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Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

April 1967

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

ARGONNE NATIONAL LABORATORY
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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

April 1967

Albert V. Crewe, Laboratory Director
Stephen Lawroski, Associate Laboratory Director

<u>Division</u>	<u>Director</u>
Chemical Engineering	R. C. Vogel
Idaho	M. Novick
Metallurgy	M. V. Nevitt
Reactor Engineering	L. J. Koch
Reactor Physics	R. Avery
Remote Control	D. P. Mingesz (Acting)

Report coordinated by
R. M. Adams and A. Glassner

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FOREWORD

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

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

The last six reports issued
in this series are:

October 1966	ANL-7267
November 1966	ANL-7279
December 1966	ANL-7286
January 1967	ANL-7302
February 1967	ANL-7308
March 1967	ANL-7317

REACTOR DEVELOPMENT PROGRAM
Highlights of Project Activities for April 1967

EBWR Plutonium Recycle

EBWR has been operating at 70 MW since the middle of March. The reactor was shut down on April 29th so that special and normal plutonium fuel pins could be removed from the reactor for subsequent isotopic analysis at Pacific Northwest Laboratory. At the time of shutdown, the average exposure of the plutonium fuel was approximately 2400 MWd/tonne. After shutdown, the reactor will be operated at 70 MW until the next shutdown scheduled for July 1.

EBR-II

An intensive program of sodium sampling to determine the concentration of copper in sodium continued. Sampling upstream and downstream of the cold trap showed that the average copper content on the effluent side of the cold trap was smaller than in the bulk sodium.

With receipt of AEC approval for power operation, Run No. 25 was begun on April 21, following several days of zero-power operation for kinetics measurements. Because of the replacement of depleted uranium with stainless steel subassemblies in Rows 7 and 8, an extremely cautious approach-to-power program was initiated. Upon reaching 22 MWt, the observed decreasing power coefficient averaged 1 lh/MWt, an operating limit of the reactor. Accordingly, the reactor was shut down and then recycled through several power operations from 0 to 45 MWt in small increments while closely monitoring the power coefficient at each step increment. At month's end, rod-drop data taken at all increments showed a consistent prompt negative power coefficient.

Nineteen additional blanket rods were removed from Rows 6, 7, 8, and 10 of the reactor for dimensional and density measurements.

Work is progressing on installation of components for the cold fabrication addition to the Fuel Cycle Facility. Building of the injection-casting equipment and design of the final assembly machine are 50% completed; pin processing and element-processing units are 75% completed. The Laboratory's recommendations for selection of an outside vendor for driver fuel have been submitted to AEC.

Sixteen subassemblies were fabricated this month.

ZPR-3

Assembly 49, the second of a planned series of plutonium cores with well-degraded spectra has been loaded into the reactor. The uncorrected critical mass for this assembly is 282 kg plutonium. Fuel-drawer worths at the edge and control- and safety-rod measurements are under way to provide data for critical-mass corrections. Analyses of data from Assemblies 48 and 48A (the latter in support of the FFTF program) are presented this month.

ZPPR

A significant amount of concrete pouring was accomplished this month; the reactor cell wall has been placed up to the lower ring beam, the roof of the fuel-storage vault-workroom area was poured, and the west wall of the support wing has been placed. Placement of backfill and interior work on finishing of walls, installation of ventilating ducts, etc., is in progress. The reactor bed and tables are now protected in on-site storage by an impact shield. The Final Safety Analysis Report is nearing completion.

AARR

The primary-pump proposals have been received and the reactor-vessel contract has been negotiated and is awaiting AEC approval.

Detail design of the beam-tube test facility has been completed.

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I. PLUTONIUM UTILIZATION--EBWR

A. Operations

After two months of operation at 70 MWt (5 MWe), the EBWR facility was shut down on April 29, 1967. This completed the second irradiation step. At the time of shutdown, the average exposure of the plutonium fuel was approximately 2400 MWd/tonne. Seventeen fuel pins will be removed from the southwest quadrant of the plutonium zone. One set of plutonium foils will be changed in the plutonium zone and a set placed in the enriched zone. The pins removed at the completion of the first irradiation step (see Progress Report for February 1967, ANL-7308, p. 1) will be removed from the EBWR containment shell for transfer to Battelle Northwest Laboratory.

On April 27, the turbine was taken off line due to a sudden increase in the vibration magnitude. As an external cause could not be found, the top turbine casing will be removed during the shutdown to locate and, hopefully, repair the cause. This will lengthen the shutdown schedule by approximately one week. Power operations are expected to resume by mid-May 1967.

The power generated by the plant was as follows:

	<u>Thermal Power</u>	<u>Electrical Power</u>
During April 1967	48,182.1 MWh	3,802.0 MWh
Total Since Start of Plutonium Recycle Program	165,533.9 MWh	16,671.0 MWh

II. LIQUID-METAL FAST BREEDER REACTORS

A. EBR-II

1. Operations

An intensive sodium-sampling program continued during the month (see Sect. II.A.2) to determine the extent of copper dissolution in the primary sodium system and to ascertain the effect of cold trapping on reducing this copper content (see Progress Report for March 1967, ANL-7317, pp. 4-7). The concentration of copper in the primary sodium averaged about 1.18 ppm for all samples, and cold trapping for about a month has not produced a noticeable change in this value. However, the data taken on samples from the effluent side of the cold trap showed a smaller average concentration of copper than did the bulk sodium, about 0.69 ppm copper for all samples taken.

Inspection of photographs taken of the primary auxiliary pump negative bus bar when it was removed from the reactor in March showed what appeared to be either a scratch or a crack. The bus bar was again removed and inspection showed that only a shallow scratch mark was present on the bus bar sheath. The scratch was buffed out and the bus bar was replaced on April 7.

All outer-blanket uranium subassemblies in Row 8 were replaced with stainless steel subassemblies and the required adjustments of fuel loading were made in preparation for Run No. 25. Because of the indication from the primary-tank cover-gas chromatograph that nitrogen content had built up in excess of 20,000 ppm, the cover gas was purged with argon to bring the nitrogen content down to less than 5000 ppm. The reactor was operated on April 19 for zero-power kinetics measurements related to the loading for Run No. 25. These included control-rod drops using the stainless steel control rod, trapezoidal oscillations from movement of a control rod, and period calibration of all control rods.

Approval for operation was received from AEC late on April 20 and the reactor was started up for power operation the next day. Because of loading changes made since the last run, the schedule for approach to power was extremely cautious. It was necessary, for example, to recalibrate the nuclear detectors because of the change in leakage flux resulting from the loading changes; this was done using the thermal output of the reactor as a reference. Measurements of power coefficient and kinetic performance of the reactor were made at small increments of increasing power.

At low powers the power coefficient was normal; however, its value decreased with increasing power until at about 22 MWt the overall power

coefficient averaged 1 lh/MW. Since this is one of the operating limits of the reactor, operation was suspended and a temporary waiver to the limit for the duration of the startup experiments was requested and obtained.

On April 24, the reactor was restarted, taking small, incremental power steps of 2.5 MW to determine precisely the power coefficients and to take rod-drop measurements at 10, 20, and 25 MW to determine the magnitude and sign of the prompt power coefficient. The reactivity data on this second run agreed well with that observed on the initial Run No. 25, and AEC approval was given to proceed above 25 MW on the basis that continued surveillance of the incremental power coefficients and the results from rod-drop tests assured a negative power coefficient and a prompt negative feedback.

The isothermal temperature coefficient of reactivity was established by noting changes in control-rod position at 50 kW when the bulk sodium temperature was reduced from 700 to 650°F. The data gave a value of 1.04 lh/°F, quite close to the value of 1.01 lh/°F established for earlier loadings.

Subsequently, the reactor was recycled through two additional power operations from 0 to 45 MW, and data were taken at 37.5 and 45 MW to determine overall and incremental power coefficients. Results are given in Table I. It will be noted that the magnitude of the power coefficient in the

TABLE I. EBR-II Power Coefficient--Run No. 25

Power (MW) ^a	$\Delta k/k$ (lh) ^a	$\left(\frac{\Delta k/k}{\text{MW}}\right)$ Overall Average (lh/MW)	$\left(\frac{\Delta k/k}{\Delta \text{MW}}\right)$ Incremental (lh/MW)
0	0		
10.0	16.2	1.62	1.62
12.5	18.2	1.46	0.80
15.0	19.6	1.31	0.56
17.5	21.0	1.20	0.56
20.5	22.5	1.10	0.50
22.5	23.6	1.05	0.55
25.0	24.8	0.99	0.48
28.0	27.0	0.96	0.88
30.5	28.3	0.93	0.52
33.0	29.6	0.90	0.52
36.0	30.0	0.89	0.80
37.5	34.5	0.92	1.00
40.0	37.0	0.93	1.00
42.5	40.2	0.95	1.28
45.0	43.2	0.96	1.20
Repeat 1			
37.5	30.3	0.81	
45.0	38.4	0.85	1.08
Repeat 2			
37.5	31.0	0.83	
45.0	39.0	0.87	1.07

^aThe power level has an uncertainty of about ± 0.5 MW and the reactivity an uncertainty of ± 0.5 lh.

two repeat runs is smaller than that of the first run, which may be due to a slight, permanent deformation of the core.

The significant difference between these last two sets of power-coefficient data and those previously taken is postulated to be due to the different heating effects and bowing characteristics of the new stainless steel reflector subassemblies in Rows 7 and 8 as compared to the previous uranium-containing subassemblies. Separating out the effects will require considerable analysis and carefully designed kinetics experiments.

By month's end, rod-drop data taken at 10, 20, 25, 30, 35, 40, and 45 MW all showed the existence of the usual prompt negative power coefficient.

2. Copper in Primary Sodium

The sampling system previously described (see Progress Report for March 1967, ANL-7317, pp. 4-7) was utilized for intensive sampling of the primary cold trap influent and effluent until late on April 3. Sampling of bulk sodium through the Fuel Transfer Port (FTP) was continued, also. The chemical analyses for copper are presented in Table II.

TABLE II. Cold-trap Performance in Copper Removal
Sample Vessels--10-ml Pyrex Beakers

Date	Time	FTP ^a (ppm) Cu	Date	Time	PIA ^b (ppm) Cu	Date	Time	PIB ^c (ppm) Cu
4/1/67	1030	3.55	4/1/67	2100	0.90	4/1/67	1125	0.45
	1500	1.32		2325	0.50		1330	0.26
	2000	0.72	4/2/67	0210	0.87		1540	3.34
4/1/67	2220	1.44	4/2/67	0440	0.40		1925	0.15
4/2/67	0055	0.70		0650	1.20		2200	0.59
	0450	0.97		2040	0.74	4/2/67	0045	0.46
	1310	0.54		2215	1.05		0250	0.79
	1625	0.79	4/3/67	0100	1.20		0540	0.25
	2140	0.60		0245	0.81		0730	0.62
All value average 1.18 ppm				0425	1.32		0920	0.42
Average without highest				0625	0.73		1050	0.25
value 0.88 ppm				0754	0.79		1345	0.31
				0955	0.67		0540	2.37
				1140	0.86		1735	0.74
				1440	0.79		1925	0.37
				1705	1.22		2000	0.54
				1855	1.04		2130	0.54
				1950	0.88	4/3/67	0015	0.25
			All value average 0.88 ppm				0200	0.82
							0345	0.65
							0545	0.48
							0705	1.95
							0900	0.25
							1105	0.37
							1345	0.93
							1630	0.61
							1733	0.29
							1923	0.30
						All value average 0.69 ppm		
						Average without values greater		
						than 2.0-0.52 ppm		

^aFTP--Fuel-transfer Port.
^bPIA--Cold-trap Influent.
^cPIB--Cold-trap Effluent.

To give a better weighting of data than that given only by samples taken after April 1, 1967, nineteen values preceding this date at 1030 for the FTP and ten values preceding this date at 2100 for cold-trap effluent (PIA) were added to make up Table III. It might be argued that this treatment is not valid due to the time lapse between samples. However, over the entire period of sampling, no trends up or down were detectable, indicating, for the period shown in the table, that there are no time-dependent concentration variations.

TABLE III. Weighted Copper Data

Date	Time	FTP (ppm) Cu	Date	Time	PIA (ppm) Cu	Date	Time	PIB (ppm) Cu
3/28/67	1550	1.91	3/24/67	0050	1.54	4/1/67	1125	0.45
	1730	1.01		1420	0.31		1330	0.26
	1940	0.72		1600	0.86		1540	3.34
	2125	2.02		2100	0.87		1925	0.15
3/29/67	0230	3.09		2245	0.73		2200	0.59
	0355	1.12	3/27/67	1355	0.88	4/2/67	0045	0.46
	1825	0.77		1730	1.58		0250	0.79
	2000	0.46		2010	0.95		0540	0.25
3/30/67	0050	1.41	3/28/67	0215	0.99		0730	0.62
	0220	0.23	3/30/67	2030	1.21		0920	0.42
	0415	2.28	4/1/67	2100	0.90		1050	0.25
	0620	0.65		2325	0.50		1345	0.31
	1100	0.42	4/2/67	0210	0.87		1540	2.37
	1300	0.41		0440	0.40		1735	0.74
	2150	0.45		0650	1.20		1925	0.37
3/31/67	0200	1.41		2040	0.74		2000	0.54
	0230	2.46		2215	1.05		2130	0.54
	1530	0.63	4/3/67	0100	1.20	4/3/67	0015	0.25
	2145	0.55		0245	0.81		0200	0.82
4/1/67	1030	3.55		0425	1.32		0345	0.65
	1500	1.32		0625	0.73		0545	0.48
	2000	0.72		0754	0.79		0705	1.95
	2220	1.44		0955	0.67		0900	0.25
4/2/67	0055	0.70		1140	0.86		1105	0.37
	0450	0.97		1440	0.79		1345	0.93
	1310	0.54		1705	1.22		1630	0.61
	1625	0.79		1855	1.04		1733	0.29
	2140	0.60		1950	0.88		1923	0.30
Total		32.63			27.76			19.35
All Value Avg.		1.16 ppm			0.92 ppm			0.69 ppm
Values > 2 Out		0.88 ppm			0.92 ppm			0.52 ppm

The results of averaging the data in Table II allow one to believe the cold trap is removing copper from the sodium. Table III, showing 28 values for each of the three points, reinforces this conclusion.

In preparing Table III, two very high values (24.25 and 9.95) for the FTP were discarded. We believe that this is justified because a relatively high copper concentration was found in a sample of sodium and sodium oxide scraped from the surfaces of the gripper guide tube in the fuel-transfer port. Even though the dip sampler used for taking samples of bulk primary sodium through the FTP was designed so as to minimize the possibility of sample contamination, complete assurance cannot be given that contamination did not occur.

The above-mentioned sample of scrapings was found to contain 39 ppm of copper. It is suspected that this copper may have been carried, as minute copper oxide particles, from the copper mesh packing of the gas purifier in the Fuel Unloading Machine (FUM) argon system. Consideration is being given to removal of the gas purifier, thus eliminating potential copper contribution from this source.

The equipment for sampling the primary sodium was modified for radioactive service. This work was completed on April 10, except for the installation of lead shielding, which was deferred pending checkout of the equipment. This sampling equipment is an adaptation, for radioactive service, of that used earlier for the intensive sampling of cold-trap influent and effluent. Samples can be taken in 10-ml Pyrex beakers from either the PI-A (cold-trap influent) or the PI-B (cold-trap effluent) sampling point. Sodium samples were taken from each sampling point on April 12 and 13 for checkout of the sampling system. Operation was satisfactory and, therefore, installation of shielding was completed. The samples were analyzed for copper content, with the results given in Table IV.

