December 1, which resulted in changes in the proposed contract documents, essentially for clarification purposes. The revised documents were sent to the proposed contractors, and contract negotiation meetings were held at General Electric Company on December 20, and at Atomics International on December 21.

F. General Research and Development

1. Fast Reactor Core Parameter Study

A summary report of the work completed to date on the fast power reactor parameter study has been prepared. This report includes the following:

³Chemical Engineering Division Semiannual Report, January-June 1965, ANL-7055, pp. 68 and 70-71.

- a. derivation of the equations which yield core composition as a function of core geometry, fuel properties, and operating conditions;
- b. basis for selection of the values used to describe the fuel properties and operating conditions;
- c. dimensional and compositional description of the reactor configurations selected for study;
- d. specific techniques employed to study the neutronic characteristics of the above reactors;
- e. background assumptions employed in the neutronics calculations;
- f. neutronic description of the above reactors.

The remaining work to complete the initial phase of the study is also outlined. This includes consideration of reactors having a core aspect ratio of 0.1, variation of both radial and axial blanket thicknesses, and calculation of sodium-void coefficients.

The initial work was based on mixed-oxide-fueled cores containing only fuel, coolant, and structure. The parametric study is now being extended to include the effects of reactor control. A computer code is being developed to select core-subassembly loadings and control dimensions that closely approximate the desired operating conditions for the cylindrical core configurations being studied. This code will later be extended to perform the thermal-hydraulic analysis of cores that have a reduced radial maximum/average power ratio resulting from radial zoning of fuel enrichment.

III. GENERAL REACTOR TECHNOLOGY

A. Applied and Reactor Physics Development

1. Li⁶ Fast Neutron Spectrometer

Measurements were made in the center of the core of ZPR-3 Assembly 48 with the Li⁶-semiconductor sandwich fast neutron spectrometer.⁴ The device is composed of two silicon surface-barrier detectors mounted with sensitive areas facing each other and separated by a distance approximately equal to the diameter of the sensitive area. Midway between the diodes is a thin organic film, less than 10 $\mu\text{g}/\text{sq cm}$, which is coated with a layer of Li⁶F. The two products T and He of the Li⁶,n reaction are detected by the surface-barrier diodes and the output pulses summed to give a pulse proportional in height to the total energy, Q plus neutron energy, of the reaction. Fast coincidence requirements are used to eliminate unrelated events in each detector.

The general response of the instrument to a neutron flux is given by

$$C_i = \int_{E_n=0}^{E_{\text{max}}} \phi(E_n) \sigma(E_n) K_i(E_n) dE_n, \quad (1)$$

where C_i is the number of events observed in channel i ; $\phi(E_n)$ is the neutron-energy spectrum; $\sigma(E_n)$ is the Li⁶(n, α) cross section; and $K_i(E_n) dE_n$ gives the probability that an event due to a neutron having an energy between E_n and $E_n + dE_n$ results in a count in channel i .

For the practical case, this equation must be approximated by a matrix equation

$$C_i = \sum_{j=1}^{j=h} (K_{ij} \sigma_j \phi_j). \quad (2)$$

The matrix $(K_{ij} \sigma_j)$ represents the response of the spectrometer. Each column of the matrix describes the spectrometer response to neutrons in the j th energy channel, that is, each column describes the response to monoenergetic neutrons.

Measurements were made at the center of the core about one inch from the midplane, which is very near the position where proton-recoil measurements were previously made at a power of about 2.5 W. Following the core runs, the Li⁶ layer was removed and the background response of the same two diodes was measured and found to be negligible.

⁴Huber, R. J., Trans. ANS 7(2), 368 (1964).

In order to compare the proton-recoil data (see Progress Report for October 1966, ANL-7267, p. 21) with the Li^6 -spectrometer data it is necessary to convert the former to what one would expect if the "sandwich" spectrometer were placed in the spectrum measured by proton-recoil proportional counting. Experience has shown that the difference between these two data at the low-energy end is particularly sensitive to the portion of the neutron spectrum below about 20 keV.

The proton-recoil data were converted by use of Eq. (2). To make the calculation of the expected pulse-height distribution, the flux per unit lethargy as given by the proton-recoil experiment was extrapolated linearly to zero at thermal energy. A more rapidly decreasing estimate would have reduced the discrepancy in the low-energy region of the pulse-height distribution. The response matrix K_{ij} was constructed from the thermal-neutron response. The full width at half-maximum of the monoenergetic response for this experiment was 72 keV.

Fig. 8. A Comparison of Spectra as Measured by the $\text{Li}^6(n,\alpha)$ Semiconductor Spectrometer and Proton-recoil Proportional Counters

Figure 8 shows the results of the two counting methods. The agreement between the two is within counting statistics (about 400 counts/channel maximum) except at the peak corresponding to very-low-energy neutrons. This small discrepancy, however, corresponds to a very small difference in total low-energy flux in the core because of the very rapid increase in $\text{Li}^6(n,\alpha)$ cross section for low energy. In general, the results from the Li^6 spectrometer are consistent with those obtained from the proton-recoil measurements.

2. Variational Methods for Two-dimensional Kinetics Calculations

An experimental method of two-space-dimensional, time-dependent reactor analysis is based on the use of the variational method to find the space-dependent flux shapes. The fluxes are fit with quadratic surfaces in the interior of each spatial region, with appropriate

boundary conditions. A code which exemplifies this method is VARI-QUIR,⁵ which has been developed at Westinghouse Astronuclear Laboratory; VARI-QUIR has been modified and compiled for execution on the CDC-3600 F at ANL.

⁵Riese, J. W., Collier, G., and Riech, C. E., VARI-QUIR, A Two-dimensional Time-dependent Multigroup Diffusion Code, WANL-TNR-133 (revised) (Dec 1965).

The modifications included substitution of a FORTRAN subroutine ICE, obtained from General Electric Co. (Nuclear Energy Division) for the FAP subroutine supplied by WANL and suitable modifications to the subroutine QUIR. The WANL test problem was successfully executed in 11 min 20 sec, and agreed with WANL results to one part in 10^5 .

Further modification includes cleaning up FORMAT statements, preparing sample feedback routines, and writing an overlay tape.

3. Effects of Cross-section Errors on Neutron Spectra

Errors in cross sections are only indirectly related to errors in spectral indices (see Progress Report for November 1966, ANL-7279, pp. 42-43); the errors in indices can be determined from comparisons of activation and perturbation measurements and calculations. The increasing ability to measure neutron spectra directly and accurately over a wide range suggests that this source be examined as a source of comparison measurements more directly related to cross-section errors.

To this end, consider a hydrogen-like medium in which the ratio of scattering to total cross section, σ_s/σ_t , is given by

$$\sigma_s/\sigma_t = 1 + \epsilon R(u),$$

where $R(u)$ is random on $(-1, 1)$ $\epsilon \ll 1$, and u is the lethargy variable. The item $\epsilon R(u)$ represents the error in σ_s/σ_t . In terms of stochastic integrals,⁶ the collision density is given by

$$F(u) = \int_{-\infty}^u F(u') [1 + \epsilon R(u')] du'.$$

Because $R(u)$ is, $F(u)$ is random. If we solve by successive approximations

$$F_n(u) = \int_{-\infty}^u F_{n-1}(u') [1 + \epsilon R(u')] du';$$

$$F_0(u) = e^u.$$

⁶Papoulis, A., Probability, Random Variables and Stochastic Processes, McGraw-Hill Book Co., Inc., (1965), pp. 323 et seq.

Then to the second order in ϵ ,

$$F_2(u) = e^u + \epsilon \left[\int_{-\infty}^u du' e^{u'} R(u') + \int_{-\infty}^u du' \int_{-\infty}^{u'} du'' e^{u''} R(u'') \right] \\ + \epsilon^2 \int_{-\infty}^u du' R(u') \int_{-\infty}^u du'' e^{u''} R(u''),$$

and

$$\phi_2(u) = [1 + R(u)] F_2(u)$$

is the neutron flux spectrum.

The average value of ϕ , the mean deviation, and the autocorrelation can all be defined, to order ϵ , from the expression for $\phi_2(u)$ and the properties of R .

Two random signals are easily used to exemplify cross-section errors:

1. the pseudorandom telegraph signal,⁷ which has the property

$$R(u) = 1 \text{ if } R(u) = 0 \text{ an even number of times in } (0, u); \\ = -1 \text{ if } R(u) = 0 \text{ an odd number of times in } (0, u),$$

and the zeros of R are distributed randomly with density λ .

2. The random telegraph signal:

$$R(u) = \text{the preceding process times } a(u)$$

where $a = \pm 1$ with equal probability.

In the first case, the expectation value of R is $e^{-2\lambda|u|}$ and the autocorrelation is $e^{-2\lambda|u-u'|}$. In the second case the expectation value is 0 and the autocorrelation is $e^{-2\lambda|u-u'|}$. By means of properties and the commutativity of the operation of integration and expected value, the expected value of ϕ_2 and its square are readily found.

⁷See Ref. 6, pp. 288-289.

4. The ARC System

a. Operation. Figure 9 schematically illustrates the operation of the ARC System. The Operating System manages the data handling and module linkages as well as dealing with user input and output information.

The standard-path modules are control programs which call ARC modules to accomplish a particular task. The module-calling sequence in a standard path can be influenced by results obtained earlier in the path. The standard paths and other ARC modules are stored in the module-library portion of the ARC Datapool.

Fig. 9. Schematic Diagram of ARC System, Showing Standard Path and F ARC Module Arrangements

Figure 9 represents a condition in the computer in which the Operating System has transferred one of the standard paths and one of the modules from the Datapool to the Fast Memory.

Table XIV reflects the current thinking in regard to the ARC module library for the system. The module names are intended to be descriptive of the function of the modules and correspond to the Module

Name or Designation in the module documentations which are now being prepared for inclusion in an ANL topical report describing the ARC System.

The listing has been grouped into five main categories. Within each category the modules are further organized into Input, Computational, and Output Edit modules. In all categories, a blank entry has been included to imply the open ended nature of the module library.

TABLE XIV. ARC System Modules

I. Cross-section-preparation Modules

A. Input Modules

1. Epithermal-cross-section Specifications
2. Thermal-cross-section Specifications
3. Broad-group Specifications
4. Broad-group Fundamental-mode Specifications
- 5.

TABLE XIV. (Contd.)

B. Computational Modules

1. Resonance Cross Sections
2. Nonresonant Cross Sections and Fundamental-mode Spectrum
3. Thermal Cross Sections
4. Broad-group Cross Sections
5. Broad-group Fundamental-mode Spectrum
- 6.

C. Output Edit Modules

1. Resonance-cross-section Edit
2. Nonresonant-cross-section and Spectrum Edit
3. Thermal-cross-section Edit
4. Broad-group Edit
5. Broad-group Fundamental-mode Edit
- 6.

II. Neutronics Calculations Modules

A. Input Modules

1. Neutronics Specifications
2. Geometry Preparation
3. Cross-section-homogenization Specifications
4. 1D-Diffusion Specifications
5. 1D-Transport Specifications
6. 2D-Diffusion Specifications
7. 2D-Transport Specifications
8. Diffusion Mesh-refinement Specifications
9. Transport Mesh-refinement Specifications
10. Neutronics Output Manipulation Specifications
- 11.