TABLE IV. Copper Analyses of Cold-trap Influent and Effluent

Date	Time	Sample Point	Cu (ppm)
4/12/67	1030	PI-A	0.95
4/12/67	1230	PI-B	0.33
4/12/67	1430	PI-A	0.61
4/12/67	1715	PI-B	0.28
4/13/67	1845	PI-A	0.49
4/13/67	2240	PI-B	0.72

A series of sodium samples was taken from the secondary sodium system on April 14, using the same sampling equipment as had been used for the primary sodium sampling of April 1-3. The purpose of this work was to check the scatter of analytical results for samples from a large system which should be uniform and constant in copper concentration. The results appear in Table V.

TABLE V. Copper Analyses of Secondary Sodium on April 14, 1967

Time	Copper (ppm)	Time	Copper (ppm)
1755	0.33	2145	0.36
1835	less than 0.25	2215	less than 0.25
1920	0.27	2245	0.41
2010	0.28	2315	0.25
2040	0.44	2345	less than 0.25
2115	0.27		

On April 18 and 19, 1967, samples were taken from the primary cold-trap influent and effluent with different flowrates through the cold trap. The copper analyses for these samples are presented in Table VI. There is no obvious effect of flowrate upon copper concentrations.

TABLE VI. Copper Analyses for April 18 and 19, 1967

Date	Time	Sample Point	Cold-trap Flowrate (gpm)	Copper (ppm)
4/18/67	1215	PI-A	15	0.45
4/18/67	1550	PI-B	15	1.89
4/18/67	1730	PI-A	15	0.49
4/18/67	1850	PI-B	15	0.45
4/18/67	1945	PI-A	15	0.43
4/18/67	2040	PI-B	15	0.40
4/19/67	0930	PI-A	10	0.43
4/19/67	1030	PI-B	10	0.31
4/19/67	1140	PI-A	10	1.20
4/19/67	1230	PI-B	10	0.64
4/19/67	1310	PI-A	5	0.32
4/19/67	1345	PI-B	5	0.38
4/19/67	1405	PI-A	5	0.46
4/19/67	1430	PI-B	5	less than 0.25
4/19/67	1450	PI-A	5	0.66

3. EBR-II Driver Fuel Development and Metallurgical Assistance

a. Anomalous Swelling of Mark-IA Fuel. A series of experiments are being undertaken to determine in CP-5 the effects of various parameters such as pressure, heat treatment, and impurities, acting individually and combined, on the anomalous irradiation swelling behavior of U-5 w/o Fs alloy pins (see Progress Report for March 1967, ANL-7317, pp. 8-12). Now in preparation, the first of these experiments will be to investigate the effect of superimposed pressure on fuel-pin swelling. To achieve this, six 0.144-in.-dia by 1.00-in.-long specimens cut from a single EBR-II fuel pin are being prepared for encapsulation under gas pressure inside 0.25-in.-OD by 0.15-in.-wall Inconel-625 tubes. Each specimen will be pressurized at a different internal pressure to enable an evaluation to be made of the effect of external swelling on swelling rate of the fuel alloy. The range from 50 to 2000 psi has been selected for study. This range will be covered in steps with a single specimen at each of the following pressures: 22, 44, 110, 220, 440, and 880 psi, which have been calculated to increase under irradiation conditions and temperature to 50, 100, 250, 500, 1000, and 2000 psi, respectively.

A procedure has been developed for using a Swagelok fitting on each jacketed specimen tube so as to permit the final closure to be electron-beam welded without loss of internal pressure while the interior is under the desired pressure. Methods for leak testing the final closure and for verifying that no pressure loss has occurred during final closure have been experimentally demonstrated. In the leak testing, the helium gas sealed within each individual tube is searched for with a helium mass-spectrometer leak detector. Verification of internal pressure is achieved by means of a miniature strain gauge cemented on the outer diameter of the specimen jacket. Strain gauges will be used on the jackets of all the encapsulated specimens used in the reactor experiment. These gauges and the Swagelok fitting will be removed before assembly into the test capsule.

b. Mark-II Fuel-element Development. Calculations have been completed for the Safety Analysis Report for irradiation of encapsulated Mark-II elements in EBR-II.

Sixty-two elements are being fabricated. Thirty-seven will constitute the initial subassembly loading; the remainder will be utilized as replacements in the subassembly as elements are removed for inspection at 1, 2, 3, 4, and 5 a/o burnup.

4. Reactor Improvements

a. Irradiated-fuel Monitor. Because of the long (30-week) delivery schedule for a monoball-type bearing planned for use in the mechanism, an available substitute will be used. This has necessitated modification of parts in the connection between the drive mechanism and the sensor extension. Fabrication of hardware (see Progress Report for January 1967, ANL-7302, pp. 11-12) is ~80% complete.

b. Sodium-chemistry Technology. In connection with analyses of possible causes of and problems for copper contamination in the primary-system sodium (see Progress Report for March 1967, ANL-7317, pp. 4-7), the original plugging-loop (PI-A) economizer is being examined destructively, and plans are being made for a new plugging loop and sampling station.

(i) Examination of Plugging-loop Economizer. The entire economizer was X-rayed in a search for gross deposits of Cu, Na₂O, or other material; no deposit of any kind was found. However, the radiographs did reveal a deformed or flattened section of the 0.5-in.-OD tube within the 0.75-in. (nominal-pipe-size) Schedule-40 shell. The radiographic evidence revealed that the flattened region is ~13 in. long and centrally located (that is, ~8 in. from either end of the economizer); measurements of several sets of radiographs indicated that the major and minor axes of the deformed-tube cross section are 11/16 and 5/16 in.

A 3-in.-long section was cut from the upper and lower ends of the economizer so surfaces could be inspected and samples of frozen sodium obtained for analysis. After the shell halves of the two end sections were slit and laid open, frozen-sodium samples were taken and are being analyzed. The sodium remaining on these shell pieces was dissolved with ethanol, and the surfaces were then examined and photographed. On the surfaces on which copper had been deposited, the deposit is friable and a considerable portion sloughed off because of the agitation when the sodium and ethanol reacted. Close examination of the shell halves during and after the reaction revealed copper deposited in the lower 3-in. section, but no visual evidence of copper on the upper 3-in. section.

This information indicates that between the time sodium enters at the bottom of the shell and the time it emerges at the top, changes in flow, temperature, and copper solubility bring about conditions that result in copper deposition at the lower end of the economizer but no deposition at the top.

Planned examination of the entire inner surface of the shell will disclose the exact location of the transition region. Chemical analyses of sodium samples taken from various sections and recovery of the deposited copper from all sections will furnish a profile of copper concentration for the entire economizer.

(ii) New Plugging Loop and Sampling Station. A new sodium plugging indicator and sodium-bypass freeze sampler were proposed for installation in the EBR-II Fuel Element Rupture Detection (FERD) loop. Preliminary concepts have been designed. All details of the plugging indicator have been decided, and final design is underway. However, some aspects of the concept for the freeze sampler are still under consideration.

The new proposed sodium plugging-indicator loop will be located on the discharge side of the main FERD loop pump and will consist of a plugging valve with serrated stem (12 grooves, each 0.031 in. deep by 0.031 in. wide), a sodium-to-sodium heat economizer, a magnetic flowmeter, a small electromagnetic pump ($\sim 3/4$ gpm), and two isolation valves. Except for having the pump control the sodium flow rather than having a throttle controlling system pressure drop, the loop is essentially similar to the ones now used at EBR-II. Means will be provided for controlled air cooling and plugging-valve maintenance and/or removal.

The sodium-bypass freeze sampler, as initially proposed, would consist of a 0.5-in.-dia U-shaped nickel tube with a total length of ~ 24 in. The sample tube would be connected across the FERD-loop main pump and be subjected to its normal differential pressure; estimated flow through the tube would be 11 ft/sec. The tube, which would be at the highest

point of the FERD loop, would be easily accessible from the operating floor and would require a minimum of modifications and additions to the radiation shielding.

c. Rotating-plug Seal Cleaning System. Improvements were made in the oxide-removal devices and the circulation system for the bismuth-tin alloy. The shape of the collector tank for the skimming mechanism (see Monthly Progress Report for March 1967, ANL-7317, pp. 12-13) has been modified to increase the efficiency of oxide removal. The new tank configuration performed very well in tests in the laboratory mockup, but further improvement is expected as a result of design changes being made on the skimming mechanism. The alloy return line and discharge nozzle are being modified to improve flow characteristics. A prototype centrifugal-type oxide separator is being fabricated for initial testing in the laboratory mockup of the system.

d. Irradiation Subassembly Mark C. Hardware was received for hydraulic studies of the Mark-C19 irradiation subassembly. This subassembly contains nineteen elements of 0.290-in. OD and 44-in. length, and incorporates the coolant-bypass feature that permits acceptable subassembly mixed-coolant outlet temperature with much higher coolant temperatures at the element exit. Hydraulic studies of the Mark-C37 subassembly (see Progress Report for March 1967, ANL-7317, p. 13) and Mark-C mixing studies were completed.

e. Irradiation Subassembly Mark E. Preliminary design concepts have been initiated to provide a subassembly capable of withstanding the physical damage resulting from a postulated failure of fuel-element cladding. The subassembly structure must contain all fragments or portions of the fuel element without adverse effect or damage to adjacent subassemblies and other reactor components. Calculations and analyses on the first concept led to a second concept that incorporates an increased quantity of temperature-resistant material; these modifications are being evaluated.

f. Fuel Subassembly Mark-II. Drawings of dummy test fuel elements and four designs of shield configurations have been completed. The Mark-I fuel subassembly Model-STB was tested hydraulically to measure the pressure drop in the existing major components, specifically the lower shield, fuel elements, and the upper shield. These measurements will be compared with those of the new Mark-II components to match the pressure drop of the Mark-II subassembly with existing fuel subassemblies.

g. Instrumented-subassembly System. Effort continued toward developing the systems concept and equipment requirements necessary to place instrumented subassemblies in the reactor core.

(i) Operating Mechanism. A reference design for the operating mechanism has been completed, in which the subassembly is moved vertically so that the rotating plug can be indexed. As shown in Fig. 1, the various mechanism parts have been arranged to form a compact unitized structure, which can be readily attached to the upper section of the central control-rod support column. Because of its compactness, the mechanism can be installed in any of the 12 spaces now occupied by control-rod drives.

The essential parts of the mechanism are: (a) drive apparatus for rotating the lead screw, installed on top of the mechanism, (b) drive screw for changing rotating to vertical motion of the elevator plate, (c) a terminal box for instrument leads, attached to the elevator plate, and (d) the motorized extension-tube seal and bellows. The seal surrounds the extension tube inserted between the instrumented subassembly and the instrument-lead terminal box (see Progress Report for February 1967, ANL-7308, p. 5).

Figure 1 also shows the method for mechanically coupling the extension tube and terminal box to the elevator plate such that the forces required to raise or lower the instrumented subassembly can be observed at all times. Note the location of the limit switches that arrest further motion if frictional forces in the seal or extension tube become high.

h. Higher-worth Control Rod. So that up to four of the present 12 control-rod locations in the core could be made available for installation of an oscillating rod, instrumented irradiation subassemblies, or other special in-core experimental devices, studies are being made of methods by which to increase the reactivity effectiveness of the remaining eight control rods, which now contain fuel. Preliminary dimensional layout and analysis show that this rod will likely be an all-absorber rod rather than consisting of fuel rods with an absorber follower. The gas space required in the top of the fuel elements limits the proximity of any absorber section to the fuel section to the extent that only an inadequate portion of the absorber section could enter the reactor core. Efforts are being made to increase the boron carbide absorber material commensurate with cooling requirements.

i. Oscillator Rod Mark II. During routine transfer-function measurements of the EBR-II, the effort required to turn the Mark-II oscillator rod increased markedly at very slow speeds and high reactor temperature differentials. Removal and subsequent inspection of the rod revealed rubbing marks on one side of the rod. It was concluded that these rubbing marks were caused by a combination of rod bowing and thimble bowing at the lower speeds and higher temperature differentials.

Fig. 1. Instrumented-subassembly Operating Mechanism; Overall at Left, Detail at Right

As a first step to correct this condition, design changes are being studied. If the rod were 0.2 in. smaller in diameter, the rod and thimble could bow somewhat without interfering with each other. A more effective balance of heat generation within the rod would lessen the bowing of the rod; a scheme being studied would involve balancing the boron carbide capsule within the oscillator rod with a uranium oxide rod.

A special thimble for use with the oscillator-rod assembly would replace the standard control-rod thimble in the reactor. The bottom of the thimble would have a spring-loaded bayonet, which, when locked to the reactor grid pin, would cause the thimble to exert a downward seating force of about 230 lb on the top grid plate. This seating force would stabilize the thimble and provide a firm housing for the oscillator-rod lower bearing. The thimble would have a pin fastened to its lower end, to which the oscillator-rod assembly bayonet would lock. Thus, by locking the oscillator-rod assembly to the thimble pin instead of to the grid pin, the oscillator-rod assembly would be isolated completely from the reactor grid, thereby reducing the possibility of handling difficulties.

The oscillator-rod assembly also could be shortened to permit its storage in the storage basket. To accommodate the oscillator-rod assembly, a sleeve for receiving the rod assembly would be locked into a storage tube in the outer row of the storage basket. A special gripper would be used to install and lock the sleeve in the storage-basket tube. The gripper may also be used to remove the sleeve from the basket.

j. Modification for Increased Fuel Production

(i) Bonders. The new bonders have been delivered. Calibration of the assembled system is scheduled to start the second week in May. Additional testing will be performed on this unit to prove a new zone-heater control design. Optimum thermocouple locations will be selected prior to installation in the Argon Cell.

(ii) Bond Tester. The readout equipment has been field-tested on an existing system and has been accepted for operation. Work on the controls is approximately 60% complete.

k. Nuclear Instrumentation. Circuit-design work has started for adaptation of the new equipment to the existing reactor protective circuit and readout equipment. Life testing of relays used in the system for final control has continued.

A check with the equipment manufacturer was made and his report is that the equipment deliveries remain firm as stated.

1. Reactor Systems Instrumentation

(i) Temperature-monitoring Devices. The detailed circuit modifications necessary to interconnect the new equipment to the existing systems is approximately 60% complete. The monitor switches ordered have been delivered, and the modifications necessary for fail-safe operation will start the first week in May. No change in delivery of the remaining equipment is anticipated.

(ii) Miniature Recorders. Work was started on the alarm and control functions, and then stopped to complete work on the bulk-sodium-level system. Completion of the necessary engineering will be scheduled for May.

The float-operated, bulk-sodium-level indicator has been installed in the primary tank, and connected to the readout and control instrumentation. The unit will be monitored closely for operation for one or more reactor operating periods before substituting it for existing level equipment. Two sets of alarms have been set up for monitoring. One set of high and low alarms have been preset to actuate a mechanical counter at ± 1 in. from normal level and a second set at ± 3 in. The normal reactor scram points for the system are ± 3 in. Since the system has been placed into operation, no trips have occurred due to noise.

m. Selected Parameter Data Processing System

(i) 100-point System. Manufacturing follow-up has continued; although the manufacturer states he will deliver on time, it is somewhat doubtful.

(ii) 50-point Scanner. The 50-point system has been installed and the operation debugged. Work is in progress aligning the system for operation on plant parameters. The system will be in operation by April 26, 1967.