B. Computational Modules

1. Cross-section Homogenization
2. 1D-Diffusion
3. 1D-Transport
4. 2D-Diffusion
5. 2D-Transport
6. Diffusion Mesh Refinement
7. Transport Mesh Refinement
8. Criticality Search
9. Neutronics Output Manipulation
- 10.

C. Output Edit Modules

1. 1D-Diffusion Edit
2. 1D-Transport Edit
3. 2D-Diffusion Edit
4. 2D-Transport Edit
- 5.

TABLE XIV. (Contd.)

III. Adjunct Calculations Modules

A. Input Modules

1. Neutron-inventory Specifications
2. Diffusion-perturbation Specifications
3. Transport-perturbation Specifications
4. Spatial-synthesis Specifications
5. Group-collapse Specifications
6. Bivariate Cross-section-interpolation Specifications
- 7.

B. Computational Modules

1. Neutron Inventory
2. Diffusion Perturbation
3. Transport Perturbation
4. Spatial Synthesis
5. Group Collapse
6. Bivariate Cross-section Interpolation
- 7.

C. Output Edit Modules

1. Neutron-inventory Edit
2. Diffusion-perturbation Edit
3. Transport-perturbation Edit
4. Spatial-synthesis Edit
5. Group-collapse Edit
6. Bivariate Cross-section-interpolation Edit
- 7.

IV. Fuel Cycle and Depletion Calculations Modules

A. Input Modules

1. Fuel-cycle Specifications
2. Economics Specifications
- 3.

B. Computational Modules

1. Fuel Cycle
2. Economics
- 3.

C. Output Edit Modules

1. Fuel-cycle Edit
2. Economics Edit
- 3.

V. Safety and Accident Calculations Modules

A. Input Modules

1. Point-kinetics Specifications
2. Spatial-kinetics Specifications

TABLE XIV. (Contd.)

3. Heat-transfer Specifications
 4. Two-phase Coolant-flow Specifications
 5. Core-compaction Specifications
 6. Compressible-fluid-hydrodynamics Specifications
 7. Fuel-element-deformation Specifications
 8. Initiating-mechanism Specifications
 9. Fuel-Coolant Interaction Specifications
 - 10.
- B. Computational Modules
1. Point Kinetics
 2. Spatial Kinetics
 3. Heat Transfer
 4. Two-phase Coolant Flow
 5. Core Compaction
 6. Compressible Fluid Hydrodynamics
 7. Fuel-element Deformations
 8. Initiating Mechanisms
 9. Fuel-Coolant Interaction
 - 10.
- C. Output Edit Modules
1. Point-kinetics Edit
 2. Spatial-kinetics Edit
 3. Heat-transfer Edit
 4. Two-phase Coolant-flow Edit
 5. Core-compaction Edit
 6. Compressible-fluid-hydrodynamics Edit
 7. Fuel-element-deformation Edit
 8. Initiating-mechanism Edit
 9. Fuel-Coolant Interaction Edit
 - 10.

b. Burnup Calculations. The Phase-zero burnup package effort (see Progress Report for August 1966, ANL-7249, pp. 40-41) is complete in most essential features. An equilibrium test case has been successfully compared with the CYCLE-1 code. Methods for accelerating the iteration steps for the external cycle are being developed. Further improvement of the core cycle search procedures may be possible, depending on the results of this effort.

Preliminary ARC documentation of the Phase-1 (see Progress Report for May 1966, ANL-7219, pp. 34-35) specifications has been completed. The Phase-1 package for the IBM 360 Model 50 has been completely flowcharted and coded. Debugging of this package will begin in the near future.

The input modules have BCD upper interface data sets while the computational modules deal only with binary data sets. The edit modules have both BCD and binary upper interface data sets. Eventually, quite selective edits should be possible, depending upon user choice. At the moment, most edit modules will probably be similar to the classical code output routines in that the user either does or does not get the programmed output.

B. Fuels and Cladding

1. Irradiation of High-temperature Materials

Last month UC-20 w/o PuC, PuC and US, each in an Nb-1 w/o Zr jacket at 750°C (max), had attained maximum burnups of 6.9, 1.8, and 7.2 a/o of (U+Pu), Pu, or U, respectively, and electron-microscopical studies were initiated of fission-product distribution (gas and solid) of vibrationally compacted UC-20 w/o Pu after 6.9 a/o burnup. The irradiations, which are being made in instrumented capsules in MTR, are summarized in Table XV.

TABLE XV. 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
56-11	MV-2	UC-20 PuC	79	0.281	0.021	1.2	470	6.6	17.0
56-8	MV-3	UC-20 PuC	81	0.281	0.012	1.2	715	7.1	18.8
56-8	MV-5	UC-20 PuC	80	0.281	0.012	1.2	705	6.8	17.8
56-11	MV-6	UC-20 PuC	80	0.281	0.012	1.2	480	7.1	18.6
56-13	Z-4	UC-20 PuC	79	0.174	0.015	1.2	665	2.0	5.2
56-13	Z-5	UC-20 PuC	79	0.174	0.015	1.2	585	2.0	5.2
56-13	Z-7	UC-20 PuC	79	0.174	0.015	1.2	570	2.0	5.2
56-13	C-45	PuC	84	0.174	0.009	1.4	700	2.0	5.5
56-8	S-7	US	80	0.281	0.012	1.0	535	5.4	10.5
56-8	S-8	US	89	0.281	0.012	1.0	725	7.4	16.0
56-8	S-9	US	76	0.281	0.012	1.0	750	7.4	13.6
56-8	S-10	US	91	0.281	0.012	1.0	690	7.4	16.4
56-11	S-15	US	82	0.281	0.012	1.0	380	4.5	9.0
56-11	S-16	US	90	0.281	0.012	1.0	510	6.4	14.0
56-11	S-17	US	88	0.281	0.012	1.0	500	5.1	10.9
56-11	S-18	US	77	0.281	0.012	1.0	610	6.9	12.9

^aBased on effective density.

2. Radiation Damage to Structural Materials

a. Effect of Fast-neutron Irradiation on Jacket Materials. An investigation of irradiation with respect to tensile, creep, creep-rupture, and tube-rupture properties 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 is in progress for the exposure range of 10^{21} to 10^{23} n/cm² at temperatures in the range from 400 to 750°C. Twenty-six tensile-type specimens of V-20 w/o Ti, three of Hastelloy-X, and ten of Type 304 stainless steel were removed from EBR-II last November after an exposure of 2.7×10^{22} n/cm² at 670°C, but 24 capsules, containing 103 tube-burst specimens and 321 tensile-type specimens are being irradiated in EBR-II subassemblies XA08, XO10, and XO18 at temperatures between 500 and 650°C. These specimens have accumulated maximum total neutron exposures in the range from 4×10^{20} up to 1.6×10^{22} n/cm². The status and identification of these capsules are given in Table XVI.

TABLE XVI. 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	3.4×10^{22}
XA08	AS-2	V-20 w/o Ti	Tube-burst	12	Argon-helium	540	3.4×10^{22}
XA08	AS-3	Hastelloy-X	Tube-burst	12	Argon-helium	540	3.6×10^{22}
XA08	AS-4	Hastelloy-X	Tube-burst	12	Argon-helium	540	3.6×10^{22}
XA08	AS-5	304 SS	Tube-burst	12	Argon-helium	540	3.6×10^{22}
XA08	AS-6	V-20 w/o Ti	Tensile	16	Argon-helium	580	3.7×10^{22}
XA08	AS-7	Hastelloy-X	Tensile	16	Argon-helium	580	3.4×10^{22}
XA08	AS-8	304 SS	Tensile	16	Argon-helium	580	3.4×10^{22}
XA08	AS-12	V-20 w/o Ti	Tensile	16	Argon-helium	580	3.3×10^{22}
XO10	AS-16	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.2×10^{22}
XO10	AS-17	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.3×10^{22}
XO10	AS-18	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.2×10^{22}
XO10	AS-19	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.2×10^{22}
XO10	AS-20	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.4×10^{22}
XO10	AS-21	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.6×10^{22}
XO10	AS-22	Hastelloy-X	Tensile	13	Argon-helium	500	1.4×10^{22}
XO10	AS-23	304 SS	Tensile	13	Argon-helium	500	1.6×10^{22}
XO10	AS-24	304 SS	Tensile	13	Argon-helium	500	1.4×10^{22}
XO10	AS-25	304 SS	Tensile	13	Argon-helium	500	1.3×10^{22}
XO10	AS-26	304 SS	Tensile	13	Argon-helium	500	1.3×10^{22}
XO18	AS-56	V-20 w/o Ti, V-15 w/o Ti- 7.5 w/o Cr	Tensile, Tube-burst	75	NaK	650	4.0×10^{20}
XO18	AS-57	304 SS, Hastelloy-X	Tensile, Tube-burst	75	NaK	650	4.0×10^{20}
XO18	AS-58	V-20 w/o Ti, V-15 w/o Ti- 7.5 w/o Cr	Tensile, Tube-burst	25	NaK	650	4.0×10^{20}

EBR-II subassembly XA07, containing 48 tensile-type specimens made severally of V-20 w/o Ti, Hastelloy-X, and Type 304 stainless steel, was removed after accumulating a maximum total neutron exposure of 3.8×10^{22} n/cm². Arrangements are now being made to ship the specimens to the Illinois site. EBR-II subassembly XO18, containing 63 tube-burst specimens and 112 tensile-type specimens of V-20 w/o Ti, V-15 w/o Ti-7.5 w/o Cr, Hastelloy-X, and Type 304 stainless steel, was inserted in Row No. 2 during this report period for irradiation to 1×10^{23} n/cm² at 650°C.

As an aid to an interpretation of the results being obtained, a characterization of the neutron spectra (flux and energies) in EBR-II has become highly desirable. A special short run at low power is therefore to be made at EBR-II when the core is loaded to full size. Assemblies have been constructed (by Argonne National Laboratory, Idaho Division) that can accommodate capsules for irradiation of detecting foils.

Stainless steel capillary tubes have been loaded with iron, nickel, copper, titanium, and aluminum-cobalt wires. These have been shipped to EBR-II for inclusion with other experiments in the special capsules. Calculations of the neutron spectra in the EBR-II core and blanket are being analyzed for application to the experimental data that will be obtained from the foils.

3. Techniques of Fabrication and Testing

a. Tungsten-base Alloy Tubing. For the past several years, a long-range program has been under way at Argonne National Laboratory to devise and develop a practical technology for the fabrication of tungsten and tungsten-alloy tubing for high-temperature applications. Since July 1966, a change in programming has permitted a much-needed concentration on process development with less emphasis on the delivery of high-quality finished tubing for application evaluation.

The existing technology⁸ consists of primary fabrication by the "filled-billet" extrusion technique. Although this method has consistently produced high-quality tube-blanks, minor surface striations have plagued subsequent processing. Secondary fabrication has followed two alternative warm-drawing routes: ductile-core drawing or plug-drawing. In the former method the metal core is retained in the tube until after extrusion, and drawing to the final tube size. The core is then removed by acid dissolution. This is an unattractive and time-consuming process, the time for core removal being dependent upon the final inner diameter and length of the tubing. Plug-drawing is performed on the extruded tube-blank after core removal.