5. Surveillance

a. Effect of Stress on Type 304 Stainless Steel Exposed to Sodium. Earlier work¹ indicated that stressed Type 304 stainless steel is affected more adversely by 1200°F sodium than is the unstressed material. Further screening tests on as-received samples of EBR-II radial-blanket tubing have continued (see Progress Report for February 1967, ANL-7308, pp. 5-7). After samples of Type 304 stainless steel tubing were exposed for 6 days to various environments in stressed or unstressed conditions, the specimens

¹Smith, F. A., et al., 1200°F Sodium-free-surface, Stress, and Wear Effects on Reactor-component Metals, IAEA Symposium, SM-85/22, Vienna, December 1966.

were exposed to Strauss reagent for various times, after which the electrical resistivities of the specimens were measured. The Strauss reagent accelerates grain-boundary or sensitization effects. A deadweight method was used to apply an arbitrarily chosen stress of 59% of the yield strength at 1200°F. The same deadweight loading fixture was used for exposures to both air and sodium. Test conditions and data are summarized in Fig. 2, from which it can be seen that:

(i) For the as-received samples, the Strauss test did not affect the resistivity of either the stressed or unstressed samples.

(ii) For the samples exposed to 1200°F air (after having been pretreated in a 1200°F vacuum for 35 days), the Strauss test increased the resistivity only slightly; there is little difference between the stressed and unstressed samples. These results are in general agreement with the effect of sensitization in air at 1200°F to increase the resistivity of Type 304 stainless steel.

(iii) For the samples exposed to 1200°F sodium (after no pretreatment), the resistivities of both the stressed and unstressed samples increased markedly as a result of the Strauss tests, but the stressed samples were affected much more than were the unstressed samples. The large difference between results for the stressed and unstressed samples was unexpected because the stress was relatively modest (59% of yield strength). Duplicate samples were tested to evaluate the reliability of the measurements; the spread in the measurements may be related to inclusions or metallurgical differences.

(iv) For the samples exposed to 1200°F sodium (after having been sensitized in 1200°F vacuum), the unstressed sample was affected considerably, but the stressed sample was affected catastrophically by only 15-min exposure to Strauss reagent. This indicates that presensitized Type 304 stainless steel that is exposed to 1200°F sodium when under stress has a microstructure very susceptible to intergranular corrosion.

6. Experimental Irradiations

The status of all experimental irradiations in the reactor and the approximate exposure at month's end is shown in Table VII.

Subassembly XO14, containing structural materials samples from GE, NRL, and PNWL, was discharged, disassembled, and the test capsules shipped to the TAN Hot Shop for interim storage and transfer to the customers' shipping containers. The nominal exposure at the midplane position to these capsules was approximately 1.7×10^{22} nvt (total flux). Removal was requested by one of the experimenters, and this request received concurrence from the other experimenters and LDO.

Fig. 2. Rates by Which Strauss Reagent Attacks Samples of Type 304 Stainless Steel That Had Been Stressed (Solid Data Points) or Unstressed (Hollow Data Points) While Being Exposed for 6 days to 1200°F Sodium or to 1200°F or Room-temperature Air

TABLE VII. Status of Experimental Irradiations in EBR-II

Sub-assembly	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	9,844	13,600
XG03	7/16/65	UO ₂ -20 w/o PuO ₂ (2) Stainless Dummies (17)	GE	9,844	19,450
XG04	7/16/65	UO ₂ -20 w/o PuO ₂ (2) Stainless Dummies (17)	GE	9,844	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	9,417	14,750
XA08	12/13/65	UC-20 w/o PuC (8) Structural (11)	ANL	7,595	19,800
XO10	3/24/66	UO ₂ -20 w/o PuO ₂ (4) Structural (11) Structural (4)	GE ANL PNWL	6,775	19,600
XO11	5/9/66	UO ₂ -20 w/o PuO ₂ (7) UO ₂ -20 w/o PuO ₂ (9) 304 SS-30 w/o PuO ₂ (1) 304 SS-20 w/o PuO ₂ (1) 304 SS-20 w/o UO ₂ (1)	ANL GE PNWL PNWL PNWL	6,204	8,300
XO12	8/10/66	UO ₂ -20 w/o PuO ₂ (19)	NUMEC	3,075	20,600
XO15	11/15/66	PuO ₂ -UO ₂ (11) PuO ₂ -UO ₂ (2) (Pu,U)C (4) MK-IA (Metal) (2)	NUMEC GE ANL ANL	1,420	11,000
XO16	1/13/67	Structural (9) Structural (10)	ANL GE	100	3,000
XO17	11/15/66	PuO ₂ -UO ₂ (11) (Pu,U)C (3) MK-IA (Metal) (5)	NUMEC UNC ANL	1,420	6,500
XO18	12/6/66	Structural (3) Structural (1) Structural (2) Structural and Heavy Metal Fission Yield Samples (1)	GE PNWL ANL ANL	730	21,300
XO19	1/13/67	UO ₂ -20 w/o PuO ₂ (7) (U _{0.8} Pu _{0.2})C (3) Structural (8) Graphite (1)	GE UNC PNWL PNWL	100	7,500 ^a
XO20	1/13/67	UO ₂ -20 w/o PuO ₂ (9) (U _{0.8} Pu _{0.2})C (3) Structural (4) Structural (2) Graphite (1)	GE UNC PNWL ANL PNWL	100	7,500 ^a
XO21	2/27/67	Structural (7)	PNWL	100	21,500
XO22	2/27/67	Structural (7)	PNWL	100	8,000

^aBeing considered for further exposure.

7. Vertical Assembler-Disassembler (VAD)

Assembly of the VAD mechanical components was completed on April 24, 1967; assembly and wiring of the control console has almost been completed. Preparation of installation drawings is still under way.

Operational tests are continuing and have been expanded to the testing of interrelated machine systems. Satisfactory results have been obtained in a limited number of tests of the hex tube cutter, the welding heads, and power supply. The positioning accuracy of the vertical drive units, as well as force-limiting and -measuring features of the machine are better than the design requirements. A test of ease of remote disassembly and reassembly was made. A number of handling fixtures and modifications, seen to be needed at that time, have now been completed. We have been able to make up the time lost during contracting and fabrication of the machine components and are now on schedule.

A final test and rehearsal of operating and installation procedures will be conducted between May 1 and 5. It is planned to carry out final plating, greasing, and other preparations prior to packing for shipment, scheduled for May 19.

8. Fuel Cycle Facility (FCF)

a. Examination of Irradiated Radial Blanket Rods. Nineteen additional rods from 6th, 7th, 8th, and 10th row subassemblies have been sent to Test Area North (TAN) for examination. Diametral measurements will be obtained for the jackets and for the declad rods. Length and density values will be obtained for the declad rods. No significant additional data are yet available.

b. Fabrication of Unirradiated Fuel in the FCF (Cold Line). Installation of the alloy-preparation furnace for cold fuel is substantially complete. When this is available, complete test runs with unenriched uranium will be made. The injection-casting equipment is about 50% complete; the pin-processing and element-processing units are about 75% complete. The design of the final assembly machine is about 50% complete, but existing equipment can be utilized for this purpose. Preparation of the inert-atmosphere glovebox for installation is largely accomplished. These components will be installed and tested as soon as they are completed.

The layout work on the Cold-line Assembly Machine is almost completed. This machine will: 1) hold the components of the EBR-II fuel subassemblies in position during the manual assembly of the fuel elements onto the support grid, 2) provide a device to push on the outer hexagonal tube, and 3) align the subassembly during the welding of the hex tube. A simple and inexpensive method of incorporating a subassembly pull test has been devised and is being added to the machine.

Detailing of the major components has begun and some drawings should be ready for release to fabricators by May 15, 1967. All parts are scheduled to be fabricated by October 1, 1967 and the machine installed in Idaho by November 30, 1967.

The status of several modifications are as follows:

(i) Leak Detectors. Control-circuit and component-interconnecting wiring was started; this work is 40% complete. The solenoid valves ordered for this unit were rescheduled for delivery by the manufacturer. The new delivery date submitted would have delayed the completion of the cold line. By redesign of the system controls, a second manufacturer's valves can be used. These valves were ordered with delivery scheduled for May 5, 1967, and the original valve order was canceled.

(ii) Bonder. In-panel control wiring is approximately 90% complete.

(iii) Pin Processor. In-panel wiring is approximately 70% complete. The balance of the necessary components has been received and can be installed as scheduled.

(iv) Alloy Furnace. Control-circuit and component-interconnecting wiring is 50% complete. All equipment and hardware have been received for completion of this unit.

c. Procurement of Driver Fuel from Outside Vendors. The Laboratory's recommendations for selection of the vendor of the driver fuel have been submitted to the AEC for action.

d. Facility Modifications and Equipment Additions to the FCF. The vertical assembly-disassembly machine (VAD) is near completion and ready for operational testing in Illinois (see Sect. II.A.7). It will then be reassembled in the FCF mockup area for final testing and training operations. A floor penetration in the mockup area has been provided for this purpose.

Preliminary designs are complete for a neutron radiography source to be located in the Argon Cell for use in nondestructive testing of fuel, blanket, and experimental capsules. The "negative film" will be introduced into the cell via a sealed and shielded feedthrough from the subcell, where it will be exposed and then returned to the basement area. The film-transfer mechanism will be completely isolated from the cell atmosphere.

To reduce the number of transfers through the Air Cell, a new opening has been made in the wall of the transfer subcell between the Air and Argon Cells. A window and transfer lock will be installed in this opening,

and two master-slave manipulators will be installed above them. This transfer system will allow all the unirradiated materials, such as crucibles, molds, and waste cans, to be introduced directly into the Argon Cell via the large lock rather than having to traverse the whole length of the Air Cell. When the mobile decontamination facility is complete, this lock system will also be used to remove equipment components for cleaning and repair.

The design of the top loading-top unloading coffin for transfer of subassemblies to the storage facility of the Idaho Chemical Processing Plant is about 75% complete.

e. Operations. Subassembly dismantling, processing, and fabrication operations were continued routinely. Both unirradiated and irradiated fuel was used for fuel element fabrication. A total of 16 subassemblies were completed as shown in the operation summary in Table VIII.

TABLE VIII. Production Summary for April 1967

	<u>April</u>	<u>Total This Year</u>
Subassemblies received:		
Core Control, Safety (for processing)	19	39
Other (XO14)	1	93
Subassemblies dismantled (for processing):	16	40
Subassemblies dismantled (for examination, etc.):	1	45
Subassemblies fabricated: reprocessed	16	60
unirradiated	0	4
Reprocessed subassemblies transferred to reactor:	14	50
Elements decanned:		
From irradiated subassemblies		1027
rejects		<u>77</u>
Total Decanned:		1104

Melt Refining

	<u>Irradiated Fuel</u>	<u>Recycle Material</u>	<u>New Fuel</u>
No. of runs	5	4	0
Average pour yield, %	92.5	93.8	-

Processing

Injection-casting runs (total number):	11	
Elements processed:		
Accepted:	1200	
Rejects:	216	
Elements welded:	1128	Rewelded: 0
Elements leak tested:		
Accepted:	1133	
Rejects:	19	
Elements bonded (including recycle):	1516	
Elements bond tested:		
Accepted	1475	
Rejects:	41	
Elements to surveillance:	80 from 8 subassemblies	

Waste Shipments

Cans to burial ground:	16
Oxide and glass scrap to ICPP (No. of shipments):	1

B. Physics Development

I. ZPR-3

a. Reactivity Sample Traverses in Assemblies 48 and 48A.

Analyses of the data from the traverses of reactivity samples through Assemblies 48 and 48A have been completed. The results obtained for four sets of reactivity sample traverse experiments are given below; they are for the radial traverses through Assembly 48 (with a depleted uranium radial blanket), and for traverses in Assembly 48A with Inconel radial reflector segments (performed in support of the FFTF program) done radially through the reflector, axially through the reflector at the core edge, and radially through B₄C mockup rods in the reflector. The specifications of the samples used in these experiments are given in Table IX.

TABLE IX. Specifications of Reactivity Samples Used in Assembly-48 Traverses

Sample Material	Geometry	Material Dimensions (in.)			Material Mass (g)	Can Mass ^a (g)	Material Composition (w/o)
		Length	OD	Thickness			
Plutonium	Annulus	1.86	0.34	0.020	9.187	6.63	95.05 Pu ²³⁹ , 4.50 Pu ²⁴⁰ , 0.45 Pu ²⁴¹
Enriched Uranium	Annulus	1.90	0.39	0.010	7.128	1.79	93.16 U ²³⁵ , 6.84 U ²³⁸
Depleted Uranium	Cylinder	2.00	0.19	-	19.63	-	99.78 U ²³⁸ , 0.21 U ²³⁵
Enriched Boron	Annulus	1.90	0.39	0.010	0.406	11.55	85.48 B ¹⁰ , 7.32 B ¹¹ , 7.2 impurity ^b
Tantalum	Annulus	1.98	0.38	0.015	9.67	1.77	>99% Ta
Stainless Steel	Cylinder	2.00	0.42	-	35.90	-	Type 304, approx 72 Fe, 18 Cr, 10 Ni
Polyethylene	Cylinder	2.00	0.42	-	4.072	-	CH ₂

^aStainless steel can or bracket.

^bThis composition is suspect, pending confirmation of additional analyses.

Table X presents the results of the radial traverses through Assembly 48 with the reactivity samples. The traverse tube ran through the P-row drawers of Half No. 1 about 1 in. back from the interface. In the

TABLE X. Radial Traverses of Reactivity Samples in Assembly 48

Radius (cm)	Reactivity Coefficient (1h/kg)						Stainless Steel ±0.3
	Plutonium (5% Pu ²⁴⁰) ±1	Enriched Uranium ±2	Depleted Uranium ±0.4	Enriched Boron ±20	Tantalum ±1	Polyethylene ±3	
-73.66	-	-	-	-	-	-4	-
-66.04	1	-1	0.4	-35	0.9	-6	0.5
-60.96	-	-	-	-	-	-13	-
-55.88	6	8	0.5	2	3.0	-45	0.6
-48.26	24	21	0.0	-181	-0.7	-131	2.4
-44.45	49	43	1.0	-335	-2.4	-150	5.5
-41.59	76	64	2.0	-736	-7	+4	8.2
-38.89	103	82	0.6	-1127	-15	238	7.6
-33.34	-	-	-1.4	-	-	-	-
-27.78	229	167	-5.7	-3567	-64	916	0.4
-16.67	367	264	-13.7	-6033	-114	1338	-8.8
-5.56	452	324	-19.0	-7502	-150	1558	-14.3
0.00	464	329	-20.7	-7679	-151	1585	-15.4
5.56	450	322	-18.7	-7394	-145	1544	-14.7
16.67	361	255	-12.4	-5868	-112	1310	-7.9
27.78	223	162	-5.1	-3422	-62	872	+1.0
33.34	-	-	-1.8	-	-	-	-
38.89	101	79	+1.6	-1084	-14	211	7.3
41.59	76	58	2.4	-628	-2.4	-10	7.0
44.45	47	39	1.7	-345	-1.5	-146	4.8
48.26	25	21	0.7	-124	-1.1	-125	2.1
55.88	6	6	0.5	-7	+1.0	-32	0.6
60.96	-	-	-	-	-	-22	-

table, the negative radii refer to positions in matrix column numbers less than 16, i.e., the axis of the core runs through P-16. The data display a distinct shift of the worth profiles in the negative direction. This reflects the asymmetry of the core drawer loading for this assembly, wherein the left half of a Half No. 1 core drawer (in the negative direction) contains about 90% more plutonium than the right half. The accuracies quoted at the column heads in the table represent the average reproducibility found in repeated measurements. However, the overall accuracy of the quoted values is limited to about $\pm 3\%$ because of uncertainties in the calibration of the autorod used for the experiments.

Tables XI and XII present the results of the radial traverses in Assembly 48A (see Progress Report for February 1967, ANL-7308, p. 13, Fig. 4). These traverses were also made through the P-row drawers of Half No. 1. The traverses show the effect of the symmetrically located B_4C poison rods. The effect of these rods on the reaction rates has previously been presented (ANL-7308, p. 13) and these results show the same asymmetries as those in Table X for Assembly 48.

Table XIII presents the results of the axial traverses taken in the reflector sector. These traverses were made in the center of a reflector drawer one inch from the core-reflector interface. The drawers used were 1 and 2-0-9 and the negative position locations indicate Half No. 2.

TABLE XI. Radial Traverses of Reactivity Samples in Assembly 48A^a

Radial Position (cm)	Reactivity Coefficient (lh/kg)				
	Plutonium (5% Pu ²⁴⁰)	Enriched Boron	Depleted Uranium	Tantalum	Stainless Steel
-66.0	4.2	-31	-	-	-0.2
-55.9	38.7	-120	0.9	-2.0	0.3
-48.3	92.1	-605	0.8	-11.0	2.9
-43.2	129.5	-1735	1.2	-29.2	5.7
-40.6	138.0	-2619	-1.7	-46.2	6.2
-38.1	141.6	-3490	-3.1	-62.6	6.7
-35.6	156.4	-3184	-2.4	-69.5	7.1
-33.3	179.1	-3914	-3.0	-70.0	5.7
-22.2	328.4	-5504	-	-	-4.0
-11.1	453.6	-7508	-19.6	-143.5	-14.0
-5.6	-	-8110	-21.8	-157.1	-16.3
0.0	494.7	-8285	-22.2	-162.6	-16.7
5.6	475.1	-7992	-19.9	-153.8	-15.4
11.1	443.7	-7375	-18.5	-141.4	-12.9
16.7	386.4	-6477	-15.5	-123.0	-8.5
22.2	313.7	-5345	-	-	-3.8
27.8	225.4	-5237	-13.6	-93.4	-2.7
33.3	167.2	-3806	-3.0	-66.5	5.7
35.6	145.4	-3813	-4.6	-68.1	6.1
38.1	132.2	-3401	-3.8	59.3	6.6
40.6	128.5	-2571	-2.7	44.2	5.6
43.2	122.8	-1673	-0.6	-28.1	4.7
48.3	89.3	-539	0.9	-6.5	2.2
55.9	36.6	-82	0.2	-1.0	0.2

^aCore boundary was at 36 and -36 cm.

TABLE XII. Radial Traverses of Reactivity Samples in Assembly 48A
with B₄C Poison Rods Inserted^a

Radial Position (cm)	Reactivity Coefficient (1h/kg)			
	Plutonium (5% Pu ²⁴⁰)	Enriched Boron	Depleted Uranium	Stainless Steel
-61.0	11.0	-83	-0.2	-0.2
-49.8	64.0	-163	0.7	0.4
-44.5	79.0	-344	0.6	1.8
-38.7	84.8	-345	5.4	9.8
-33.3	159.0	-1867	4.1	14.2
-27.6	236.8	-3702	-5.7	3.4
-22.2	308.7	-	-	-2.0
-16.5	393.1	-6282	-15.5	-8.4
-11.1	-	-	-	-13.4
-5.7	477.1	-7822	-20.4	-15.3
-2.9	-	-	-	-
0.0	492.1	-8000	-21.0	-16.0
6.4	470.8	-7720	-20.2	-15.2
11.1	433.2	-7086	-	-13.0
16.5	372.9	-6156	-16.7	-9.3
27.6	224.0	-3351	-4.7	4.4
33.3	152.0	-1700	3.8	14.6
38.7	81.6	-242	2.8	8.5
44.5	68.1	-330	-2.4	1.5
49.8	56.6	-140	-	1.9
61.0	7.1	-40	-0.8	0.8

^aThe rod locations were P-9 and P-23, i.e., rod centers at 38.76 and -38.76 cm. These rods were located at the core-reflector interface but completely in the reflector.

TABLE XIII. Axial Traverses of Reactivity Samples in Assembly 48A^a

Axial Position (cm)	Reactivity Coefficient (1h/kg)			
	Plutonium (5% Pu ²⁴⁰)	Enriched Boron	Tantalum	Stainless Steel
-68.6	-	-24	0.5	0.5
-61.0	2.7	-48	-1.5	-0.4
-53.3	5.3	-9	-1.5	-0.3
-45.7	12.4	-125	-3.3	-0.02
-40.6	-	-	-	1.0
-38.1	34.8	-554	-10.4	1.4
-35.6	-	-	-	1.9
-33.0	53.7	-1052	-17.5	3.1
-25.4	88.2	-1898	-31.5	4.0
-17.8	122.9	-2602	-47.9	4.5
-10.2	133.7	-3111	-59.1	4.5
-5.1	137.9	-3284	-62.3	5.1
-2.5	-	-	-	5.6
0.0	143.1	-3318	-65.5	5.7
2.5	-	-3322	-	5.9
5.1	139.0	-3406	-60.5	6.1
10.2	131.1	-3248	-57.1	5.4
17.8	114.9	-2805	-46.5	4.8
25.4	76.8	-2075	-33.3	4.0
33.0	49.2	-1259	-20.5	2.9
38.1	38.4	-733	-9.8	2.3

^aIn the Inconel reflector one inch from the core-reflector interface, down the center of 1-0-9 and 2-0-9.

b. Calculations for the Assembly-48A Results. Values of the B^{10} and plutonium reaction rates were computed with the use of 22 neutron-energy groups by MACH-I code in redimensional cylindrical geometry. Cases considered were with and without control rods.

In Figs. 3 and 4 the measured and computed rates for B^{10} and plutonium are compared.

Fig. 3a. Radial Profiles of Boron-10 Reaction Rate through Assembly 48A without Mockup B_4C Rods

Fig. 3b. Radial Profiles of Boron-10 Reaction Rate through Assembly 48A with Mockup B_4C Rods

Fig. 4a. Radial Profiles of Plutonium Fission Rate through Assembly 48A without Mockup B_4C Rods

Fig. 4b. Radial Profiles of Plutonium Fission Rate through Assembly 48A with Mockup B_4C Rods

Figure 4 shows three computed curves: the upper curve comes from blanket-averaged cross sections, the middle one is based on reflector-averaged cross sections, and the lowest one is derived from core-averaged cross sections. Each curve should have been plotted only in the region for which its cross-section input applied, but this would have implied a neutron-flux discontinuity at the boundaries between core and reflector and between

reflector and blanket. Also, because of neutron-spectrum mixing at the boundaries, many more than three sets of cross sections would be needed to give a quasi-continuous reaction rate curve. Since only one cross section was available for B^{10} , only one computed curve appears in Fig. 3.

The cases without control rods are better simulated by one-dimensional cylindrical computations than the case with control rods, since the rods grossly violate the cylindrical symmetry. For the computations with rods, a region was set up in the form of a symmetrical cylindrical shell with a thickness equal to the rod thickness and with inner and outer radii that included both rods. The rod and reflector materials in this region were considered uniformly diluted. The experimental reaction-rate traverse along the radius, on the other hand, passed through the control rod where the B_4C is concentrated. Because of this situation it was found necessary to double the concentration of rod material in the cylindrical shell in order to depress the reaction-rate maximum in the reflector to the measured value. No attempt was made to add enough rod material to depress the reaction rate in the shell to that measured in the rod, because reaction-rate peak in the reflector would then be depressed far below the measured value. This difficulty may possibly be circumvented if a two-dimensional computation is used in noncylindrical symmetry.

c. Assembly 49. Assembly 49 is now loaded in ZPR-3. This is the second in the planned series of plutonium assemblies with a well-degraded neutron spectrum. This series of assemblies aims to provide a set of integral data which can be used for the evaluation of calculation methods and nuclear cross-section compilations employed in the design of large plutonium-fueled power reactors.

The first assembly in this series, Assembly 48, had a core composition with a U^{238}/Pu^{239} ratio of 4.5, and graphite, sodium, and stainless steel. Assembly 49 has the same core composition as Assembly 48 except that all of the sodium has been removed. Experimental data to be obtained with Assembly 49 will provide important information on the sodium-loss and Doppler coefficients in cores with and without sodium when compared to results from Assembly 48.

The uncorrected critical mass of Assembly 49 is 282 kg of plutonium. Currently, edge fuel drawer and control and safety rod measurements are being made to provide data for critical mass corrections.

2. ZPR-6

A series of experiments designed to vary the potential scattering cross sections σ_p of U^{238} and consequently vary the effective absorption resonance integral in this material, was performed with Assembly 5 of ZPR-6, a 2600-liter uranium carbide core. Four related effects associated with the variation of σ_p of U^{238} were experimentally investigated. These effects were:

- the effect of decreasing σ_p on the critical mass;
- the effect of decreasing σ_p on the sodium-void coefficient;
- the environmental effects on the bunching of U^{238} plates;
- the effect of σ_p on the absolute fission rates of U^{235} and U^{238} and absolute capture rate of U^{238} as a function of position in the three-drawer cell.

The bunching of U^{238} was achieved in two steps. First, the normal three-drawer cell loading (see Fig. 5) was reloaded as shown in Fig. 6. This step was deemed necessary so that the bunching of U^{238} , shown in Fig. 7, would

Fig. 5. Normal Loading Pattern of Three-drawer Cell in Assembly 5 of ZPR-6

Fig. 6. Reference Loading Pattern of Three-drawer Cell for Bunching U^{238} in Assembly 5 of ZPR-6

Fig. 7. Pattern for Bunched U^{238} Loading in Three-drawer Cell of Assembly 5 of ZPR-6

not require any shifting in position of either the U^{235} plates or the sodium cans. It should be noted that the spacing between the U^{235} plates in the three-drawer cell is essentially equidistant. The region in which the loading pattern was changed from that shown in Fig. 5 to that shown in Fig. 6 and finally to that shown in Fig. 7, was a square comprising the central 9 by 9 drawers (49.72 by 49.72 cm) in each half of the reactor. At a radius of about 20 cm from the center of this region, there were four control rods per reactor half, equally spaced about the center. The loading pattern of these control rods (or drawers) was the same as that of the central drawer shown in Fig. 5, and this pattern was maintained in all experiments. The number of drawers in which the loading patterns were changed were 77 per reactor half.

a. The Effect of Varying σ_p of U^{238} on Critical Mass. The reactivity change in the system due to the change in loading pattern from that shown in Fig. 5 to that shown in Fig. 6 was -13.4 ± 0.4 lh for the 154 drawers involved in both halves. This reactivity decrease may be due to either a slight increase in σ_p of U^{238} relative to the normal loading pattern of Fig. 5 or to an increase in the streaming through the light-density channels of sodium.

The change in pattern loading from the reference case of Fig. 6 to the bunched case of Fig. 7 was carried out in steps. The first step was changing the loading pattern in the 9 central drawers per half (a total of 18 drawers). The second and subsequent steps were bunching the U^{238} in rings around the central 9 drawers per half. In Table XIV are summarized the experimental results. The total increase in reactivity in this region was $+131.6$ lh ($0.275\% \Delta k/k$). The experimentally determined decrease in the critical mass that is equivalent to this reactivity increase was 51.6 kg of U^{235} .

TABLE XIV. Reactivity Change due to Bunching U^{238} in Assembly 5 of ZPR-6

$R_i - R_o$ (cm) ^a	No. of Drawers	Reactivity Change (lh) ^b
0-9.35	18	$+17.5 \pm 0.4$
9.35-15.85	32	$+30.6 \pm 0.4$
15.85-21.95	41	$+33.6 \pm 0.4$
21.95-28.45	63	$+49.9 \pm 0.4$

^a R_i and R_o are the equivalent inner and outer radii of cylindrical rings.

^bThe bunching in a ring was performed around a bunched region.

The data shown in Table XIV is compared with a $J_0^2(2.405 r/\tilde{R})$ distribution in Fig. 8. It is seen that the fit is reasonable over the range of measurements. If it is assumed that the J_0^2 distribution is reasonable

Fig. 8

Extrapolation of the Worth of U^{238}
Bunching in Assembly 5 of ZPR-6

for the entire core, then the extrapolated increase in reactivity due to bunching of U^{238} is 218 lh. This increase in reactivity corresponds to a 5.6% decrease in the critical mass (87.2 kg).

b. The Effect of Varying σ_p of U^{238} on Sodium-void Coefficient. The sodium-void coefficient was measured in the central 18 drawers with the loading patterns shown in Figs. 5, 6, 7, and 9. In each case the sodium was removed from the front 4 in., from the front 8 in., and from the entire axial length (28 in.) in each half of the reactor. The difference between the loading patterns in Figs. 7 and 9 is simply that the bunched U^{238} was moved away from the sodium cans by inserting a 1/8-in. plate of graphite between these two materials. The results are given in Table XV. It is to be noted that the difference in the sodium-void coefficient obtained with the loading patterns of Figs. 6 and 7 is mainly due to the decrease in σ_p of U^{238} for the bunched case. Preliminary calculations support this conclusion. The difference between the sodium-void coefficient obtained with the normal loading (see Fig. 5) and that obtained with the reference case (see Fig. 6) cannot be accounted for on the basis of variation in σ_p of U^{238} . It is not expected that the value of σ_p for the loading pattern in Fig. 6 would be lower than that of the normal loading. It is conceivable that the streaming effect in the reference case is more prominent than in the normal loading because the sodium cans in all three drawers of the reference case are bunched. The streaming effect will be investigated shortly.

Fig. 9

Loading Pattern Used in Measuring Sodium-void Coefficient in Assembly 5 of ZPR-6

TABLE XV. Dependence of Sodium-void Coefficient on Loading Pattern

Section Voided (kg)	Sodium Removed (kg)	Loading Pattern	Reactivity Effect of Sample (Ih)
Front 4 in.	1.772	Normal Loading (Fig. 5)	$+2.21 \pm 0.20$
	1.955	Reference Case (Fig. 6)	$+1.51 \pm 0.10$
	1.955	Bunched Case (Fig. 7)	-0.017 ± 0.30
	1.955	Position Effect of Bunching (Fig. 9)	$+0.10 \pm 0.30$
Front 8 in.	3.727	Normal Loading (Fig. 5)	$+4.11 \pm 0.20^a$
	3.91	Reference Case (Fig. 6)	$+2.70 \pm 0.24$
	3.91	Bunched Case (Fig. 7)	$+0.13 \pm 0.20$
28 in.	13.50	Normal Loading (Fig. 5)	-3.95 ± 0.20
	13.62	Reference Case (Fig. 6)	-10.17 ± 0.02
	13.62	Bunched Case (Fig. 7)	-16.97 ± 0.03
	13.62	Position Effect of Bunching (Fig. 9)	-17.11 ± 0.55

^aThis number is the sum of two adjacent 4-in. sections.

There was no significant effect on the sodium-void coefficient when the loading pattern of Fig. 7 was replaced by that shown in Fig. 9.

c. Environmental Effect on the Bunching of U^{238} Plates. The environmental effect, which is primarily due to the Dancoff interaction, was investigated by: (i) measuring the reactivity change associated with bunching the U^{238} in only the central drawer of each half with the surrounding drawers having the loading pattern shown in Fig. 6; (ii) repeating the same measurement with the bunched U^{238} in the surrounding 8-drawer square ring per reactor half; and (iii) performing the same measurement once again with the surrounding 8 drawers unbunched but with a square ring (16 drawers per half) adjacent to the 8-drawer ring, bunched. This square ring was one drawer away from the central drawer. The reactivity change due to the bunching of the two central drawers alone was $+1.56 \pm 0.08$ Ih. The reactivity change that resulted from bunching the two central drawers when the surrounding 8 drawers per half were bunched was $+2.02 \pm 0.07$ Ih. The reactivity change that resulted from the last case was $+1.54 \pm 0.07$ Ih. These results show that the effect of bunching of the U^{238} in the 8 drawers per reactor half square ring adjacent to the central drawers increased the reactivity due to the bunching in the two central drawers by about 25%. Such an effect was totally absent when the bunching was in the ring that was one drawer away from the central drawers.