All of the above fabrication operations have been beset with difficulties such as surface striations resulting from the extrusion operation, lubrication problems, swage pointing failures, tooling inadequacies, and many others. Some of these difficulties have received attention; some remain to be resolved.

The continued application of the extrusion technique to billets that are "filled" or canned for mandrel use requires that means for economic conditioning of the extruded tube-blank be considered. Since one such method is tube-reduction, the Argonne National Laboratory three-roll tube-reducer is being modified for warm working operations (see Progress Report for November 1966, ANL-7279, pp. 57-58). By tube-reduction of some extruded W-Re-Mo alloy, we have successfully improved both the outer and inner surfaces. This work will continue and, it is hoped, be extended to tungsten and to tungsten-rhenium alloys, both of which are more difficult to fabricate than the ternary alloy.

A variety of lubricants are being evaluated. Early results indicate that an alcohol dispersion of WS_2 can produce a better drawn surface than that obtained with the formerly used graphite-sugar-water mixture. Conventional cold-swaging dies have been substituted for the hot-working dies previously used, and furnace-tip heating has been replaced by inductive (RF) heating. Considerably improved swaged-tip surfaces have resulted.

⁸Burt, W. R., Jr., et al., ANL-7151 (July 1966), Development of Techniques for Fabrication of Small-diameter Thin-wall Tungsten and Tungsten-alloy Tubing, Interim Progress Report Covering the Period from January 1, 1964 through December 31, 1965.

Improvements in swage-pointing have resulted in more successful final drawing operations. Attempts to improve the economics of these operations by eliminating both drawing and annealing sequences have met with some success and continued effort will be concentrated in this area.

C. Engineering Development

1. Electric Master-Slave Manipulator, Mark E4A

The electrically actuated brakes for the slave-arm motions (see Progress Report for November 1966, ANL-7279, p. 60) have been completed in design and detail; some parts are on order. Clutches, preset at about $3/4$ of the maximum load capacity of each motion, are incorporated to protect the manipulator from external loads when the brakes are on. The brakes will be automatically set by springs whenever there is an electrical interruption or electrical malfunction as indicated by a special safety circuit. The brakes can also be set by switches on the handle.

The stiffness of the Z motion, when in 1:1 force ratio, has been increased by a factor of three. This was brought about by changes in the amplifier, that is, by use of a ring modulator and by increasing the tachometer and positional gains. High-precision bearings on the output shaft of the Z-motion gear box were installed to reduce the mechanical backlash. The increased stiffness and decreased backlash improve the "feel" considerably, and it is planned to make these changes in the other motions.

A new synchro gear system has been designed and is being detailed. The new gear ratio will increase the synchro null spacings from about 8 to 32 in. (as measured at the wrist joint) in the X, Y, and Z motions. This should eliminate inadvertent "jumping" of nulls when the slave arm is overloaded. The position signal will be reduced by a factor of four, but tests show that the signal-to-noise ratio is still acceptable.

2. Studies of Structural Dynamics of Core

By means of a combined theoretical and experimental program, structure-fluid interactions will be studied as they relate to the performance, stability, and reliability of reactor core components.

The initial effort, related to the theoretical phase, will be an in-depth analysis of literature in the field of flow-induced vibration. The outcome will be a survey report discussing possible excitation mechanisms and the nature of fluid-structure coupling, presenting the state-of-the-art in explaining and predicting observed vibration phenomena, and outlining theoretical approaches to be pursued.

An experimental program will be run concurrently. The first phase will be a laboratory experiment to demonstrate the phenomena of parallel flow-induced vibration of rods. Towards this end, a small bench-top flow loop is presently being constructed.

The pump, chosen because of its availability, has a maximum flow-rate of approximately 30 gpm. The test set-up features a removable test section, $2\frac{3}{16}$ in. in diameter and 34 in. long. The test section will accommodate rods, up to 30 in. in length, of various materials and cross sections. For initial tests, circular brass rods of 0.063-, 0.090-, and 0.125-in. dia have been chosen.

The tests are intended to demonstrate measurable oscillations of the rods. Future tests will be instrumented to measure pressure, displacements and strains, thus yielding pressure- and displacement-time histories which can be analyzed for harmonic content or correlated, leading to further insight into the overall problem and design of experiments.

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

a. Boiling Liquid Metal Technology

(i) Niobium-1% Zirconium Loop. This facility is designed to investigate the heat transfer and two-phase flow characteristics of boiling sodium to a temperature of 2100°F and a pressure of approximately 8 atm. Among the variables to be investigated are boiling heat flux and temperature difference up to the occurrence of critical flux, boiling and adiabatic two-phase pressure losses, vapor volume fraction, boiling-stability parameters, and ultimately the transient behavior of some of these same quantities.

Operation of the vacuum chamber was initiated early in December. After several days of pump-down, malfunctions of the partial-pressure analyzer and of one ion gauge were detected and testing discontinued. Inspection of the mass-spectrometer tube revealed a thin film of copper oxide over several surfaces; the tube was dismantled and thoroughly cleaned. The ion gauge has been replaced.

Several other modifications were completed: an alarm was incorporated into the ion-gauge circuit to warn of very small increases in vacuum chamber pressure; a 500-W quartz lamp was installed to provide better porthole observation of loop components during high-temperature operation; a 50-kW wattmeter was installed for monitoring boiler power; several auxiliary thermocouples were installed at supplementary loop positions; the new power supplies for the pressure transducers were installed; an initial sodium sample was taken from the dump tank, the first of several samples to be taken during long-term loop operation; the argon-clean-up system is being modified to allow the use of very pure argon for field-welding experiments.

(ii) Electron-bombardment Heater (EBH) Experiment. This experiment has been shut down due to a failure at the outside wall of the sodium vessel. This vessel has been repeatedly exposed to temperature greater than 1600°F, and it is questionable whether or not this facility is still useful. A total of 900 hr of testing at heat fluxes up to 350,000 B/hr-ft² (anode flux) have been completed, and this is in excess of the originally planned life of the experiments. Although a short, high-powered energy burst was planned for the final test, it appears that tests at higher flux should be made in the forthcoming EBH test facility now under design. A description of this new facility and its application to EBH designs for liquid metal loops will be described in a subsequent progress report.

b. General Heat Transfer

(i) Countercurrent Turbulent Liquid Metal Flow. The impeller of the canned-rotor pump has been "reworked" for mercury operation and replaced in the pump. Tests in the mercury flow loop showed that the pump now performs satisfactorily. Installation of the second heat-exchanger test section, as well as loop clean-up, are in progress. Design of the third test section continues.

Recent analytical studies concentrated on extensions of the special treatment required for the case of equal heat capacity mass flow-rates on both sides of the exchanger, and on attempts to find an improved computational procedure for applying the existing mathematical solution for counter-flow heat exchangers. The former is being reformulated for application to cases of practical interest and future publication; the latter has not been successful.

(ii) Nonsymmetrical Geometries. Fabrication of a small heat transfer loop for the purpose of performing experiments with sodium in model heat exchangers has begun.

(iii) Liquid-metal-heated Steam Generators. Basic research on heat exchangers for use in liquid-metal-heated steam generators has begun. Information related to the heat transfer design and operation of existing liquid-metal-heated steam generators is being collected.

c. Effects of Interfacial Transport Processes on Film Condensation. Modification of the film injector has been completed, and the loop is operative. Flow patterns in the film have been studied by use of the dye-injection technique. At both high and low water and air flows, the streamlines across the channel width were found to be parallel. These results indicate that the desired two-dimensional flow pattern has been achieved and that secondary flows have been eliminated.

The entrance effects resulting from the exchange of momentum between the air and water streams will now be studied. During these runs, different types of instrumentation for measuring film thickness will be tested.

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 fluorinating UO_2 - PuO_2 pellets containing fission products. Current emphasis in these studies is on the use of BrF_5 as a selective fluorinating agent for conversion of uranium oxides in uranium-plutonium mixtures to volatile UF_6 . In this reaction, 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. A recent experiment (Purse-13) was performed as one of a series of experiments to determine the effect of the temperature of the BrF_5 fluorination step upon plutonium retention in the final alumina bed. In previous experiments of this series, BrF_5 fluorination temperatures of 200°C (see Progress Report for April 1966, ANL-7204, p. 63), 300°C (see Progress Report for February 1966, ANL-7176, p. 65), and 400°C (see Progress Report for July 1966, ANL-7245, pp. 45-46) were evaluated. In the recent experiment, the BrF_5 fluorination step was carried out at 250°C . The charge to the reactor in Run Purse-13 consisted of 650 g of pellets and 1100 g of alumina containing 0.6 g CsF and 0.15 g RbF. The processing sequence consisted of: oxidation of the pellets for 4 hr at 450°C with 20 v/o oxygen, fluorination with 8-10 v/o BrF_5 at 250°C for 2.8 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 .

The final alumina bed contained 0.005 w/o uranium and 0.007 w/o plutonium, which corresponds to the removal of more than 99.9% of the uranium and 97.3% of the plutonium present in the charge. These residual concentrations are nearly identical to those obtained using a BrF_5 fluorination temperature of 300°C (see ANL-7176, p. 65), which were the lowest values achieved in the three prior experiments. Plutonium removals of 99% can be achieved by reuse of the alumina bed (see Progress Report for October 1966, ANL-7267, p. 66). On the basis of the present data, it appears that the best temperature for conducting the BrF_5 fluorination step is within the range from 250 to 300°C . However, to explore this area more completely, experiment Purse-15 has been performed with a BrF_5 fluorination step at 350°C . The results of analyses of the final bed from this experiment are not available.

A series of two experiments were performed to determine if the presence of BrF_3 in fluorine would suppress fluorination of plutonium. The experiments were carried out in a horizontal tubular reactor using simulated fuel mixtures containing 10 g uranium as U_3O_8 , 0.3 g plutonium as PuO_2 , 0.2 g nonradioactive fission product oxides and 12 g of alumina. A gas stream containing 93 v/o fluorine-7 v/o BrF_3 was passed over the fuel mixtures at 250°C for 6 hr in the first test and at 350°C for 3 hr in the second test. In the first experiment, about 40% of the plutonium was volatilized as PuF_6 ; about 80% was volatilized in the second experiment. Apparently, the presence of BrF_3 at this concentration level in fluorine does not suppress the fluorination of plutonium.

(ii) Neptunium Fluoride Chemistry. Laboratory-scale studies are being carried out to determine the behavior of neptunium when oxidized nuclear fuel is reacted with bromine pentafluoride. Previous experiments have shown that NpF_6 reacts readily with bromine (the product of the reaction between U_3O_8 and BrF_5) at 30°C to produce solid NpF_4 and BrF_3 (see Progress Report for November 1966, ANL-7279, pp. 69-70). It was observed that the reaction proceeded by an initial period at a high rate followed by a period at a lower reaction rate. This observation suggested that a secondary reaction between NpF_6 and BrF_3 to form BrF_5 , as mentioned in the literature,⁹ might be occurring. This behavior was evaluated in two experiments: in the first the total pressure of a gaseous mixture of BrF_3 and NpF_6 was measured over a 2-hr period, and in the second the infrared absorption of a BrF_3 - NpF_6 mixture was followed for 2 hr. In neither case was any evidence for the formation of BrF_5 observed; no change in pressure occurred, and there was no absorption at 644 cm^{-1} , where BrF_5 shows characteristic absorption.