The same types of measurements were performed to determine the environmental effects on the sodium-void coefficient in the two central drawers. It was necessary to remove all the sodium from the two central drawers in order to effect a measurable change in reactivity. The effects from the three different environments, described earlier, on the sodium-void coefficient were not measurable. Further examination of this phenomenon indicated that the worth of the sodium in the bunched two central drawers was largely due to the leakage component since the spectral component at center, as seen from Table XV, was nearly zero. The environmental effect on the leakage component of the sodium-void coefficient is small (it is a second-order effect) and hence more difficult to measure with the present technique.

The uncertainties in the measured reactivity values are based on the computed variance in the data and the propagation of error. The values reported are the difference in averages of at least two data runs and two reference runs. In some cases when the reproducibility was poor (approx 0.4 lh), four or five runs were performed. Interpolation between reference runs to account for what seemed to be systematic reactor drift was not used because the reactivity values were not reproducible within the uncertainty derived from such techniques.

d. The Effect of σ_p on Fission and Capture Rates. The effect of varying σ_p of U^{238} on the capture and fission in U^{238} and fission in U^{235} was investigated by measuring these quantities in the loading patterns of Figs. 6 and 7. Depleted uranium foils, 5 mils thick by $5/8$ in. in diameter, were placed in such a manner that there was a foil next to every side of every plate in the three-drawer cell. The foils were positioned at an axial distance of 2 in. from the center of the core in one half of the assembly. The bottom edges of the foils were touching the stainless steel drawers. Similarly, the U^{235} foils, 5 mils thick by $3/8$ in. in diameter, were placed in the other half of the assembly at a comparable position with respect to the U^{238} foils from the center of the core. The foils were irradiated for one hour at about 50 W. Five hours after the irradiation the activities of the U^{238} foils were counted as function of time for fission fragments by counting gamma rays with energy of about 0.41 MeV and for capture by counting the 106-keV gamma rays from excited Pu^{239} in coincidence with X rays produced by internal K-conversion electrons. The 5-hr waiting period was necessary to allow sufficient time for the buildup of Np^{239} from U^{239} . The U^{235} foils were counted as a function of time for fission fragments. After counting, a few foils of each type were radiochemically analyzed for the absolute number of fissions and captures in the U^{238} foils and for the absolute number of fissions in the U^{235} foils. The radiochemical results were used in conjunction with the results from the counting techniques to obtain efficiency factors to convert the rest of the data to absolute numbers. The results obtained with the loading pattern of Fig. 7 (bunched case) are reported here. The results obtained with the loading pattern of Fig. 6 are presently being analyzed.

Figure 10 shows the absolute fission rates per gram of U^{235} as a function of position in the three-drawer cells. The relative uncertainty in each point due to the inherent counting statistics is 0.2%. The absolute uncertainty which is based on the statistics of the radiochemical analysis is about 2%. It is noted from Fig. 10 that there are more fissions in the graphite regions than anywhere else and that the fission rates in the bunched U^{238} are at a minimum.

Fig. 10. Absolute Fission Rates per Gram U^{235} in Bunched U^{238} Case of ZPR-6 Assembly 5

Figure 11 shows the fission rates in U^{238} as a function of position. The peaks occur in the vicinity of U^{235} fuel plates.

Fig. 11. Absolute Fission Rates per Gram U^{238} in Bunched U^{238} Case of ZPR-6 Assembly 5

Figure 12 shows the absolute capture rates per gram of U^{238} . The size of the flag on the data is due to the error in the absolute measurements. The relative error is typically 0.2%. It is noted from Fig. 10 that there are considerable dips in the capture rates within the bunched U^{238} .

Fig. 12. Absolute Capture Rates per Gram U^{238} in Bunched U^{238} Case of ZPR-6 Assembly 5

Figures 13 and 14 show the ratios of U^{238} captures to U^{235} fissions and of U^{238} fissions to U^{235} fissions as a function of position, respectively. It is noted that these ratios vary as much as 30% within the three-drawer cell.

Fig. 13. Absolute Number of U^{238} Captures per U^{235} Fission in Bunched U^{238} Case of ZPR-6 Assembly 5

Fig. 14. Absolute Number of U^{238} Fissions per U^{235} Fission in Bunched U^{238} Case of ZPR-6 Assembly 5

3. ZPR-9

Results of Doppler measurements made in Assembly 16, the third in a series of polyethylene-softened assemblies, indicated that a fourth and final assembly should be built to conclude the investigation into the systematics of the U^{235} Doppler coefficient.

In this assembly (Assembly 17) the 10.6 v/o depleted uranium of Assembly 16 will be replaced with carbon. The final material composition will be 8.7 v/o polyethylene, 30 v/o carbon, 42.5 v/o sodium, 15.1 v/o stainless steel, and 3.5 v/o 93% U^{235} enriched fuel.

Preliminary U^{235} Doppler measurements have been made in a small-zoned version of the full-size assembly. These results indicate that the U^{235} Doppler coefficient will be strongly negative in Assembly 17. The full-size version of Assembly 17 is in the process of being installed on the ZPR-9 machine.

4. ZPPR

The construction contractor has poured the concrete for the cell wall up to the lower edge of the ring beam. The concrete roof of the fuel-storage vault-workroom interior equipment room area was poured. The reinforcing steel is being placed for the remaining portions of the access corridor, the blanket-storage room, the emergency escape corridor, and

the connecting corridor to the support wing. The west wall of the support wing has been installed, and work is continuing on the north and south walls. The placing of backfill, wall finish, lighting fixtures, and ventilation ducts is continuing.

The preliminary processing of the sand for the cell roof at the Central Facilities has been completed. The sand has been transported to Idaho Falls for washing and final grading. An additional 30 cu yd are being processed concurrently for the ZPR-6 and ZPR-9 emergency filters.

The wood impact shield has been placed above the bed and tables which are stored in the reactor cell. A fire-detection system is being prepared for installation in the temporary storage area of the bed and tables. The waterproof covering of the bed and tables was removed and the units were inspected for rust, since there has been evidence of free moisture inside the storage structure. The critical areas of the bed and tables appear to be free of corrosion.

The Final Safety Analysis Report is in its final stages of preparation. The accumulation of data and equipment manuals for the operating manual has been started.

The present status of the reactor components which have been received this month or have not yet been received is as follows:

Matrix Drawers. The narrow front and rear drawers have been received and accepted. This completes the matrix drawers.

Poison Rod Drives. The vendor is checking and debugging the assembled rod drives. An ANL representative is scheduled to visit the vendor the first week of May.

Personnel Shields. The vendor has received all components for the personnel shields except the tractor drives. Assembly of the shield units has begun.

Matrix Alignment. The castings of the alignment plates have been poured. Machining of the first plate has begun.

Source Drives. The source drives have been assembled and checked by the manufacturer. An ANL representative is scheduled to inspect the assembled units at the vendor's plant during the first week of May.

Rod Drive Mounting Plates. Because it was necessary to repour the second plate, the scheduling of machining has been delayed for four weeks. Attempts are being made to reduce this delay.

Fuel Control Drawers. The purchase order to the new vendor, Stainless Steel Equipment Company of Denver, Colorado, was issued on March 31. Delivery is scheduled for July.

Poison Safety Blade Clevises. The vendor is reworking the clevises.

Boron Fabrication. The vendor is assembling a new furnace capable of the 2000°C operating temperature necessary for pressing the boron carbide plates.

Stack Particulate Monitor. The stack monitor has been repaired, checked out, and accepted.

An abbreviated mockup of the ZPPR matrix, using the knees and 5 columns of tubes, 43 tubes high, has been assembled to check the deflections due to core loads. The clamping system performed as designed and deflections of the clamped tubes under load were under 0.010 in. The mockup will also be used to checkout the installation procedures for the rod drives and experimental equipment.

5. Plutonium Fuel Procurement

a. Programmatic Requirements. More than 3600 kg of plutonium is required to measure criticality and neutronic effects in very large reactor assemblies to be built in ZPPR and in ZPR-6 and ZPR-9. About 2900 kg of the plutonium are being fabricated as U-28.3 w/o Pu-2.5 w/o Mo alloy plates, which will be jacketed in stainless steel. The plutonium will be 88.3% fissile ($\text{Pu}^{239} + \text{Pu}^{241}$) and 11.5% Pu^{240} . An additional 200 kg of plutonium will be derived from highly irradiated fuel compositions. This material, which will contain increased amounts of Pu^{238} , Pu^{240} , Pu^{241} , and Pu^{242} , will be fabricated into metal plates, oxide-loaded elements, and instrumented test specimens, which will be used to measure the effect of plutonium isotopic composition. There is an additional requirement for 500 kg of plutonium containing 11.5% Pu^{240} to be manufactured into (U,Pu) O_2 and (U,Pu)C pellet-loaded tubes and plates to study light-element dilution effects. These elements may also be used to confirm temperature-related effects, which are also being studied in Doppler effect test elements.

b. Plutonium Ingotting. The critical and neutron effect experiments require fuel of a closely controlled isotopic composition. The plutonium which is available to the program from several different sources varies in composition. The work of blending and ingotting this plutonium to the required isotopic composition is being done under contract with Isochem Inc. at Richland, Washington. Three-kilogram batches of plutonium are being melted and cast into ingots of about one kilogram each. Isotopic and spectrochemical analyses are being furnished with each batch of plutonium blended. A total of 806 kg of plutonium was blended to $11.50 \pm 0.50 \text{ Pu}^{240}$ by

May 1, and the analyses were completed on about two-thirds of the material reserved for fabrication at Rocky Flats. An additional 600 kg of plutonium are being blended during FY 1967, which will be reserved for the first commercial fabrication of the fuel elements. The sampling and ingot identification of this material is being witnessed by an observer from Battelle Pacific Northwest Laboratory.

c. Fuel-element Fabrication. About 300 kg of plutonium will be required as U-28.3 w/o Pu-25 w/o Mo fuel elements for an experiment scheduled in October-November 1967 and an additional 1100 kg must be received for ZPPR experiments by March 1968. In order to assure that fuel will be available for these experiments, an order has been placed with the Rocky Flats Division of the Dow Chemical Company for delivery of elements containing 700 kg of plutonium by May 1967. This company had previously made zero-power reactor elements and the fabrication can start quite quickly. Fabrication is scheduled on a single shift so that, if commercial fabrication should be delayed, the total fuel requirement could be met at Rocky Flats with two- or three-shift production. The planning and setup of fuel-element production equipment was nearly complete by May 1, and experimental production of fuel elements is scheduled to begin at Rocky Flats during May 1967. The processes to be used will be essentially those that were developed at ANL, and arrangements have been made for close ANL supervision of the production.

Negotiations have been completed with one commercial source, to establish a contract for part of the alloy fuel-element production, and detailed contract recommendations will be submitted to AEC-CH for the commercial fabrication of ZPPR fuel elements during the first week in May.

The plutonium alloy plates are jacketed in rectangular-section, stainless steel sleeves with welded end plugs. Vacuum-melted, Type 304L stainless steel has been fabricated into 1.350-in.-dia x 0.016-in.-wall-thickness tubing by Pacific Tube Company. Nine-hundred feet of this tubing were inspected by ANL representatives and approved for shipment to ANL. Difficulty was experienced in obtaining a commercial source for the forming of jacket sleeves to ZPPR tolerance requirements. Production tooling for stretch forming this tubing into rectangular sleeves has been completed in ANL shops, and an order has been placed for the first 8000 jacket sleeves, which will be used for the immediate requirement and will serve to demonstrate to commercial fabricators that the jacket sleeves can be fabricated as specified. The end plugs are being cut from precision-formed rectangular wire drawn by Major Wire Company.

C. Other Fast Reactor Physics

1. Plutonium Air Monitor for Use in ZPR Areas

A plutonium air monitor has been designed which utilizes the high sampling rates of the impactor monitor² and has the pulse-height discrimination advantages of low-sampling-rated filtration monitors.³ In the new design the dust is impacted directly onto the face of a 2π passivated solid-state detector.⁴ Alpha emissions due to plutonium are discriminated from those due to radon-thoron daughter products or due to uranium by a single-channel analyzer. A second channel is used to follow the higher-energy-alpha emissions of the radon-thoron daughter products so that background corrections are made in the plutonium measurement.

The dust accumulation on the solid-state detector is removed by wiping once a day or more frequently. A thin film of grease is used on the detector face to cause the impacted particles to adhere. No degradation in resolution has been encountered after two weeks of initial "life" tests, and there is nothing that would indicate such degradation would be expected in the near future. "Life" tests are continuing.

D. Component Development

1. Sodium Technology and Development

a. Fission-gas Pressure Transducer. To obtain an indication of the fission-gas pressure buildup in individual fuel pins while they are operating in the reactor, commercially available pressure transducers are being reviewed and evaluated for conditions expected in the Fast Flux Test Facility (FFTF), and will be tested out-of-pile and in-pile.

(i) NaK-filled Capillary System. A prototype transmitter of this type was built and tested. It has a 0.5-in.-dia stainless-steel welded bellows welded to a 1/8-in.-OD by 1/16-in.-ID 20-ft-long capillary tube leading to a 300-psi pressure transducer. The system was evacuated at the transducer end and backfilled with NaK; the bellows end was sealed in a pressure chamber supplied with 150-psig argon.

The 20-ft capillary tube was wound in a 2.5-ft dia coil; a system curve of voltage output from the transducer to gauge pressure was generated. The tube then was unwound and another test curve was obtained. The two curves checked, indicating that bends in the capillary tube did not affect accuracy.

²Tait, G. W. C., *Nucleonics* 14, 53-55 (1965); Alexander, J. M., *Health Physics* 12, 553-556 (1966).

³Phillips, W. A., and Lindeken, C. L., *Health Physics* 9, 299 (1963); Hazards Control Quarterly Report 4, 10-15, UCRL-14351 (1965).

⁴Simtec, Ltd., 3400 Metropolitan Blvd. East, Montreal 38, Canada, 200 mm² and 500 mm².

The pressure chamber containing the bellows was placed in a furnace and heated to 1200°F. System thermal-expansion effects showed a new positive pressure on the transducer of 0.2 mV. Although the exact curve of this zero shift versus temperature has not been obtained yet, the data indicated that 8 mV is worth 100 psi. Therefore, on a linear scale, 0.2-mV zero shift is equivalent to 2.5 psig. A 9-ft length of capillary tube will be heated to 1200°F to determine effect on calibration.

The bellows end has been heated to 1200°F many times; the consistency of voltage output versus pressure is very encouraging and indicates that accuracy can be within $\pm 1/4$ psig. Temperature effects are not serious if the working range of bellows temperature is known.

This system can be miniaturized to a 1/4-in. bellows. Smaller size would be quite difficult because of the NaK expansion effects and the inability of the bellows to expand sufficiently. Thus, if accuracy is to be maintained despite limited bellows expansion, when a smaller bellows is used, the capillary tube must be smaller also. For diameters less than 1/4 in., electrodeposited bellows made of stainless steel are not available; they probably would have to be made of nickel. Further investigation is underway.

(ii) Strain-gauge Systems. Work has continued on the Statham Model-PA812 thin-film transducer (see Progress Report for February 1967, ANL-7308, p. 23). Examination of the bridge portion indicates the the gold wire was in almost intimate contact with the header support, which probably was what caused the insulation resistance to drop to ~3 kilohms. When the wires were reoriented again, the insulation resistance increased to over 200 megohms. A deposit of gold was noted on and near the header-pin support.

The transducer bridge has been reconnected to the lead wires, and tests of the bridge will continue above 600°F to determine whether the sagging wires were, in fact, the cause of the previously observed permanent change in insulation resistance. During the previous tests, no increase in bridge resistance was measured, thus indicating little, if any, oxidation. Any evidence of strain-element oxidation will be observed by continuously checking the bridge resistance as well as the insulation resistance.

b. Failed-fuel Locator. To find a method for determining which subassembly within a shutdown reactor core has one or more failed fuel pins, the feasibility of sensing the fission-gas release from the subassembly is being investigated. To prove the principle, a scheme will be demonstrated for efficiently disengaging noble gases from a continuously circulating liquid-sodium system at 1200°F. The equipment will include devices for disengaging the gas, trapping sodium vapor, viewing, monitoring radiation, and for injecting gas.

Work has continued on modifications to an existing sodium vapor-trap flow system (see Progress Report for February 1967, ANL-7308, pp. 23-24) that will be used to evaluate methods for detecting small quantities of simulated fission-product gas. The sodium-supply tank was filled with sodium, new heaters were installed, and new insulation was applied; temperature-control wiring remains to be installed.

The literature is being surveyed to gain a better understanding of the adequacy and operability of previously designed methods for detecting and locating failed fuel cladding. For liquid-cooled reactors, most such detectors must contend with interference radiation from corrosion products, coolant impurities, system contaminants, and the coolant itself, which tend to mask the signal being sought. A considerable portion of most such detector systems is devoted to the reduction of the undesirable background radiation by means such as delay lines, ion exchanges, gaseous-fission-product stripping from liquid coolants, electrostatic precipitation, and gas chromatography, coupled with energy discrimination or spectrographic analysis of the output from a wide variety of detection instruments.

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

The rebuilt test tank and associated equipment have been checked out and made operational. Performance appears satisfactory; difficulties in sodium transfer, system tightness, and repeatable measurements have been overcome. A prototype connector assembly is being tested. The test consists, mainly, of cycling temperature between 250 and 600°C while the connector is submerged continually in sodium. Leakrates have been measured at various temperatures; it appears that a correlation between leakrate and rate of change of temperature can be established. While the leakrate averages about 2×10^{-2} std cm³ of argon/sec, variations ranged up to about twice this value during the first 7 days of this test.

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

(i) Thermal and Mechanical Tests of Magnet Materials.

Thermal tests of the initial Alnico-V rod magnet having a length-to-diameter ratio of 6 have continued (see Progress Report for February 1967, ANL-7308, pp. 24-25, and for March 1967, ANL-7317, p. 48); this magnet is designated as sample A5-1. It has accumulated more than 1100 hr at 1200°F and there is now a definite indication that the magnetic flux has departed from the initial value; the exact cause of the change remains to be determined.

Most of the Alnico-V and some of the Alnico-VI and -VIII rod magnets for the out-of-pile and in-pile tests have been delivered, so tests can proceed to determine the irreversible and material effects at temperatures up to 1200°F for these magnets having various length-to-diameter ratios. The change in the shape of the hysteresis loop due to material effects in these alloys will also be determined at room temperature.

So that reversible changes in magnet properties can be measured continuously at high temperatures by use of a magnetometer, a turntable has been designed to swing the test furnace, with the magnet inside, slowly and smoothly through 180°, thus providing two tests positions. The furnace apparatus is being fabricated.

Five magnet manufacturers were requested to quote on supplying typical flowmeter magnets of cast Alnico-VI and -VIII alloys. Only one quotation for Alnico-VI was received; all five vendors declined to quote on the Alnico-VIII magnets. Two vendors who gave reasons for declining to quote cited orientation and wall thinness for the length; another vendor declined because of the small quantity involved. Further discussions are underway to learn whether there is a configuration of Alnico-VIII that can be manufactured suitable for flowmeter application.

(ii) Magnetic Flowmeter Prototype. The preliminary prototype permanent-magnet flowmeter has been installed in the calibration loop and the loop has been made operational. Preliminary flowmeter calibrations have begun in preparation for tests of signal long-term stability.

(iii) Magnet Irradiation Tests. The capsule configuration for irradiating rod magnets in EBR-II has been designed. Sample temperatures of 1200°F can be achieved easily; thermal-radiation shields in the capsules will be used to adjust sample temperature to desired levels. To determine sample test temperatures, wires having selected melting points will be installed in the capsules. Rod magnets are being encapsulated in stainless steel cans, and stabilization of the Alnico-V magnets is about to commence. In addition to the magnet samples, specimens of Type 304 stainless steel will be irradiated in some capsules to determine magnetic changes caused by the combined radiation and high-temperature environment.

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

The spectrochemical analysis of the 99.9%-vitrified thoria insulators shows that purity is satisfactory. Coaxial resistivity probes have been assembled with these insulators and tantalum electrodes; preparations have been made to measure the resistivity of the vitrified thoria in a high-temperature furnace.

Bare-wire W-3%Re/W-25%Re thermocouples have been assembled for long-term tests of thermoelectric stability in the high-temperature furnace.

For study of the compatibility of tantalum and W-25%Re with UO_2 , the capsule (designated TU-1) that had been tested previously (see Progress Report for March 1967, ANL-7317, p. 49) at 2000°C for ~33 hr is being examined metallographically. An identical tantalum capsule designated TU-2, has been assembled and is being tested in a 1-spig dry-helium environment at 2400°C; the exposure goal is 720 hr.

2. Liquid Sodium Coolant Chemistry

a. Dispersion of Carbon in Liquid Sodium. In connection with the studies of the mechanism of carburization and decarburization of metals in liquid sodium, efforts to prepare dispersions of carbon in liquid sodium are continuing.⁵ Carbon-sodium pastes are being prepared, since the carbon in such pastes has been wet by the liquid sodium and wetting of the carbon is considered to be a prerequisite to achieving stable dispersions. A minimum temperature of ~370°C was found to be necessary for liquid sodium to wet 90-Å carbon black.

A carbon-sodium paste with the nominal composition 9.5 w/o carbon was prepared by adding 90-Å carbon black (7.4 w/o oxygen) to liquid sodium at ~450°C. Samples of the paste were taken through 5μ-porosity stainless steel filters. Analysis of the samples showed no significant concentration of carbon and, therefore, that the 90-Å particles had agglomerated. Reduction of the oxygen content of the carbon black to 0.3 w/o (by vacuum-heating) did not alter the results.

⁵For previous reports, see Reactor Development Program Progress Reports for January 1967, ANL-7302, p. 41; November 1966, ANL-7279, p. 20; September 1966, ANL-7255, p. 25.

Portions of a sodium-carbon paste were added to a large batch of sodium (final nominal composition ~740 ppm carbon) being stirred at ~1000 rpm in a blender at 150°C. The stirrer speed was increased to 14,000 rpm during several mixing periods, after each of which the mixture was allowed to settle for about 5 to 10 min and then sampled. Analysis of the samples for carbon showed that the agglomerated particles had not been dispersed. The mixture was allowed to settle for 28 hr during which it was sampled periodically. From the carbon analyses and calculations based on the rate of sedimentation, it is concluded that the carbon agglomerates were $\geq 10\mu$ in diameter.

E. Fuel Development

1. Metal Fuels

a. Irradiation of Uranium-Plutonium Alloys. Neutron radiographs of two elements, fueled with U-15 w/o Pu-10 w/o Zr and with U-15 w/o Pu-10 w/o Ti in separate jackets of V-20 w/o Ti (see Progress Report for March 1967, ANL-7317, p. 50), indicated fuel-pin elongations of 3.6 and 8.8%, respectively, after irradiation in EBR-II to about 5 a/o burnup at maximum jacket temperatures of 540°C. Although both fuel jackets appeared intact, nearly one-fourth of the elongation of the titanium-bearing fuel was around and above the lower part of the upper fuel restrainer.

Accordingly, fuel pins of U-15 w/o Pu-10 w/o Zr only were jacketed and were being encapsulated for irradiation to higher burnups. Known as Group M-3 elements, the jacketed fuels, fifteen in number, have now been encapsulated and delivered to EBR-II. The several jacket materials are V-20 w/o Ti, Type 304 stainless steel, and Type 316 stainless steel. These elements will be irradiated along with four encapsulated EBR-II Mark-IA production fuel elements (the total makes up a full experimental subassembly).

The M-3 elements are similar to the XA07 elements, which were irradiated to 4.6 a/o burnup. The contemplated reactor exposure of the M-3 Group will result in maximum jacket temperatures near 600°C.

Group M-3 will be removed at 10 a/o burnup, but the Mark-IA fuel will operate at jacket temperatures near 500°C and will be removed at 3 a/o burnup. Table XVI lists the specimen and jacket dimensions of Group M-3.

TABLE XVI. Group M-3 Specimen and Jacket Dimensions

Fuel Material: U-15 w/o Pu-10 w/o Zr (93%) enriched
 Fuel Dimensions: 13.5 in. x 0.144 in. in diameter

Capsule/Specimen Number	Cladding			Specimen Length (in.)	Plenum Length (in.)	Specimen Plenum Volume (cc) (RT)
	Material	ID (in.)	Wall (in.)			
ANL-BA-01	304 SS	0.166	0.015	33.00	17.97	6.51
ANL-BA-02	304 SS	0.166	0.015	33.00	17.97	6.51
ANL-BA-03	304 SS	0.166	0.015	33.00	17.97	6.51
ANL-BA-04	304 SS	0.166	0.015	33.00	17.97	6.51
ANL-BA-05	304 SS	0.166	0.015	32.61	17.57	6.36
ANL-BB-01	304 SS	0.166	0.020	29.80	14.76	5.35
ANL-BC-01	316 SS	0.166	0.015	33.00	17.97	6.51
ANL-BC-02	316 SS	0.166	0.015	33.00	17.97	6.51
ANL-BC-03	316 SS	0.166	0.015	33.00	17.97	6.51
ANL-BD-03	V-20 Ti	0.166	0.015	24.68	9.65	3.49
ANL-BD-04	V-20 Ti	0.166	0.015	24.68	9.65	3.49
ANL-BD-05	V-20 Ti	0.166	0.015	24.68	9.65	3.49
ANL-BE-01	V-20 Ti	0.166	0.015	22.70	7.67	2.77
ANL-BE-02	V-20 Ti	0.166	0.015	22.70	7.67	2.77
ANL-BE-03	V-20 Ti	0.166	0.015	22.70	7.67	2.77

2. Carbide Fuels

a. Compatibility of (U,Pu) Carbides with Potential Jacket Materials

(i) (U,Pu) Carbides versus Hastelloy-X. No reaction was observed between Hastelloy-X and $(U_{0.8}Pu_{0.2})C$ after heat treatment at 700°C for 4000 hr. Hastelloy-X and a hyperstoichiometric (U,Pu)C composition containing approximately 20 v/o $(U,Pu)_2C_3$ form a reaction zone 4 μ in depth. The 4 μ average reaction zone consists of a 2 μ nickel-rich zone on the fuel side of the interface plus a 2 μ chromium-rich zone on the jacket side. Although these zones are too small for quantitative analysis, the nickel-rich zone is very probably the compound $(U,Pu)Ni_5$, identified by electron microprobe analysis in a couple containing hyperstoichiometric (U,Pu)C versus Inconel 625.⁶ UNi_5 has been shown to be the principal product formed in a diffusion couple of UC versus Ni.⁷ The extent of reaction was equal to that observed previously (see Progress Report for June 1966, ANL-7230, pp. 17-18) at this temperature for 1000 hr.

Another test involving Hastelloy-X with a (U,Pu) carbide composition containing 6.75 w/o equivalent carbon and consisting of approximately 90 v/o $(U,Pu)_2C_3$ substantiates the finding that the sesquicarbide phase is the only phase that has reacted in the 700°C tests of hyperstoichiometric (U,Pu)C. Examination of a compatibility couple containing this

⁶ANL Metallurgy Division Monthly Report, March 1967, pp. 55-56.

⁷Anselin, F., Colais, D., and Passefort, J. C., Etude du Diagramme Ternaire Uranium-Carbone-Nickel, CEA-R 2845 (Aug 1965).

composition and heat treated for 1000 hr revealed a 4μ reaction layer and also an adjacent 8μ layer in the fuel that was reduced from sesquicarbide to monocarbide. This observation provides an explanation, in the case of the hyperstoichiometric carbide composition, for (1) no increase in the size of the reaction zone after 4000 hr as compared with 1000 hr, and (2) the absence of sesquicarbide grains at the Hastelloy-X interface, since these grains were all less than 10μ in diameter.

(ii) $(U_{0.8}Pu_{0.2})Fe_2$ versus Type 304 Stainless Steel. Although iron additions to hypostoichiometric UC are reported⁸ to result in improved compatibility with stainless steel through reaction with the excess uranium metal phase to form UFe_2 , no direct compatibility test with this compound has been reported.

Attempts to fabricate $(U_{0.8}Pu_{0.2})Fe_2$ pellets were previously reported (see Progress Report for November 1966, ANL-7279, p. 27). After recrushing and repressing these pellets, and firing at $1100^\circ C$, an average geometrical density of 9.45 g/cm^3 (72% of theoretical) was obtained; this was approximately equal to that of the earlier work. The pellets were essentially single-phase; chemical analysis showed a slight excess (0.4 w/o) of iron over that required for stoichiometry, and metallography revealed a small amount of second phase appearing as discrete particles.

Some reaction with the tungsten setter plate was observed. An unidentified phase was found in the tungsten to a depth of 245μ in the $(U,Pu)Fe_2$.

After grinding the ends of the pellets to remove the reaction product, this material was held in contact with Type 304 stainless steel at $800^\circ C$. After 1000 hr, only a small layer of about 2μ was observed at the interface.

b. Performance Studies of Mixed-carbide Fuel. Postirradiation examination of three mixed-carbide elements continued in the Metallurgy Alpha-Gamma Hot Cell (see Progress Report for March 1967, ANL-7317, pp. 54-55). An additional 15 elements remained in EBR-II for continued irradiation.

Metallographic examination continued with the sectioning and preparation of element No. SMV-1. In contrast to the sintering behavior in solid-solution $(U-20\text{ w/o Pu})C$ in element No. VMV-1, the physically mixed $UC-20\text{ w/o PuC}$ in element No. SMV-1 exhibited no indications of sintering even in the hottest area of the fuel column. At a location in VMV-1 comparable to the maximum temperature in SMV-1, the sintered diameter of solid-solution fuel was approximately 0.078 in. or 30% of the diameter of the fuel.

⁸Herbst, R. J., and Jordan, K. R., Compatibility of Modified Monocarbide Fuels with Stainless Steel, Trans. Am. Nuc. Soc. p. 17 (June 1966).

Fission-product gas release from the solid-solution (irradiation sintered) fuel in VMV-1 was smaller than that from the physically mixed fuel in SMV-1. Preliminary results were reported in ANL-7317. Values corrected for analytical results are as in Table XVII.

TABLE XVII. Fission-gas Release from Mixed-carbide Fuel

Specimen Number	Fuel Description	Gas Release (% of Theoretical)	Max Est (kW/ft)	Burnup (a/o)
SMP-1	Solid solution, pellets	4.1	18.9	2.1 ^a
VMV-1	Solid solution, vibratory compacted	8.6	29.5	3.02
SMV-1	Physically mixed, vibratory compacted	13.4	25.3	2.59

^aCalculated--to be measured.