Since the previously reported reactions of NpF_6 with bromine were performed with NpF_6 partially in the solid state, it is suggested that the change in reaction rate observed might have been due to the formation of solid NpF_4 at the interface between solid NpF_6 and the liquid phase, which would establish a diffusion barrier towards further reaction.

(iii) Separation of Neptunium from Plutonium. Thermal decomposition of PuF_6 to solid PuF_4 is being considered as a technique for separating plutonium from mixtures containing UF_6 and other fluorides. Since neptunium may be present as NpF_6 in these mixtures, the behavior of NpF_6 during thermal decomposition of PuF_6 must be determined.

Two experiments were performed in which gaseous mixtures of NpF_6 and PuF_6 were circulated through a vessel at 300°C . Each experiment was interrupted at hourly intervals to measure the quantity of solids deposited and to obtain samples for analysis. It was observed that

⁹Malm, J. G., Weinstock, B., and Weaver, E. E., J. Phys. Chem. 62, 1506 (1958).

a small fraction of the neptunium ($\sim 0.2\%$) codeposited with the plutonium. The concentration of the neptunium in the deposited solids was constant throughout the entire deposition experiment. The fraction of the total neptunium in the gas mixture that codeposited with the plutonium was found to be dependent on the total quantity of plutonium deposited. The results suggest that selective thermal decomposition may be a feasible method for separating NpF_6 from PuF_6 .

b. Recovery of Uranium and Plutonium from Low-enrichment Fuels: Engineering Work. Engineering development work is primarily directed toward the demonstration of various process steps in the fluoride volatility processing of oxide fuel using bromine pentafluoride as the fluorinating agent. The processing steps are shown in Fig. 10. In the process, Zircaloy-clad fuel is immersed in a fluid bed of refractory alumina and then contacted with hydrogen chloride at 350°C to remove the cladding as a volatile chloride. Since the direct reaction of sintered, massive oxide fuel with BrF_5 does not proceed readily, the oxide fuel is pulverized to powder by reaction with oxygen prior to fluorination. Recovery of uranium is effected by selective fluorination (Fluorination I) of uranium to UF_6 using 12 v/o BrF_5 at 300°C , while plutonium is converted to non-volatile PuF_4 and remains in the fluid bed. The UF_6 is collected in a condenser with excess BrF_5 and with the reaction product bromine. These materials, together with those fission product fluorides that are volatile, are separated by fractional distillation, resulting in a completely decontaminated UF_6 product. The bromine, with some BrF_3 , is refluorinated to BrF_5 by reaction with fluorine, and the BrF_5 is recycled.

Fig. 10. Interhalogen Flowsheet for Reprocessing of UO_2 Fuel Material Using Bromine Pentafluoride

In the second fluorination step (Fluorination II), plutonium is recovered as PuF_6 by fluorination of the PuF_4 with fluorine gas at 300 to 550°C.

Engineering-scale studies currently under way include:

(1) determining the effects of process variables on the fluorination of oxidized UO_2 with BrF_5 in a 3-in.-dia fluid-bed reactor; (2) investigating the behavior of PuF_6 in process equipment (engineering-scale alpha facility) during fluorination of PuF_4 with fluorine; and (3) determining the distribution of fission products during the processing of highly irradiated UO_2 fuel in a $1\frac{1}{2}$ -in.-dia fluid-bed reactor.

c. Engineering-scale Alpha Facility. The major objectives of experiments conducted in the engineering-scale alpha facility are to demonstrate handling of PuF_6 on an engineering scale and to determine the feasibility of fluoride volatility flowsheets. The facility comprises two large alpha boxes: one contains process equipment, the other scrubbers and auxiliary equipment. The process equipment includes a fluid-bed fluorinator for fluorinating mixed oxide fuel to UF_6 and PuF_6 , a converter reactor for converting PuF_6 to PuF_4 by thermal decomposition or for converting mixed UF_6 and PuF_6 to mixed oxide, and a system of condensers and chemical traps for collecting hexafluoride products.

Recent experiments in the facility have involved the fluorination of PuF_4 powder in a fluidized bed of alumina to simulate the Fluorination II step of the interhalogen flowsheet (Fig. 10). In each of these experiments (Runs Pu-10, -11, and -13) approximately 135 g of PuF_4 in a bed of alumina (6,700 g) was fluorinated with fluorine at 200 to 550°C. In Run Pu-10, 41.5 g UF_4 and 41.8 g CsF (simulating fission product cesium) were added to the bed; in Run Pu-11, 39.2 g of CsF were added. No cesium was added in Run Pu-13. The fluorination times were 14 hr in Run Pu-10, 52 hr in Run Pu-11, and 29 hr in Run Pu-13. Analyses of bed samples taken periodically throughout the runs indicated that the rate of plutonium removal by fluorination was less in runs in which CsF was present, Runs Pu-10 and Pu-11, compared to runs in which no CsF was present, Pu-13 and Pu-6 (see Progress Report for July 1966, ANL-7245, p. 47). The concentration of plutonium in the alumina beds after completion of the fluorination was higher (~0.10 w/o) in Runs Pu-10 and Pu-11 than in Run Pu-13 (0.004 w/o).

After Run Pu-13, while the alumina bed was still in the fluorinator, the entire system was fluorinated for 17.5 hr to remove any plutonium from process vessels and lines. In this fluorination period, the bed was maintained at 550°C, the sintered metal filters at 300°C, and process lines at 300°C. It was observed that the plutonium concentration in the bed did not decrease as a result of the additional exposure of the bed to fluorine at 550°C.

Run Pu-14 was performed in the alpha facility with feed material generated in experiments that had been carried out previously in the laboratory-scale fluid-bed facility. This material represented the fluid bed after the uranium had been recovered by fluorination of oxidized UO_2 - PuO_2 -fission product pellets with BrF_5 . The charge to the fluorinator consisted of 6400 g of the mixture which contained 25.7 g of plutonium and 3 g of CsF . This material was contacted with 80 v/o fluorine for 9.7 hr while the bed temperature was increased from 300 to 550°C, and for 5.2 hr at 550°C. The final bed after fluorination contained approximately 0.01 w/o plutonium which represents the removal of about 98% of the plutonium. An additional clean-up fluorination step for 10 hr at 550°C did not result in the fluorination of additional plutonium. This high plutonium removal suggests that the effect of CsF on plutonium retention may be less marked in the presence of composite fission products.

Inspection of the fluid-bed reactor after Run Pu-11 revealed that the two sintered metal filters in the disengaging zone of the fluorinator were severely corroded. The temperature of the filter zone during the major portion of the fluorination is maintained at 125°C. However, near the end of the fluorination step, while the bed temperature is 550°C, the filter temperature is raised to 300°C to assure fluorination of any plutonium-containing fines in the vicinity of the filters. The filters used in these experiments are fabricated from sintered Monel powder. It is believed that the corrosion of the filters resulted from exposure to fluorine at temperatures above 300°C (e.g., at 375°C for a brief period). Both filters showed evidence of a nickel-fluorine reaction, and both filters were replaced after Run Pu-11. Filters composed of sintered nickel fibers have recently become commercially available. These filters are known to have good corrosion resistance--superior to sintered Monel--under these fluorination conditions. Filters of this type have been ordered for future use.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. General

The architect-engineer, Burns and Roe, Inc., has completed the final draft of the specifications for Construction Package No. 1, primary pumps, and primary heat exchangers. These specifications are being reviewed by the project staff and the AEC.

Meetings were held with containment-structure contractors and the architect to discuss the containment liner and erection process. Based upon the information derived from these meetings a construction sequence was evolved for the reactor containment building.

The architect has started work on the outside utilities and the re-vamp of the 549A substation. Authorization has been given for the start of detailed architectural and structural design of the laboratory and office building and of the reactor containment building.

A final draft of the report on the computer control study by Burns and Roe has been received and is presently being reviewed by the project instrument and control group.

2. Studies of Materials Compatibility and Corrosion

Progress and results of this work, conducted at ORNL, have been reported regularly.¹⁰ The experimental portion was completed in July 1966 and the final report issued in November 1966.¹¹ The following is an abstract from this report.

The purpose of the studies was to investigate the potential of corrosion to be expected in the multimaterial primary loop. This circuit includes the beryllium reflector, the aluminum beam tubes, and the stainless steel fuel element cladding,¹² all of which are exposed to the high-purity primary coolant. All tests were consequently conducted in deionized water with a specific resistivity of 10^6 ohm-cm or greater.

The beryllium exposure was made in a pump loop with water flowing past the specimen at 44 fps. The effect of exposed aluminum upon the

¹⁰ Reactor Development Progress Reports ANL-7082, 7090, 7105, 7115, 7122, 7132, 7152, 7176, 7193, 7204, 7219, and 7245.

¹¹ Griess, J. C., and English, J. L., Materials Compatibility and Corrosion Studies for the Argonne Advanced Research Reactor, ORNL-4034 (Nov 1966).

¹² The AARR will utilize aluminum cores (HFIR) in lieu of the initially planned stainless steel fuel.

corrosion of beryllium in the loop was also investigated. Although the corrosion of beryllium showed a marked effect on the ratio of material surface to water volume, neither crevices nor contact with stainless steel or aluminum produced adverse effects.

The direct effect upon beryllium corrosion of aluminum corroding in the same loop is not clear.

A few randomly distributed pits were present on most of the beryllium specimens, the deepest being 3.2 mils. Except for these few pits, the attack was uniform. The ratio of beryllium surface to volume of water in the AARR will place the corrosion to be expected in the range between 1.1 and 2.8 mpy, either of which would be acceptable.

Aluminum corrosion under conditions expected to exist at the tips of the 6061-T6 aluminum beam tubes with different cooling-water temperature and different heat fluxes was studied. Two tests were performed (see ANL-7176 and ANL-7219). The first test, designed to simulate conditions with 135°F cooling water of a velocity of 10 fps, resulted in the formation of a 3-mil-thick corrosion product layer in 75 days. The temperature drop across the corrosion product layer was 150°F. A thermal cycle caused part of the corrosion product to spall from the surface, but on continuation of the test the corrosion product reformed at about the original rate. The heat flux was 820,000 Btu/hr-ft² and the water-oxide interface temperature was 260°F.

The second test, simulating a 120°F cooling water inlet temperature was conducted under similar conditions except that the heat flux could be reduced to 400,000 Btu/hr-ft² in order to maintain the required interface temperature of 200°F. Under these conditions only about 0.65 mil of corrosion product formed during an 85-day exposure. These results indicate that if the surface temperature of the beam tube tips can be maintained at 200°F or less, the buildup rate of corrosion product will be low enough to prevent excessive temperatures in the beam-tube walls.

The effects of heat flux on the corrosion of stainless steel was tested with beryllium and aluminum exposed in the loop. The results indicate that Type 304 stainless steel will not develop appreciable deposits on the heat-transfer surfaces when exposed under thermal conditions similar to and even higher than those anticipated in the AARR.