A continuous profilometer scan of the surface contour of element No. SMP-1, containing pellets of solid solution (U-20 w/o Pu)C, did not reveal ridges or irregularities that would correlate with pellet lengths. Gamma-scan indications interpreted to be separations between pellets could not be repeated when the element was reversed (end-for-end) in the vertical scanning device. Neutron radiography subsequently showed five or six loose pellets at the top of the column and one pellet fractured in the transverse direction.

c. Processes for Large-scale Preparation of (U,Pu)C. A fluidized-bed process for the preparation of (U,Pu)C has been demonstrated in 15 runs in which high-purity (U,Pu)C was prepared on a scale of 300 to 600 g per batch by hydriding U-15 w/o Pu alloy and then carburizing by reaction with methane-hydrogen fluidizing gas.⁹ Carbide fuel for irradiation testing is also being prepared by this technique. This process appears promising for large-scale production of carbides; calculations show that a single fluidized-bed reactor of 8-in. dia could prepare 100 kg/day of (U,Pu)C if operated at 10 atm pressure and 800°C. However, some work remains to be done to perfect this process for large-scale synthesis of (U,Pu)C.

To study problems of scaling up the fluidized-bed process, a semiworks-scale fluidized-bed reactor with an internal diameter of $2\frac{1}{2}$ in. is being operated for preparation of UC on a scale of 1 to 2 kg per batch. The reactor was designed to be operated in an air environment [the fluidized-bed reactor for (U,Pu)C preparation is operated in a helium-atmosphere glovebox]. It is also designed to discharge the product by conveying it

⁹See Progress Report for December 1966, ANL-7286, p. 26, and Progress Report for February 1967, ANL-7308, p. 31.

pneumatically through a discharge tube to a product receiver that can be removed and opened in a helium-atmosphere glovebox. The reactor and its operation with once-through gas flow were described in the Progress Report for July 1966, ANL-7245, p. 8. With once-through gas flow the operating pressure was limited to about 2 atm because of the large flow of gas required at higher pressures. A diaphragm compressor was therefore installed to recirculate the off-gas from the fluidized bed and allow operation at the reactor design pressure of 10 atm.

Uranium in the form of acid-cleaned rods, 3/8-in. in diameter by 1 in. long, is charged to the reactor through a lock which prevents the admission of air to the system. The uranium metal is reacted with hydrogen at about 250-300°C for 0.5 to 1 hr to produce finely divided uranium hydride (UH₃), which decomposes to uranium metal as the temperature is raised. The metal particles are then reacted with hydrogen-methane gas at 4 to 10 atm pressure and 600 to 800°C. The reaction of uranium metal with methane yields two moles of hydrogen for each mole of uranium reacted. The excess gas is released from the system through a back-pressure regulator; the rate of release is measured with a wet-test meter.

The methane concentration is automatically controlled by an infrared analyzer coupled with a recorder-controller. The amount of methane added to maintain the concentration is determined by the decrease in pressure in a small supply tank.

Analyses of the products of the four latest experiments (see Table XVIII) indicate that the carbide products have carbon contents close to the stoichiometric value and that low oxygen contents can be achieved. The fluidized-bed reactor is operated in an ambient air atmosphere rather

TABLE XVIII. Preparation of UC in a Fluidized Bed at 10 atm Pressure
(1 kg/batch)

Expt No.	Temp ^a (°C)	Gas Velocity (cm/sec)	CH ₄ Conc ^b (v/o)	Reaction Time (hr)	Product Analysis ^c (w/o)	
					Oxygen	Carbon ^d
16	850	12	5	11	0.4	4.66
17	810	18	5	11	0.094	4.85
18	750	18 ^e	5	14.2	0.11	4.73
19	775	12	2.5 ^f	16.2	0.068	4.24 ^g

^aTemperature was measured at the top of the fluidized bed.

^bBalance of the gas was hydrogen.

^cAverage of two analyses.

^dStoichiometric UC contains 4.80 w/o carbon.

^eGas velocity lowered to 12 cm/sec for two short periods during run (see Fig. 15).

^fCH₄ concentration raised to 5 v/o after 7 3/4 hr.

^gExperiment was terminated before carburizing was complete.

than in a helium-atmosphere glovebox such as is used for the (U,Pu)C preparations cited above. The product is transferred by pneumatic conveyance to a receiver that is subsequently removed and opened in a helium-atmosphere glovebox. The transfer of the product by pneumatic conveyance can usually be carried out by a single transfer, but occasionally repeated transfers are required to remove the product completely.

Fig. 15. Fluidized-bed Reaction of Uranium with Methane-Hydrogen Mixtures (1 kg/batch)

curve and by comparing both curves. It is also of interest to note the enhanced rate of reaction when the powder is rehydrided on cooling the reactor overnight and reheating the next morning. This effect has been noted in the uranium-plutonium experiments and is attributed to the exposure of fresh surfaces as the uranium rehydrides.

One of the goals of the experimental program is to be able to predict the carbon content of the product from the methane utilization during the run. For these experiments, the amount of methane reacted was compared with the chemically determined carbon content of the powder at various stages of the reaction. For the recent experiments, the results calculated from the methane consumption (correcting for methane in the gas released in maintaining the pressure) give a calculated carbon value for the final product which is within 0.1 w/o of the carbon content actually determined by chemical analyses.

After 19 experiments, it was necessary to replace the reactor heaters. At the same time, the reactor was opened and examined for any evidence of deterioration. The reactor vessel itself was used in approximately 10 experiments or for approximately 100 hr at temperature. An X-ray examination of the vessel gave no indication of any deterioration of the vessel wall.

The gas-inlet tube which extends from the top to the bottom of the reactor along the centerline was more extensively examined. The tube

was sectioned and six samples were prepared for metallographic examination. Four of the samples were taken from the lower end of the tube, one from the midpoint of the gas phase, and one at the coldest portion of the tube. There was no evidence of attack in any of the samples except one, which was located near a weld and suffered grain separation to a depth of $1\frac{1}{2}$ miles. This latter sample was examined with an electron microprobe. There was no indication of any alloying with uranium, nor was there any evidence for the accumulation of carbon on the outside wall of the gas entry line.

These results indicate that Type 304 stainless steel can be used as a material of construction for the fluidized-bed preparation of UC by the uranium-methane reaction. The reactors for preparing (U,Pu)C are also constructed of Type 304 stainless steel and they too have shown no evidence of deterioration.

3. Fuel Cladding and Structure

a. Tube-burst Tests of Vanadium Alloys. Although tube-burst tests have been carried out on unirradiated stainless steel (see Progress Report for May 1966, ANL-7219, p. 13) and on stainless steel irradiated at temperatures in the range 371 to 520°C (see Progress Report for January 1967, ANL-7302, pp. 20-21), burst tests were made for the first time on tubes of V-15 w/o Ti-7.5 w/o Cr in the present reporting period.

Eight inner capsule tubes of V-15 w/o Ti-7.5 w/o Cr from sub-assembly XO13 were on hand. These had initially been annealed for one hour at 1100°C. The maximum fluence (at reactor midplane) was 6×10^{21} n/cm², and irradiation temperature was about 600 to 650°C. Each tube was cut into eight sections, about 4 in. long. A few tests have been made on the 4-in. sections at 550, 650, and 750°C. Unirradiated control specimens were also tested. The results are shown in Table XIX. Ductility has been indicated

TABLE XIX. Burst Strength and Ductility of V-15 w/o Ti-7.5 w/o Cr Tubing

Test Temp (°C)	Specimen Location ^a	Max Tangential Stress		Diameter change (% $\Delta D/D$)		Time at Test Temp (min)
		psi	kg/mm ²	Range	Avg	
550	Unirradiated	100,100	70.4	b	b	60
	Unirradiated	101,300	71.3	b	b	60
	-10 to -20 cm	113,800	80.0	0.8-2.3	1.4	130
	0 to -10 cm	128,000	90.0	1.8-2.4	2.0	90
	+50 to +60 cm	110,500	77.7	2.2-3.6	2.8	30
650	Unirradiated	97,800	68.8	7.4-8.8	8.1	45
	-10 to -20 cm	111,000	78.1	2.6-3.2	2.9	35
	0 to -10 cm	112,000	78.8	1.2-1.7	1.4	120
	+50 to +60 cm	88,000	61.9	b	b	60
750	Unirradiated	90,200	63.4	6.9-8.6	7.8	60
	+50 to +60 cm	89,300	62.8	2.2-6.8	3.7	95

^aSpecimen location above or below reactor midplane in cm.

^bShattered or split so that no diameter measurements were possible.

by the percent change in the diameter of the tube. In some cases, the tube shattered to such an extent that no diameter measurements were possible. The time at test temperature is shown because of its possible relation to annealing effects, particularly at the higher temperatures. The variation in the time at temperature resulted from problems in getting a tight system.

At 550°C the maximum increase in burst strength is 27%; at 650°C it is 13%, and at 750°C (one specimen) there is no increase in strength due to the irradiation. At both 650 and 750°C the unirradiated specimens showed about twice the ductility of the irradiated. Presumably the shattered unirradiated specimens at 550°C test temperature had negligible ductility. One specimen at 650°C split open along its entire length.

Bohm *et al.*,¹⁰ indicated that complete recovery of both strength and ductility of tensile specimens irradiated at 50°C occurred with vanadium alloys tested at 650 and 750°C. The results of our biaxial tests are at variance with these data.

b. Development of Refractory-metal Alloys for Service in Oxygen-contaminated Sodium. Small weight gains shown by V, V-5 w/o Cr, and V-15 w/o Ti-7.5 w/o Cr exposed for 88 days to flowing (6.1 m/sec) sodium containing about 7 wt ppm of oxygen were reported (see Progress Report for March 1967, ANL-7317, p. 58) to differ from one another less in tests at 550°C than at 650°C. The same trend prevailed in April after 117 days of exposure, with the oxygen level now found by analysis to be 8 wt ppm (cold trap at 120°C).

The alloys have a faint brown tint, but otherwise appear metallic. The weight gains shown in Table XX are believed to fairly represent the relative pickup of impurities (mainly oxygen) from the flowing stream.

Table XX. Corrosion of Vanadium Alloys in Sodium at 550°C and Flowing with 6.1-m/sec Velocity

Alloy	Weight Gain, mg/cm ² at Indicated Time (days)				
	6.9	20.9	53.8	88.7	116.5
V	0	0.06	0.08	0.16	0.16
V-5 Cr	0	0.032	0.064	0.075	0.075
	0	0.043	0.065	0.086	0.086
V-20 Ti	0.089	0.20	0.25	0.29	0.31
	0.079	0.21	0.26	0.30	0.33
V-15 Ti-7.5 Cr	0.077	0.15	0.22	0.30	0.32
	0.088	0.17	0.22	0.30	0.32
Oxygen level (Hg amalg)	-	7.8	9.7	6.5	8.5

¹⁰Bohm, H., *et al.*, Irradiation Effects on the Mechanical Properties of Vanadium-base Alloys, presented at the Sixty-Ninth Annual Meeting of ASTM, Atlantic City, New Jersey, June 27-July 1, 1966, Paper No. 75.

A newer high-strength alloy (V-15 w/o Cr-5 w/o Ti) became available at the last inspection, and specimens are now included in the continuing test. It is presently planned to terminate the 550°C dynamic testing after an additional two exposures of one month each.

Static (oxygen refreshed) testing at 450°C has started. Samples of various vanadium alloys exposed in low-oxygen sodium (CT 110°C, 1.5 ppm O by distillation) lost a very small amount of weight (less than 0.02 mg/cm²) in 7 days.

In sodium of higher oxygen concentration (CT 175°C, 13 ppm O by distillation) V-20 w/o Ti samples gained about 0.08 mg/cm², while V-15 w/o Cr-5 w/o Ti lost about 0.6 mg/cm² after 6.8-day exposure at 450°C.

c. Effect of Mechanical Properties on Nickel-base Alloys in 650°C Sodium. Nimonic 80A specimens were selected as control material for conducting mechanical property tests in 650°C sodium for Ni-Cr-Fe alloys (see Progress Report for February 1967, ANL-7308, pp. 34-35). These control specimens were machined from doubly annealed stock (1080°C/8 hr/air cooled and 200°C/16 hr/air cooled) and then creep tested at 650°C in commercial tensile machines (Instron), adapted, with special furnaces for testing in low-oxygen sodium and in vacuum at 10⁻⁶ Torr. In the case of sodium, the oxygen content was maintained below 5 ppm by hot gettering with zirconium chips. Oxygen measurements were made using internal-friction techniques. A stress of 46.2 kg/mm² was applied to the specimen after the furnace had been at 650°C temperature for 24 hr.

The results of these tests were that specimens of Nimonic 80A exposed in 650°C sodium prematurely rupture in 212 hr with a true strain of 1.2%, whereas a corresponding specimen in vacuum at 650°C ruptured in 765 hr with a true strain of 4.60%. There was also a 25% reduction in strain rate in the third stage of creep in sodium as compared to that in vacuum, while there was no change in corresponding rates during the first and second stages of creep. These results indicate the surface energy of Nimonic 80A is lowered by liquid sodium penetrating into the grain boundaries of the alloy. Since this test was conducted with no mass flow in an isothermal chamber of Type 304 stainless steel, it can only be used as a screening test for studying the effect of sodium exposure on mechanical properties of Ni-Cr-Fe alloys under the specific test conditions. A program (and the necessary equipment) for a more meaningful study has been proposed.

d. Effect of High-temperature Sodium on Type 304 Stainless Steel. Deleterious effects of 650°C sodium on stainless steel, as evidenced by accelerated attack of Strauss reagent or aluminum sulfate solution on sodium exposed samples has been reported (e.g., Progress Report for January 1967, ANL-7302, pp. 42-45). In cooperation with the Reactor Engineering Division, the Applied Corrosion Group of the Metallurgy Division is trying to determine the cause of these effects.

Preliminary experiments have been carried out in low-oxygen (8 ppm by mercury amalgamation technique) sodium and in vacuum at 650°C. After eight days at 650°C there was essentially no difference in response to Strauss reagent of samples exposed in the relatively pure sodium and in vacuum. It is thus tentatively concluded that the effect of sodium on stainless steel is dependent on impurities in the sodium.

4. Fuel Reprocessing

a. Processes for Fast Reactor Fuels. Pyrochemical processing of fast reactor fuels of the ceramic type (e.g., oxide or carbide) clad with stainless steel is being investigated. A conceptual flowsheet currently under investigation has been described previously (see Progress Report for September 1966, ANL-7255, pp. 30-31). The process utilizes liquid metal-molten salt extractions and salt-transport separations for the separation of fissile and fertile constituents of the fuel from the fission products. Recent laboratory investigations of the current flowsheet are discussed below.

The first step after the decladding of mixed oxide ($\text{PuO}_2\text{-UO}_2$) fuels is the reduction of the oxide to metal (plutonium and uranium) by a liquid Cu-33 w/o Mg alloy in contact with a molten salt (see ANL-7255, pp. 30-31). The alkali, alkaline earth, and rare earth fission products and the solid MgO formed as a byproduct of the reduction reaction remain in the waste salt. The plutonium, uranium, and noble and refractory metal fission products are transferred into the Cu-Mg alloy; the plutonium is in solution and the uranium is present as a precipitated metal phase. Laboratory-scale investigations of the variables which influence the reduction of UO_2 in molten salt-liquid metal systems have continued.

Several experiments were performed to study the effect of agitation speed on the reduction rate of UO_2 in a 42.5 m/o NaCl-37.5 m/o CaCl_2 -15 m/o MgCl_2 -5 m/o CaF_2 /Cu-33 w/o Mg system at 800°C. Tests were made at agitation speeds of 600, 800, and 1000 rpm, and oxide loadings of 2 and 5 w/o. The results show that variations in the agitation speed (between 600 and 1000 rpm) have no effect on the reduction rate in this system. The reductions achieved with this salt mixture have been only ~90% complete in 4 hr, which indicates that this mixture is not satisfactory as a reduction salt. However, reductions of >99% are currently being achieved in 2 hr with a salt containing 47.5 m/o MgCl_2 -47.5 m/o CaCl_2 -5 m/o CaF_2 .