At the conclusion of each of the two tests, the heat-transfer surfaces of the specimens were covered with a thin tarnish-like deposit. Metallographical examination revealed the presence of a film averaging about 0.1 mil in thickness. The development of this film was not observed in the temperature measurement across the interface, probably because of a high thermal conductivity of the film.

Special galvanic and crevice corrosion tests were conducted in an isothermal loop. Those tests lasted for an uninterrupted 3655 hr. Different combinations of stainless steel, aluminum, and beryllium were bolted together and exposed to deionized water at 200°F, and at high (44 fps) as well as very low (2 fpm) flow velocity.

Examination of the specimen at the completion of the test revealed nearly complete absence of any nonuniform attack either in the crevice or on the exposed part of the specimen. Some of the aluminum coupons showed a few isolated pits, the deepest of which was only 1.5 mils. One of the beryllium surfaces in contact with stainless steel showed a single pit, 0.8 mil deep. All other surfaces, whether exposed to high or low velocity, were free of localized attack.

No indication of a galvanic effect was found in any of the tests.

3. Steady-state Heat-transfer Program

In the AARR steady-state heat-transfer program, the effects of channel spacing, flow, temperature of inlet water, pressure and power upon pressure differential along the channel, void distribution, and critical heat flux are measured in electrically heated, rectangular test sections that simulate the flat- or involute-plate geometrical arrangement of the AARR core. The tests discussed here were run at approximately 655 psia.

With a 30-mil test section, it was found that the channel had distorted under pressure (see Progress Report for May 1966, ANL-7219, p. 59); the center spacing was about 50 mils, while the spacing at the corners remained about 30 mils. Tests were also run with a 50-mil test section. It is possible to calculate an equivalent channel spacing from the adiabatic pressure-drop data. The results are: for the 50-mil test section, the equivalent channel spacing is 51 mils; for the 30-mil test section, 47 mils. These values, 51 mils and 47 mils, were used in the analysis reported here.

In examining the test results, it was found that near burnout a considerable amount of steam evidently exists at the outlet, even though the temperature of the bulk liquid is slightly subcooled. This was checked for a burnout point at 100 kW, assuming a true heat balance of 98%. If the outlet thermocouple temperature (486°F) is assumed to represent the bulk-liquid temperature, and if the steam is assumed to exist at saturation temperature (496°F), then the flow quality is calculated to be 5.70%. If a slip of 1.5 be assumed, a steam volume fraction of 58.1% was estimated. The void fraction actually measured for this case by gamma attenuation was 60%.

In an alternative approach, the exit flow steam quality that would exist if equilibrium conditions prevailed was calculated as 4.2%. When this

result was used with the method devised by Levy¹³ to calculate the true non-equilibrium exit flow quality, a quality of 6.2% was obtained. If it be assumed again that a slip of 1.5 existed, a steam volume fraction of 60.4% was found. These calculations indicate good agreement with the experimental data.

The single-channel burnout points were compared with the additive or superposition type of critical heat-flux correlation proposed by Gambill.¹⁴ For the forced-convection contribution, the McAdams equation for the heat-transfer coefficient with a constant of 0.023 was used. For the pool-boiling contribution, the Zuber equation with the theoretical coefficient of $\pi/24$ was used. A value of 1.4 was used for the ratio maximum to average heat flux which occurs at the exit of the test section.

Table XVII shows a comparison of the experimental points with the correlation. The correlation predicts the results with deviations ranging from 4 to 26%. This is well within the 40% range claimed by Gambill.

TABLE XVII. Comparison of Predicted and Experimental Critical Heat Flux q'' (in Btu/hr-ft²)

	<u>51-mil Case</u>			
Power, kW	100	125	150	175
Correlation q'' ($\times 10^{-6}$)	1.99	2.18	2.34	2.48
Experimental q'' ($\times 10^{-6}$)	1.91	2.38	2.86	3.34
Ratio	1.05	0.92	0.82	0.74
	<u>47-mil Case</u>			
Power, kW	78	100	140	175
Correlation q'' ($\times 10^{-6}$)	1.80	2.02	2.27	2.55
Experimental q'' ($\times 10^{-6}$)	1.51	1.94	2.70	3.33
Ratio	1.19	1.04	0.84	0.77

For multiple-channel systems, the minimum of the curve of the pressure drop versus flow is important in predicting the onset of flow instability. The design approach used for the HFIR core during normal operation is to maintain cooling conditions that prevent incipient nucleate boiling as predicted by the Bergles-Rohsenow correlation.¹⁵ The experimental pressure-drop minima were therefore compared with the flow for incipient nucleate boiling as given by this correlation. Table XVIII shows the results. The ratios of flows and pressure drops shown may be thought of as a form of safety factor, in that they indicate the ratio by which the flow or pressure

¹³Levy, S., GEAP-5157 (April 1966).

¹⁴Gambill, W. R., Chem. Eng. Progress Symposium Series 59, 71-87 (1963).

¹⁵Bergles, A. E., and Rohsenow, W. M., Trans. ASME J. Heat Transfer, pp. 365-372 (Aug 1964).

drop at incipient nucleate boiling is greater than that for the onset of flow instability. It will be noticed that the ratios of pressure drop range from 1.84 to 1.99 for the 51-mil channel, but is only 1.33 to 1.53 for the 47-mil channel. This would seem to indicate that the pressure-drop safety factor is decreasing rapidly with decreasing channel spacing. However, the results for the 47-mil channel are less suitable for close analysis due to the greater scatter of the data, making it somewhat uncertain just where the minima occur.

TABLE XVIII. Comparison of Incipient Nucleate Boiling Flow and Pressure Drop with the Flow and Pressure Drop Corresponding to the Minimum of the Curve of Pressure Drop versus Flow

<u>51-mil Case</u>				
Power, kW	100	125	150	175
W_{inb} , lb/min	27.0	35.2	43.1	50.4
W_{mpd} , lb/min	15.2	19.3	23.8	28.0
Ratio	1.78	1.82	1.81	1.80
Δp_{inb} , psi	14.2	22.0	32.2	43.8
Δp_{mpd} , psi	7.7	11.7	16.8	22.0
Ratio	1.84	1.88	1.92	1.99
<u>47-mil Case</u>				
Power, kW	78	100	140	175
W_{inb} , lb/hr	1161	1547	2216	2824
W_{mpd} , lb/hr	640	880	1230	1468 ^a
Ratio	1.81	1.76	1.80	1.92 ^a
Δp_{inb} , psi	13.6	23.1	49.0	66.3
Δp_{mpd} , psi	9.9	17.4	32.0	43.8 ^a
Ratio	1.37	1.33	1.53	1.51 ^a

Subscripts

inb = incipient nucleate boiling

mpd = minimum of pressure drop versus flow curve.

^aRelated to single-channel burnout point, not minimum of pressure drop curve.

Another method of examining the flow instability is to consider the ratio of the exit temperature rise above the inlet temperature to the saturation temperature rise above the inlet temperature for the minima of the pressure drop versus flow curves. Table XIX gives these results. The values for the 51-mil channel range from 0.93 to 0.97 and appear reasonable based on previous studies using the computer code STDY-3.¹⁶ The values for the 47-mil channel are somewhat more uncertain because of the greater scatter of data.

¹⁶STDY-3, A Program for the Thermal Analysis of a Pressurized Water Nuclear Reactor during Steady-state Operation, WAPD-TM-213 (June 1960).

TABLE XIX. Tabulation of the Quantity r Corresponding to the Minimum of the Curve of Pressure Drop versus Flow

$$r = \frac{\text{Exit temperature minus inlet temperature}}{\text{Saturation temperature minus inlet temperature}}$$

51-mil Case				
Power, kW	100	125	150	175
r	0.972	0.956	0.934	0.928
47-mil Case				
Power, kW	78	100	140	175
r	1.00	0.97	0.97	1.00 ^a

^aRelated to single-channel burnout point, not minimum of pressure-drop curve.

It should be noted that in Tables XVIII and XIX the values for 175 kW in the 47-mil channel do not relate the single-channel burnout point to the minimum of the pressure-drop curve, since burnout occurred before a minimum pressure drop was established.

A comparison has also been made between the experimental pressure drop versus flow curves for heated test sections and the results as calculated with the computer code STDY-3. Figures 11 through 14 show the comparisons

Fig. 11. Comparison of Experimental and STDY-3 Curves of Pressure Drop vs. Flow for 50-mil Channel at 100 kW

Fig. 12. Comparison of Experimental and STDY-3 Curves of Pressure Drop vs. Flow for 50-mil Channel at 125 kW

Fig. 13. Comparison of Experimental and STDY-3 Curves of Pressure Drop vs. Flow for 50-mil Channel at 150 kW

Fig. 14. Comparison of Experimental and STDY-3 Curves of Pressure Drop vs. Flow for 50-mil Channel at 175 kW

for the 50-mil channel at power levels of 100, 125, 150, and 175 kW. Also shown are the flows for incipient nucleate boiling as obtained from STDY-3 and as calculated from the Bergles and Rohsenow correlation (for these curves, velocity in ft/sec = flow in lb/min times 0.815). The curves agree quite well.

In general, the behavior as predicted by STDY-3 is as follows: At high flows the curve of pressure drop versus flow is a straight line on the log-log graph, corresponding to the conventional behavior of a single-phase fluid. As the flow is reduced, local boiling begins and the pressure-drop curve deviates from the straight line and becomes more nearly horizontal. Finally, at low flow, bulk boiling begins and the pressure drop jumps up sharply, causing a minimum in the curve, at the point of local boiling. As discussed above, the true behavior in the experimental situation appears to be more complicated; a nonequilibrium condition exists in which, at low flow, considerable steam exists at the channel exit even though the liquid is still slightly subcooled. However, the experimental and computed situations are sufficiently close so that the curves appear to agree fairly well.

Figure 15 shows the results for the 30-mil test section at 100 kW. This test section distorted under pressure, expanding to approximately 50 mils in the central region, as shown in Fig. 16. The equivalent channel spacing for this case, as determined from the adiabatic pressure-drop curve, was 47 mils. Use of 47 mils in STDY-3 gives the lower pressure-drop curve shown in Fig. 15. It can be seen that, as the flow is reduced, the experimental curve begins to flatten out considerably earlier than does this lower, computed curve. This fact can probably be attributed to the beginning of local boiling in the corner regions of the channel, which would of course receive less flow than the central section, since the corners remained near 30 mils while the central section expanded to 50 mils. If a channel spacing of 30 mils is used in STDY-3, the upper curve in Fig. 15

Fig. 15. Comparison of Experimental and Calculated Values of Pressure Drop vs. Flow for 30-mil (nominal) Channel Operating at 100 kW

Fig. 16. Schematic of 30-mil Flow Channel Showing Expansion to Approximately 50 mils at Center Section

is obtained. It can be seen that for the flows at which boiling is taking place, the experimental results might be thought of as being composed of an average or superposition of the upper and lower computed curves, since a portion of the channel area is behaving according to each of the two curves. The upper curve would at least be conservative in guiding reactor design for

Fig. 17. Plan View of Hypothetical Configuration Showing Narrowing of Channel Spacing

a hypothetically deformed channel spacing such as shown in Fig. 17. If a pressure drop corresponding to incipient nucleate boiling on the upper curve were applied across the reactor, it would produce sufficient flow to prevent failure via flow instability on the experimental curve. It should be noted, however, that in general the smaller the channel spacing, the closer will the flow for incipient nucleate boiling be to the flow for

flow instability if the entire channel is at the small channel spacing. In other words, the safety factor associated with operating at incipient nucleate boiling is reduced if the channel spacing is very small.

4. Physics Experiments and Analysis

Analyses of the reactor physics characteristics of the AARR with HFIR-type fuel elements are continuing to supply important results of effects of perturbations, e.g., void coefficients of reactivity in the core and in the ITC. It has become increasingly apparent that more detail is required in the specification of region compositions, especially the removable portions of the beryllium reflector, to determine the absolute reactivity status. Such detail is not necessary, however, to determine the reactivity effects of the perturbations studied to date. Nevertheless, the specifications of all region dimensions and material compositions are being reviewed in fine detail.

Computations of the reactivity gain which would occur in the event of voidings of ITC water have consistently underestimated the measured reactivity gain. Detailed void-coefficient experiments have been reported previously (see Progress Reports for November 1965, ANL-7122, pp. 73-74; for December 1965, ANL-7132, pp. 55-56; for August 1966, ANL-7249, p. 69). These experiments involved ITC's of two different radial dimensions, with heavily loaded fuel zones (SS-U²³⁵ fuel plates). It is believed that the loading of the fuel zone has only a second-order effect on the reactivity coefficient of voiding of ITC water, and analysis of these systems therefore has direct relevance to the ITC void coefficients in the AARR. Two-dimensional analyses of the 1215/1620 loading, with the small ITC, have confirmed that the one-dimensional calculations underestimated the reactivity coefficients

of ITC voidings principally because of the assumption that the effective bare-region height was unaffected by the voiding. The earlier one-dimensional calculations implied a reactivity change of 1.1% for 50% void of the 18-in.-high active portion of the 100%-H₂O ITC. The two-dimensional calculations yielded a reactivity gain of 1.50% for a 50% void, in excellent agreement with the measured value of 1.47% for a 49% void. The two-dimensional problems were solved with the 2-D CANDID program on the CDC-3600 computer, in 6-group diffusion theory.

The N¹⁶ production rate in the operating reactor has been calculated. Approximately 1% of the fission-spectrum neutrons born with energy above 11 MeV are captured by O¹⁶ to produce N¹⁶ by the O¹⁶(n,p)N¹⁶ reaction. The principal uncertainty in the analysis is the density of fission neutrons in this energy range. Rather than the Watt fitting to the fission spectrum,¹⁷ the more conservative formula $f(E) = a\sqrt{E} \exp(-E/T)$ has been used.¹⁸ At 11 MeV, the neutron density is roughly 3 times as large as the value implied by the Watt formula. This is a reflection of the uncertainty in the measurements of the fission spectrum above 10 MeV.

The production rate of O¹⁹, from O¹⁸(n,γ)O¹⁹, has been shown to be unimportant by comparison with the N¹⁶ production. The O¹⁹ isotope has a half-life which is 4 times that of the N¹⁶, but the N¹⁶ activity dominates even after one minute of decay, which is the cycle time for the primary coolant flow.

¹⁷Reactor Physics Constants, ANL-5800 (First Edition), pp. 16-20.

¹⁸Reactor Physics Constants, ANL-5800 (Second Edition), July 1963, Table 1-10, p. 14.

V. NUCLEAR SAFETY

A. Coolant Dynamics

1. Coolant (Water) Expulsion Studies

By means of a digital computer, all data for the runs in which complete expulsion occurred has been reduced. Analysis of this data has commenced.

Work on the data from the runs in which oscillatory flow resulted is still being programmed by the Applied Math Division.

2. Superheat Experiments

This experiment is designed to measure the degree of liquid superheat required to initiate nucleate boiling in sodium under various conditions simulating a reactor environment. Typical parameters will be systematically varied to determine their independent and combined effects upon the liquid superheat necessary to initiate nucleation. The parameters to be examined are: (a) pressure, (b) dissolved-gas content, (c) heat flux, (d) surface characteristics, and (e) the pressure-temperature history of the system.

Approximately 100 test runs have been completed. The superheat test runs to date include: (a) pressures ranging up to one atmosphere, (b) heat fluxes ranging to approximately 0.3×10^5 Btu/hr-ft², (c) liquid superheats required to initiate boiling, ranging up to approximately 200°F, (d) steady-state superheats ranging from 20 to 40°F, and (e) transient pressure pulses ranging up to 60 psia. Liquid sodium temperatures ranging up to approximately 1800°F have been obtained.

3. Studies of Critical Flow

Calculations of the progression of accidents in sodium-cooled reactors reveal that critical flow may exist in the core, leading to detrimental effects such as voidage of the coolant channel, shock phenomena, and pressure buildup.

The construction of equipment to study critical flow of different fluids is continuing. The sodium loop has been completed, and the first test section (ID = 1/16 in. and L/D = 100) has been installed in the loop. The loop has been filled with sodium. Final calibration of the instrumentation is being completed.

A technique has been developed which definitely shows the existence of metastability in high-velocity, flashing, two-phase flow. The analysis is outlined briefly below.

Three assumptions are made:

- a. one-dimensional analysis;
- b. a different average velocity for each phase;
- c. no viscous dissipation; therefore the acceleration of the fluid corresponds to the total pressure drop from the initial point of flashing.

At small qualities ($x < 0.01$), the above assumptions lead to an equation for the absolute minimum average vapor velocity which may exist under thermodynamic equilibrium conditions:

$$u_{g\text{MIN},E} = \frac{x_E v_g G [G^2 v_\ell + g_c (P_1 - P_2)]}{(P_1 - P_2) g_c}, \quad (1)$$

where

$P_1 = P_{\text{SAT}}$ is the pressure at the initial point of flashing;

P_2 = pressure at the exit of the test section;

G = mass flowrate;

x = quality;

v_g = specific volume of the vapor;

v_ℓ = specific volume of the liquid;

g_c = Newton's Constant;

u_g = vapor velocity;

h_g = enthalpy of the vapor;

h_f = enthalpy of the liquid;

s_g = entropy of the vapor;

s_f = entropy of the liquid;

J = unit conversion constant.

If viscous dissipation is included, larger values of the vapor velocity will result if thermodynamic equilibrium is maintained.

However, the maximum possible vapor velocity that can be established may be calculated by assuming:

- a. thermodynamic equilibrium consistent with the above analysis;
- b. the vapor expands alone; saturated vapor expands isentropically from P_1 to P_2 .

These assumptions lead to the following equation:

$$u_{gMAX,E} = \sqrt{2g_c J \Delta H}, \quad (2)$$

where

$$\Delta H = h_{g,1} - [h_{f,2} + x_s(h_{g,2} - h_{f,2})];$$

$$x_s = \frac{s_{g,1} - s_{f,2}}{s_{g,2} - s_{f,2}}.$$

Three steam-water critical flow runs from this research have been examined with respect to the above analysis. The results shown in Table XX clearly indicate that when thermodynamic equilibrium is assumed, the two-phase vapor velocity given by Eq. (1) is considerably higher than that calculated through Eq. (2) assuming an ideal vapor expansion. It can be argued that since the velocity given by Eq. (2) is the maximum velocity the vapor can acquire, some quantity in Eq. (1) must be in error. The equilibrium quality is the only calculated quantity in Eq. (1); hence, the actual quality must be considerably less than X_E . For example, in Run 2, the actual quality can be no larger than one-sixth of the equilibrium quality.

TABLE XX. Analyses of Three Steam-Water Critical Flows

Run	G (lb _m /sec-ft ²)	P ₁ (psia)	P ₂ (psia)	X _E	u _{gMIN,E} (ft/sec)	u _{gMAX,E} (ft/sec)
1	3672	55.1	49.4	0.0046	1450	793
2	3917	39.8	37.3	0.0032	3333	555
3	3503	35.2	32.5	0.0039	3107	555

The above method definitely shows the existence of metastability, but only in a qualitative manner.

4. Effect of Rapid Heating of Reactor Components

The mathematical model of the parallel-plate coolant-structure system (see Progress Report for October 1966, ANL-7267, p. 75) being used in the preliminary study of the influence of fluid-structure coupling on the pressure pulse and coolant expulsion has been simulated on the digital computer. The computer program was recently "debugged" and two runs have been made.

For the preliminary computation, the coolant was assumed to be water at a temperature of 212°F (no subcooling). The channel cross section is 2.5 x 0.5 in., with a heated length of 20 in. The channel walls are 2.5 in. wide and 20 mils thick. The input was assumed to be a rectangular power burst of 100-kW peak power with a duration of 80 msec. The mathematical computation of such an assembly under such conditions gives a fundamental natural frequency of 697 cps. The results of one simulation are given in Fig. 18.

Fig. 18. Model Coolant-structure Response to Rectangular Power Burst

Because the period of vibration of the walls is 1.43 msec (697 cps), which is much shorter than the input period, the response of the wall follows that of the pressure. At the peak pressure of 21.2 psia (1.44 atm),

the maximum displacement of the channel walls is 30.5 mils, and the expulsion height of the water is 2.75 in.

For the large channel cross-sectional area of this system, the change in vapor volume, contributed by wall displacements of less than 30 mils, is negligible, and hence the structure would be expected to have little influence on the pressure transient and coolant expulsion.

Future runs will involve channel areas typical of those found in reactors and situations in which the structural period is of the order or greater than the input period.

B. Fuel Meltdown Studies with TREAT

1. Data from Transient 1048

TREAT reactor transients 1047 and 1048 were performed using a seven-pin cluster (one 9% enriched pin surrounded by six 7% enriched pins) of EBR-II Mark-I pins on the standard EBR-II pitch (see Progress Report for September 1966, ANL-7255, p. 83). Transient 1047 was a low-energy, check run, whereas Tr 1048 was a full-scale experiment producing gross sample failure and movements of the sodium coolant. Transient data from Tr 1048 are shown in Fig. 19. Inlet flow was measured with an electromagnetic flowmeter. The pressure transducer is located at the test-section inlet. The initial rate of coolant flow was about 3.25 m/sec through the test section. Because the pump is of the alternating-current electromagnetic type, a 120-cps pressure signal is impressed on the inlet pressure transducer during normal operation.

The temperature of the outlet thermocouple rises smoothly as energy is released in the sample cluster, until approximately 600°C is reached, coinciding with a small pressure anomaly. A pressure spike of about 3.5 atm occurs at about 2.0 sec, correlated with an abrupt deceleration of coolant, which produces a maximum coolant expulsion rate of about 5 m/sec. The time delay between the pressure spike and the beginning of the temperature response of the inlet thermocouple is about 0.068 sec. This may be compared with an estimated transit time of 0.066 sec for coolant (moving at the rates recorded by the inlet flowmeter) to travel from the center of the cluster to the inlet thermocouple.

If the sharp rises of both thermocouples between 2.0 and 2.1 sec are assumed to result from a step change in temperature of coolant environment, a thermocouple response time of about 0.020 sec can be deduced from the data. A time response of about 0.021 sec has been calculated for these thermocouples. The higher (about 5 atm) secondary pressure spike does not have a direct correlation like the first. This second spike may be associated with upward expulsion of coolant followed by fall back on hot

Fig. 19. Data Recorded for a Cluster of Seven Fuel Pins during Transient 1048 in the TREAT Facility

fuel, since the time between the first and second spikes is approximately 0.1 sec, time for an object to fall free over 5 cm. However, the test section may have been largely voided of sodium at that time, as approximately 0.05 sec later, the 120-cps pump pressure signal vanishes from the inlet pressure transducer, suggesting that a sodium void has reached the transducer location.

Some flow recovery has been recorded on the flowmeter (located 7 cm below the pressure transducer) during the absence of the 120-cps signal, which is terminated by a pressure spike of about 2 atm. A second period of absence of the 120-cps signal occurs again before sodium flow is re-established at a low rate. The apparent zero shift of the pressure transducer during the transient is in good agreement with the semi-quantitative analysis of response of this instrument to a TREAT transient of the magnitude of Tr 1048.

2. Examination of Loop Test Section after Transient 1048

The test section from Integral Loop No. 1, which was used for Tr 1048, was inspected in the ANL Hot Laboratory. Frozen fuel was seen at the top end of the fuel holder (see Fig. 20). Movement of this fuel to the outlet of the test section was vigorous. Figure 21 shows that appreciable

Fig. 20. View of the Top of the Fuel Holder Showing Deposits of Frozen Fuel Resulting from Transient 1048 Test of a Cluster of Seven Fuel Pins

Fig. 21. View of Hold-down Fitting Which Was Located above the Test Section and Shows Deposits of Frozen Fuel Resulting from Transient 1048 Test of a Cluster of Seven Fuel Pins

fuel was transported in the molten state to the openings of the hold-down fitting above the test section. The frozen fuel in the openings was about one cm above the top of the holder shown in Fig. 20. The sharp rise in outlet thermocouple temperature to about 1175°C at about 2.7 sec in Fig. 19 may represent the time of movement of this fuel. Additional fuel was found frozen at the inlet to the test section. Resolution of the sequence and timing of axial fuel movements must await analysis of the fast-neutron-hodoscope record of the experiment.

It was necessary to split the fuel holder longitudinally with a milling machine to inspect the fuel remains. Complete failure was found after the holder had been opened. Only the extreme top and bottom ends of the cladding tubes remained identifiable. A small quantity of fuel remained in some of the cladding at the bottom. Fuel deposits were more pronounced toward the bottom of the test section. Most of the visible fuel was found frozen onto the inner wall of the fuel-holder pipe. All signs of flow indicated that molten fuel movement after it struck the pipe was downward. Some evidence was found for porosity of the fuel frozen at the bottom of the test section. A sample has been taken for metallographic examination in the ANL Hot Laboratory.

3. Development of Mark-II Integral Sodium Loop Pump

A Mark-II integral sodium loop is under development for use in safety experiments to be performed in TREAT. The annular linear induction pump for the loop has been designed; shop fabrication has begun on the pump alternating current coils, pole piece laminations, and pump bodies.

The pumps are to be tested on a pump test facility (designed for general-purpose use) which is now ready for filling with sodium. The entire test loop is mounted on a balance so that mass flow from a supply tank to a dump tank can be measured. It can be run either in the once-through mode of operation or in the recirculating mode. A pressure transducer is mounted on both sides of the test-pump location so that simultaneous measurements can be made of flow and pump pressure. Loop flow can be adjusted by control valves. Additional instrumentation consists of thermocouples and a permanent magnet, direct-current electromagnetic flowmeter. Test flow meters can be installed in series with the loop flowmeter for calibration. The loop also has a small auxiliary pump, an alternating-current electromagnetic pump with a capacity of about 4 gpm at about a half-atmosphere of pressure.

C. Accident Analyses

There has existed for sometime a growing need to improve ability to analyze accident behavior of fast reactors. The nature of the problem has been changing, and the capability of the modern computer to execute

larger computations has grown rapidly. A major effort in this direction was recently established at ANL in terms of the ARC (Argonne Reactor Computing) program development (see Progress Report for September 1966, ANL-7255, pp. 39-41). The quasistative excursion code QX1 will be incorporated into the Accident Analysis package as an integral part of the ARC system.

1. Safety Analysis Computing System

a. Heat Transfer. The steady-state heat transfer routine (see Progress Report for November 1966, ANL-7279, pp. 46-47) is now essentially complete. It includes a two-level implicit scheme (or Crank-Nicolson Scheme)¹⁹ which is used to compute the radial temperature profile (at any axial increment) at the end of $(k+1)$ th time increment from the knowledge of the various quantities at the end of k th increment. There are two major aspects in which the present scheme differs from the standard C. N. Scheme:

1) The radial temperature profile is predicted for (or extrapolated to) the middle of the $(k+1)$ th time interval from the profiles at the end of $(k-1)$ th and k th time intervals. Thermal properties are evaluated for this predicted profile for use in the computation of temperatures at the end of $(k+1)$ th time interval. (See Ref. 1.) It is found that this approach is adequate to take into account the nonlinearity of the heat conduction equations and that the method does not suffer from any numerical stability limitations.

2) Radial spatial increments are not kept uniform. This is done primarily to consider the unequal thermal expansions in the radial increments.

The implicit method suggested by Back in Ref. 2 is used to obtain equations relating the fuel surface temperature and the average clad temperature.

Gauss elimination procedure is used to solve the (tridiagonal) matrix equation resulting from the implicit scheme.

Thermal expansion is considered in an "open loop" sense, in that the change in the dimensions are computed directly from the change in temperatures in any time interval. Thermal expansions may or may not be constrained. If constrained, the module for the Elastic-Plastic Deformations is used to obtain the varying dimensions. The code is capable of handling fuel melting.

¹⁹Back, L. H., Trans. ASME, J. Heat Transfer, p. 89 (Feb 1962).

b. Linking of the Heat Transfer and the Two-phase Coolant Flow Modules. Linking of the Heat Transfer and the Two-phase Coolant Flow Modules is in progress. Linking is being done in an overlay structure in accordance with the specifications of the ARC system.

The two modules are coupled by the heat flux between the clad surface and the coolant. For each axial segment j , the average heat flux $\phi_j^{k+1/2}$ for the time interval $(k+1)$ is projected from the heat flux at the end of the intervals $(k-1)$ and k . By means of this projected heat flux the Two-phase Flow module computes the coolant temperatures $T_{c_j}^{k+1}$ and communicates with the Heat Transfer Module, which in turn computes all the fuel and clad temperatures at the end of the $(k+1)$ th interval. Heat flux at the end of the $(k+1)$ st time interval is now computed from knowledge of these temperatures.

The common area in the overlay is expected to contain only the primary communication variables, namely, the projected heat flux $\phi_j^{k+1/2}$, the coolant temperatures $T_{c_j}^{k+1}$, and the time interval, Δt^{k+1} .

2. Quasistatic Excursion Code QX1

Testing of the QX1 (see Progress Report for October 1966, ANL-7267, pp. 52-53) is in progress. Figure 22 shows the space-time dependence of the fast group of a two-group thermal reactor test problem as plotted by the CDC-160A computer system. The material constants, dimensions, and ramp rate were the same as given²⁰ for the delayed critical excursion in the 240-cm core, with two exceptions. All transverse bucklings were reduced by the factor 0.9994528 to make the system critical. Also, the thermal-group velocity was 2.2×10^5 cm/sec instead of infinite, as was the case for the problem given in the reference. The QX1 fluxes at 0.8 sec were +0.27% at the larger maximum and +0.01% at the smaller maximum (as shown in Fig. 22) relative to a WIGLE²¹ run of the same problem. These same differences were present in the thermal-energy group.

Further testing and feedback routine coding are being done in conjunction with the design of the ARC version of the system.

²⁰Yasinsky, J. B., and Henry, A. F., Some Numerical Experiments Concerning Space-time Reactor Kinetics Behavior, Nucl. Sci. Eng. 22, 171-181 (1965).

²¹Cadwell, W. R., Henry, A. F., Vigilotti, A. J., WIGLE--A Program for the Solution of the Two-group Space-time Diffusion Equations in Slab Geometry, WAPD-TM-416 (Jan 1964).

Fig. 22. Space-time Dependence of Fast Flux for the Yasinsky-Henry Delayed Critical Excursion Test Problem in the 240-cm Core, as Calculated by QX1

D. TREAT

1. Operations

A sample of prototype Power Burst Facility fuel was subjected to ten transient irradiations for the Phillips Petroleum Company.

Two preirradiated UO_2 samples were irradiated for ORNL to obtain data on release of fission products.

A stainless steel-clad UO_2 pin (RP-48-5) that had been irradiated to 6 a/o burnup was subjected to two transient irradiations in a transparent capsule. Photographic data from these transient tests will be compared with similar data obtained previously using unirradiated pins.

E. Chemical and Associated Energy Problems (Thermal)

1. Metal-Water Reactions

a. Loss-of-coolant Simulation Experiments in TREAT on Zircaloy-2-clad, UO₂-core Fuel Clusters. Experiments were continued in TREAT to investigate the safety problems associated with the behavior of Zircaloy-2-clad, UO₂-core fuel rods during a loss-of-coolant incident (see Progress Report for February 1966, ANL-7176, p. 90). The purpose of the current tests is to obtain information on the chemical and physical changes which occur upon meltdown of the fuel cluster. Specific emphasis is placed on determining the extent of metal-water reaction, the temperatures and pressures produced, and the final distribution of particle size of the fragmented fuel. Three experiments have been performed, each on a progressively longer time scale. The first of these, CEN-217S, was reported previously (ANL-7176, p. 90).

The basic experimental technique consists of loading a "scale-up" autoclave with a fuel-rod cluster and water, and exposing the entire assembly to a neutron flux in TREAT. The construction of the autoclave, which is essentially an instrumented stainless steel pressure vessel, was previously described (see Progress Report for November 1964, ANL-6977, p. 83). However, in these simulated loss-of-coolant experiments, three fuel rods were used in each experiment and were located above the pool of water (instead of being submerged in it). Each fuel rod consisted of 10 sintered, fired UO₂ pellets clad with Zircaloy-2. The fission heat generated in the 10% enriched UO₂ during the TREAT transient caused meltdown of the fuel rods. To achieve a more realistic simulation of a loss-of-coolant incident, the TREAT reactor power level, and hence the neutron flux, was held as constant as possible during the experiments. These irradiations are therefore called "flat-top" transients, in contrast to the power excursion type which were studied previously in this program (see, for example, Progress Report for September 1965, ANL-7105, p. 81).

The arrangement of the fuel cluster and the support structure is shown in Fig. 23. The lower part of Fig. 23 shows a "Calrod" heater (bent into a loop to fit inside the autoclave) which is actually submerged in the water when the whole assembly is inserted into the autoclave. In two of the experiments this electrical heater raised the water temperature from the ambient 30°C to a value of 100°C just before the TREAT transient was initiated. Thus, in these experiments, a 1-atm steam environment was provided for the fuel rods, which are located immediately above the water level. A tungsten-rhenium thermocouple is spotwelded to the cladding in the central portion of one of the fuel rods. A second thermocouple with a junction about 3/4 in. above the bottom (see Fig. 23) records the bulk water temperature and indicates when the hot fragments from the fuel cluster above have fallen down into the water.

Fig. 23. Arrangement of Fuel Cluster for Loss-of-coolant Simulation Experiments in TREAT

Results from the three experiments which have been completed are summarized in Table XXI. The runs are arranged from the shortest time of fission heating (12 sec) to the longest (50 sec). (This is the time during which the reactor power is on.) However, in the third test the actual event continued for about 2 min. During the first minute, the nuclear heat generation occurred, and during the second minute the cooling process took place while the pressure in the autoclave was still rising towards its maximum value. The time span of about 2 min, even though quite short, approaches the times that are currently of interest in analyses of loss-of-coolant accidents. With decay heating, under reactor conditions, times from 10 to 15 min are generally of most interest.

One of the most significant features of the experiments is the appearance of the residues; portions of the cladding were present that retained the original cylindrical form of the cladding. These are particularly apparent in the residue from experiment CEN-217S (see ANL-7176, Fig. 28, p. 93), which had the fastest heating rate. Evidently, in this experiment, considerable quantities of molten UO_2 emptied from the Zircaloy cladding before the cladding melted. This occurred to a lesser degree in experiment CEN-220S and to a still lesser degree in experiment CEN-223S, where progressively lower rates of fission heating allowed more time for heat transfer between the UO_2 and the Zircaloy.

The best indication of the time of the failure of the fuel rods is given by the time at which the temperature indicated by the cladding thermocouple peaked. Little information, however, is apparently provided by the temperature value itself at the peak. Values of 2430, 1670, and 1570°C, respectively, were recorded for the three experiments. Table XXII gives the conditions of fuel at the time that the maximum temperature was recorded by the cladding thermocouple. It is evident from the table that sufficient fission energy had been generated at the probable time of failure to have at least partly melted the UO_2 . Another indication that the

TABLE XXI. Summary of Results of Loss-of-coolant Experiments in TREAT with Zircaloy-2-clad, UO₂-core Fuel Clusters^a

	Experiment Number		
	CEN-217S	CEN-220S	CEN-223S
Transient			
Location of Fuel Cluster	Above 30°C H ₂ O	Above 100°C H ₂ O	Above 100°C H ₂ O
Duration of Flat-top			
Transient, sec	12	32	50
Average Heat Generation Rate in UO ₂ , ^b kW/ft	40	20	11
Average Reactor Power Level, ^b MW	40	20	11
Fission Energy Input, cal/g UO ₂	560	770	630
Results			
Metal-Water Reaction, %	40	44	32
Peak Pressure Rise, psig	87	750	220
Maximum Rate of Pressure Rise, psig/sec	45	76	3
Final Appearance of UO ₂ Fuel	Complete destruction; fragments and particles; ~1/2 of cluster dropped into water	Complete destruction; fragments and particles; whole cluster dropped into water	Complete destruction; fragments and particles; whole cluster dropped into water
Average Particle Size of Residue (d _{sv}), ^c mils	91	83	-

^aTen sintered pellets of UO₂ (10% enriched in U²³⁵) per rod. Overall rod dimensions: 5.62 in. long by 0.42 in. in dia; 25-mil-thick cladding; 3-mil gap between core and cladding; helium bonded. Three rods with a weight of 254 g of UO₂ and 82 g of Zircaloy-2 per cluster; 750 g of water in autoclave.

^bThe identical values of heat-generation rate and reactor power level are coincidental.

^c $d_{sv} = \frac{1}{\sum \frac{w_i}{d_i}}$ where w_i = fraction by weight of particles with diameter d_i as determined by sieve

screen analysis.

TABLE XXII. Condition of Fuel (UO₂) at the Time of Recording of the Maximum Cladding Temperature in Loss-of-coolant Experiments in TREAT

Experiment Number	Average (Constant) Heat Generation Rate ^a (kW/ft)	Peak Recorded Cladding Temperature ^b (°C)	Energy Deposit in Fuel (cal/g UO ₂)	Adiabatic Fuel Temp (°C) and Physical State of Fuel
CEN-217S	40	2430	360	3300 (fuel fully melted)
CEN-220S	20	1670 ^c	240	2800 (fuel partly melted)
CEN-223S	11	1570	285	2850 (fuel fully melted)

^aA heat-generation rate of 1 kW/ft is typical of values of greatest interest in loss-of-coolant accident analyses. Such low values cannot presently be sustained in TREAT for a sufficient time to produce fuel failure.

^bThe apparent randomness of the indicated temperature may be associated with the formation of hot spots (and cold spots) which were noted in motion-picture studies.

^cThe cladding thermocouple may have broken prematurely, since the temperature trace was lost at this point.

time of failure of the fuel corresponds to the time of recording of the maximum cladding temperature is that this time coincides within about 1 sec in each experiment with the beginning of a significant pressure rise within the autoclave.

Considerable information regarding the nature and timing of the collapse can be obtained from the oscillograph records for the three experiments. In CEN-217S there was a very high heating rate; much of the UO_2 flowed from intact tubes of cladding; collapse into the water was probably completed in about 5 sec. The 5-sec value is the approximate time from the recording of the peak cladding temperature to the completions of the pressure rise and water temperature rise. It was only in CEN-217S that the stainless steel support structure did not completely fail. Evidently, the loss of the heat source, molten UO_2 , from above the support structure was rapid enough to preclude melting of the structure.

In CEN-220S there was a more moderate heating rate, and indications are that collapse required from 10 to 20 sec. The major indication of collapse was the erratic behavior of the water-temperature thermocouple about 10 sec after the time of recording of the peak cladding temperature. Autoclave pressure reached a peak value after another 10 sec had elapsed, suggesting that collapse had been completed.

In CEN-223S, the heating rate was 11 kW/ft for a time of 50 sec. Autoclave pressure, however, rose uniformly for about 130 sec. Indications of water temperature increase were noted during a major portion of this time. It is concluded from this that collapse of the fuel cluster was rather slow and continuous for much of the 130-sec time interval.

A very significant observation, based upon the particle-size distribution of the residues, is the absence of large quantities of very fine particles. This is a particularly encouraging result, since it suggests that molten UO_2 (and molten UO_2 -Zr-Zr O_2 mixtures) do not exhibit a marked tendency for spontaneous subdivision on being quenched in water. It is also noteworthy that the pressure-time traces showed no "spike" pressure rises, which suggests that no steam explosions or very violent boiling occurred as the molten fuel material entered the water pool, at least for the small-scale experiments performed in this study.

The total extent of metal-water reaction for the three experiments varied from 30 to 44%. A number of factors complicate any detailed numerical analysis of the metal-water reaction during the experiments. During the initial heating period, the pressure in CEN-217S was 20 psia helium and only about 0.5 psia of water vapor. In the other two experiments, there was about 15 psia of water vapor pressure in addition to 20 psia of helium. There was no forced-convection flow in any of the experiments; however, the nature of the assembly of fuel rods suggests that

a brisk circulation by natural convection would be established and that there would be considerable steam available for reaction, at least in CEN-220S and CEN-223S. The likelihood of steam being available above the support plate suggests that considerable reaction occurred prior to fuel collapse. The fact that vigorous fission heating continued after much of the fuel had fallen into the water suggests that a sustained reaction (supported by the high rate of fission heating) continued after collapse of fuel into the water. Considering that the entire quantity of fuel had melted and collapsed into the water, a reaction of only 30 to 44% is encouraging from an accident-analysis point of view. This result again provides evidence that, upon being quenched in water, Zircaloy-clad fuel is not susceptible to a self-sustained reaction to completion, so long as the cladding is not dispersed in the form of very fine molten droplets.

An analysis of energy sources and distribution was performed for the TREAT loss-of-coolant experiments, with the results summarized in Table XXIII. From the analysis it was concluded that from 20 to 26% of the total energy release was of chemical origin. The remainder was energy generated by fission. The overall thermal efficiency was the fraction of the total energy that was effective in heating and boiling water to reach the quasiequilibrium state existing at the time of peak autoclave pressure. The remaining energy had been dissipated to the autoclave walls. The efficiency varied from 40 to 70%.

TABLE XXIII. Energy Calculations for Simulated Loss-of-coolant Experiments in TREAT

	CEN-TREAT Transient Number		
	217S	220S	223S
<u>Nuclear Source:</u>			
Specific fission energy input, cal/g UO ₂	560	770	630
Total nuclear energy input, cal	142,000	195,000	160,000
<u>Chemical Source:</u>			
H ₂ evolved, STP, liters	16.1	17.7	12.9
Heat evolved from Zr-H ₂ O reaction, cal	51,000	56,000	41,000
<u>Total Energy Release:</u>			
Nuclear and chemical, cal	193,000	251,000	201,000
Chemical contribution to total, %	26	22	20
Specific energy input to water, cal/g H ₂ O	257	334	268
<u>Calculated Final Conditions at "Equilibrium"^a</u>			
Water temperature, °C	117	257	184
H ₂ O vapor pressure, psia	26	648	159
H ₂ partial pressure, psia	55	86	52
He partial pressure, psia	26	35	30
Total "autoclave" pressure, psia	107	769	241
<u>Observed Final Condition at "Equilibrium"^a</u>			
Pressure in autoclave, psia (max)	107	770	240
<u>Enthalpy Balance in System^a</u>			
Total enthalpy increase (heat out)			
evaluated at final temperature, cal	78,200	177,500	89,900
Heat lost into autoclave, %	60	30	55
Corresponding thermal efficiency, %	40	70	45

^aEvaluated at the time of peak autoclave pressure (12 sec in CEN-217S, 30 sec in CEN-220S, and 130 sec in CEN-223S).

F. Engineering Safeguards Systems

1. Containment

A thorough study of the applicability of the energy-absorption concept to water-moderated reactors has shown that it is entirely feasible.

The first step was the construction of a flow-sequence diagram identifying the major events in a loss-of-coolant accident and their inter-relationships. This was done in order that the energy-absorption concept could be viewed in its proper perspective. A study of the time duration of pressure pulses in a loss-of-coolant accident gave a range of hundreds of milliseconds to minutes, depending upon the extent to which the individual events, such as metal-water reactions, blowdown, and size of openings, influenced the overall sequence. Thus, for all practical purposes, the pressure loading on the energy-absorption container will be static. If a containment designed in accordance with the energy-absorption concept can withstand explosion of a given amount of TNT, it should be capable of withstanding an explosive accident in water-moderated reactors involving from three to ten times that energy release.

The usual assumption in a core excursion is that it is initiated at the center of the core. In reality, it is possible for the excursion to be initiated anywhere within the core volume. Thus, the eccentricity of the excursion with respect to the core center and its effect upon damage to the energy-absorption containment was considered. For present-day LMFBR, the ratio of core diameter to the concrete-wall radius is such that the difference in loading on the wall between center-initiated and noncenter-initiated excursions will be minor. This is due to the fact that the shock wave in the coolant decays quite rapidly so that the strength of the shock wave which hits the gas space before the concrete wall is practically independent of the eccentricity of the excursion. These statements do not hold true for future LMFBR designs in which the previously defined radii ratio approaches one.

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