An increasing level of iron contamination in EBR-II fuel has been reported (see Progress Report for November 1966, ANL-7279, p. 9). The skull-reclamation process developed for EBR-II, but not installed in the Fuel Cycle Facility, has a demonstrated capability of separating >99% iron from uranium.¹¹ The use of molten cerium as an extractant to remove

¹¹ANL-6569, p. 36; ANL-6413, p. 52.

iron from liquid uranium-fissium fuel has been proposed. An experiment was conducted to determine the distribution coefficient of iron between the relatively immiscible liquid cerium and uranium-fissium phases. The data from the experiment show the distribution coefficient (ratio of w/o Fe in cerium to w/o Fe in uranium-fissium) to be about 0.2 at temperatures between 1000 and 1200°C. This value indicates that iron could be extracted from uranium-fissium, using cerium, with repetitive extractions. However, because uranium is slightly miscible with cerium, some uranium would have to be subsequently recovered from the cerium.

Another experiment was conducted to determine if iron could be separated from uranium by the selective precipitation of uranium as a zinc-uranium intermetallic compound from a liquid Zn-Fe-U alloy. An alloy was made which contained about 2 w/o iron in uranium (on a zinc-free basis). After dissolving the alloy in zinc at 800°C, filtered metal samples were taken during a controlled cooling period. A one-hour mixing period and a 30-min settling period preceded sampling at each temperature. The results expressed in terms of the Doerner-Hoskins coprecipitation coefficient λ^{12} were ~0.03 from 800 to 580°C and ~0.43 from 580 to 425°C. The differences in the coprecipitation coefficients over the temperature range indicate a difference in the form of the material being precipitated. These results show that iron could be separated from uranium by a selective precipitation process, but implementation of this method of iron removal from EBR-II fuel in the Fuel Cycle Facility would require a new furnace and some modification of existing equipment, as well as new retorting facilities for recovery of the uranium product.

Engineering studies for the plutonium salt transport process have shown that reductions of 98 to 100% are obtained when 47.5 m/o MgCl_2 -47.5 m/o CaCl_2 -5 m/o CaF_2 is used as the salt phase with a Cu-33 w/o Mg alloy. However, in the subsequent extractions steps it is desirable to use a salt of lower MgCl_2 content in order to minimize plutonium losses. An experiment has been completed in which the MgCl_2 content of the salt was decreased by adding calcium and lithium metal to the salt-metal system at about 700°C. Magnesium oxide was added to the salt to simulate the reduction product, and cerium was added to determine its distribution coefficient in the resultant salt-metal system. Complete analytical results are not available, but preliminary analyses show that the chemistry of the process proceeded as expected with a resulting salt composition of about 56 m/o CaCl_2 -15 m/o MgCl_2 -24.6 m/o LiCl -4.4 m/o CaF_2 . This composition should be suitable for the extraction steps.

Engineering studies to test various equipment components in a flowing molten chloride salt stream have been completed. The system,

¹²The Doerner-Hoskins coefficient λ is defined by

$$\log \left(\frac{\text{tracer in solution}}{\text{total tracer}} \right) = \lambda \log \left(\frac{\text{carrier in solution}}{\text{total carrier}} \right).$$

which was described previously (see Progress Report for December 1966, ANL-7286, p. 33), has been operated a total of 1348 hr circulating 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl at an average temperature of 560°C and an average flow rate of 1 gpm. During the last 328 hr of operation the salt also contained UCl_3 and ZnCl_2 .

Metallographic examination of the system revealed no significant change in wall thickness, but intergranular penetration of up to 0.05 mm was observed. In stressed sections the penetration was somewhat greater. A freeze valve in the drain line of the system had been opened and closed approximately 40 times. As a result of these operations the valve was badly distorted and cracked. Although the life of this type of valve is limited, freeze valves are still useful in locations where frequent operation is not required. The centrifugal pump performed well, although an oil seal leaked some oil into the salt. The pump employed a hydrostatic bearing which supported the impeller shaft in the circulating fluid. This feature will be especially useful in pyrochemical-processing equipment because these processes use relatively deep vessels.

A niobium crucible is being tested in a materials evaluation program to find suitable materials for pyrochemical processing equipment. The crucible ($4\frac{3}{4}$ -in. in OD by $6\frac{1}{2}$ -in. high with a 0.060-in. wall) was fabricated by rolling and welding niobium sheet. Four mixing baffles extend $\frac{3}{4}$ in. into the crucible. This crucible has been tested for 528 hr with a Cu-33 w/o Mg-0.1 w/o U/ MgCl_2 -30 m/o NaCl -20 m/o KCl system at 700°C . After 384 hr the crucible was cleaned with nitric acid and inspected. No evidence of corrosion was detected. The crucible does, however, appear to be slowly losing its ductility, probably as a result of temperature cycling and salt embrittlement. After 528 hr the crucible was subjected to severe physical shock to determine if sufficient embrittlement had occurred to cause fracture. A dent was produced, but the crucible did not fracture. This study will be continued using 50-hr exposures until the crucible fails or a total exposure of 1000 hr has been obtained.

Engineering studies are under way to investigate the use of a zinc dissolution process as a simplified head-end step for decladding of oxide fuel assemblies from fast reactors. Remote disassembly and mechanical decladding involves a number of complex cutting and handling operations which require equipment that is both costly and difficult to maintain. The process being investigated involves the dissolution of stainless steel cladding and fuel-element structure in molten zinc at 800°C to form a 5 to 7 w/o solution of stainless steel in zinc. The bulk of the solution would then be transferred away, leaving the oxide fuel and residual zinc-stainless steel solution in the decladding crucible.

An experiment was performed in a retorting furnace to determine if the solvent zinc could be recovered from the zinc-stainless steel solution

discharged from the decladder. The results of the experiment show that 98% of the zinc can be recovered by retorting at 850°C at a pressure of 10 Torr. The remaining zinc was combined with the stainless steel as a hard, adherent sintered mass. Evaluation of the costs involved in a plant-scale operation will be made to determine whether such zinc recovery represents an economical advantage over discarding the solvent zinc as radioactive waste.

F. Design Concept Analyses and Advanced Systems Evaluation

1. 1000-MWe Study

a. General. Procedures for project control and reporting 1000-MWe LMFBR follow-on studies have been written; they are being reviewed by AEC.

b. Combustion Engineering, Inc. The contract with Combustion Engineering, Inc. was executed; it provides that the work be done on a cost-plus-fixed-fee basis between February 6, 1967 and August 6, 1968.

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

Program reporting and planning will be by means of the Program Evaluation and Review Technique (PERT) system.

c. Babcock & Wilcox. B&W has essentially completed the four conceptual designs in the Task I effort (see Progress Report for March 1967, ANL-7317, pp. 62-63). From an evaluation of the best features of each concept, the company is selecting the major features of a reference design. In-depth engineering of the reference design in Task II will begin as a parallel effort with trade-off studies so as to provide information necessary to choose the final reference design.

Babcock & Wilcox submitted its first formal progress report for the period October 15, 1966 to March 1, 1967.

G. General Research and Development

1. Fast Reactor Core-parameter Study

Sodium-void coefficients have been calculated using the perturbation technique and the DEL subroutine of the MACH-I code. The power density

studied was 400 kW/liter. Results were obtained running both radial and axial problems with the code. A feature of the DEL subroutine is the possibility of varying the limits of integration in the transverse direction, that is, perturbing the sodium content only in that portion of the central region corresponding to the physical core. This was the method adopted. The results show that the perturbation method is in at least approximate agreement with the Meneghetti method reported previously (see Progress Report for February 1967, ANL-7308, p. 41). For example, at a core height-to-diameter (H/D) ratio of 0.6, for which the Meneghetti method gave a sodium-void coefficient of 1.00851, the radial perturbation gives 1.00767 and the axial perturbation gives 1.00781. For a low value of H/D, such as 0.2, the axial perturbation gives results closer to the Meneghetti technique than the radial perturbation. This might be expected, because for a pancake core the axial (slab) representation is more reasonable than the radial (cylinder) representation.

Preliminary results have been obtained for a two-dimensional problem that has been set up and run on the CANDID-2D code. The reactor studied has critical compositions derived using one-dimensional self-consistent diffusion theory on the MACH-I code. It has $H/D = 0.6$ and core power density = 400 kW/liter. The two-dimensional representation consisted of core, axial blanket, radial blanket, and reflector. A space mesh of 12×12 was used first, with convergence criteria of 1.0×10^{-3} . Under these conditions the value of k converged to 1.0027. The mesh then was doubled to 24×24 and the convergence criterion tightened to 1.0×10^{-4} . Under these conditions the value of k converged to 1.0009. An effort to further double the mesh to 48×48 has caused machine trouble, apparently due to inadequate capability to handle such a large problem.

III. GENERAL REACTOR TECHNOLOGY

A. Applied and Reactor Physics Development

1. An Improved Diagnostic Technique for Input-Output Equipment

There are certain difficulties involved in the diagnosis of malfunctions in electromechanical input-output equipment for computers. For one thing, it is not always easy to determine whether the trouble is in the device itself or in the interface. Furthermore, the mechanical parts in these devices move so rapidly that direct visual observation of their action is often impossible. Also, it is often difficult to display the electrical signals involved in a meaningful way.

In the process of maintaining a DDP-24 computer system, an improved diagnostic technique has been developed which is intended to assist in eliminating these difficulties and in yielding more accurate and enlightened diagnoses. This computer, like others in its class, has a number of output control pulse (OCP) lines. A pulse may be made to appear on any of these lines by executing an OCP instruction with the appropriate address. The technique, then, involves the use of specially written diagnostic programs that are arranged to execute an OCP instruction addressed to an external line before each operation of the device under test. The OCP line so addressed is connected to the delaying sweep external-trigger input of an oscilloscope. The waveforms of various signals in the interface may be thus displayed in synchronism with the operation of the device. In this way, one may check the operation of the interface and examine the signals generated by the device. Such tests can very rapidly confirm or rule out the possibility of trouble in the interface.

In addition, the delayed trigger from the oscilloscope can be made to trigger a stroboscope illuminating the moving mechanical parts of the device. The resulting flash from the stroboscope will reveal the status of the moving part at that point in the operating cycle.

Fig. 16. Block Diagram for Diagnostic Equipment Arrangement

A block diagram of the layout is shown in Fig. 16.

The OCP line is connected to the external trigger input of time base B on a Tektronix 545A oscilloscope. With the oscilloscope probes connected to the circuits under test, the waveforms at those points will be displayed on that time base in synchronism with the operation of the device.

Time base A may be used in the normal delayed-sweep mode to display small portions of the cycle. For example, it is convenient for observing the behavior of contacts at the time of opening or closing.

The stroboscopic function is performed by a General Radio Strobotac, type 631-BL. As shown in Fig. 16, a 2D21 thyatron was connected across the CONTACTOR terminals of the Strobotac, which is operated in the CONTACTOR LOW mode. This thyatron is fired by the initiation of the +GATE A signal from the oscilloscope. With the oscilloscope operated in the B INTENSIFIED BY A mode, the left-hand edge of the intensified region indicates the instant of Strobotac firing. Thus the mechanical and electrical indications may be correlated.

A number of special diagnostic codes have been written for use with this technique. They are used for diagnosing a Selectric typewriter, a paper-tape punch, and a line printer.

The technique has also been applied to the diagnosis of central-processor malfunctions. Sometimes a machine instruction will malfunction, often only with a particular data configuration in the machine registers. A diagnostic program is then written to establish the configuration in question, pulse an OCP line, execute the instruction, and repeat. The various circuits involved can then be readily checked. Since there are so many possible situations, these programs are written as needed.

If an on-line system has its OCP lines permanently connected to various external devices, it would be convenient to reserve one line for maintenance. Various diagnostic programs could then use this line for synchronizing with different functions.

2. The ARC System

A significant improvement has been made in the MC^2 code in that the total inelastic and $n,2n$ cross sections are now evaluated at the ultrafine-group level. In the previous version of the code, these cross sections were evaluated at the fine-group level.

The inelastic and $n,2n$ transfer matrices are still obtained at the fine-group level, but these matrices are now normalized to the ultrafine-group total inelastic and $n,2n$ absorptions for the corresponding fine group.

For example, let the ultrafine group (ufg) j be contained in the fine group (fg) J . Then in the calculation of the fundamental-mode spectrum, the source into ultrafine group j due to inelastic scattering, $S_{inel,j}^{ufg}$, is now evaluated as

$$S_{inel,j}^{ufg} = \sum_{K \neq J} \sum_{k \text{ in } K} \Sigma_{inel,k}^{ufg} \phi_k^{ufg} \frac{\Sigma_{inel,K \rightarrow J}^{fg}}{\Sigma_{inel,K}^{fg}} \frac{\Delta e_j}{\Delta e_J} \quad (1)$$

In Eq. (1), $\Sigma_{inel,k}^{ufg}$ is the total macroscopic inelastic cross section for ufg k, ϕ_k^{ufg} is the flux for ufg k, $\Sigma_{inel,K \rightarrow J}^{fg}$ is the macroscopic inelastic scattering cross section from fg K to fg J, and $\Sigma_{inel,K}^{fg}$ is the total macroscopic inelastic cross section for fg K. The Δe_j and Δe_J are, respectively, the energy widths of the ufg j and the fg J.

The corresponding loss term in the denominator of the fundamental-mode flux equation is therefore given by

$$\Sigma_{inel,j}^{ufg} \left[1 - \frac{\Sigma_{inel,J \rightarrow J}^{fg}}{\Sigma_{inel,J}^{fg}} \right], \quad (2)$$

where the term in the bracket accounts for the fact that self-scatter is not included in the equation.

To illustrate the improvement in the results using the new ultrafine-group technique, MC² problems have been run for a composition typical of a large oxide-fueled fast reactor. Three problems were run with fg lethargy widths ΔU of 0.5, 0.25, and 0.166666 for both the original fg treatment and the improved ufg treatment. Improved spectra resulted.

Improvement obtained by use of the code is shown in Table XXI, where the broad-group elastic removal cross sections for Fe, Na, and O are compared for the fg and ufg inelastic treatment.

TABLE XXI. Elastic Removal Cross Sections for the Light Elements

Fine-group Treatment of Inelastic Scattering										
Broad Group	Lower Energy (MeV)	Fine Group: $\Delta U = 0.1666667, k_{\infty} = 1.026$			Fine Group: $\Delta U = 0.25, k_{\infty} = 1.029$			Fine Group: $\Delta U = 0.5, k_{\infty} = 1.043$		
		Fe	Na	O	Fe	Na	O	Fe	Na	O
1	3.68	0.0512	0.128	0.483	0.0468	0.118	0.456	0.0322	0.0818	0.322
2	2.23	0.0963	0.319	0.252	0.0914	0.304	0.249	0.0738	0.250	0.210
3	1.35	0.0988	0.314	0.622	0.0954	0.304	0.613	0.0810	0.260	0.534
4	0.82	0.200	0.604	1.17	0.195	0.592	1.17	0.178	0.544	1.09
5	0.50	0.224	0.678	0.849	0.223	0.674	0.846	0.215	0.655	0.821
6	0.30	0.156	0.780	1.43	0.157	0.786	1.44	0.159	0.793	1.45

Ultrafine-group Treatment of Inelastic Scattering										
Broad Group	Lower Energy (MeV)	Fine Group: $\Delta U = 0.1666667, k_{\infty} = 1.021$			Fine Group: $\Delta U = 0.25, k_{\infty} = 1.023$			Fine Group: $\Delta U = 0.5, k_{\infty} = 1.035$		
		Fe	Na	O	Fe	Na	O	Fe	Na	O
1	3.68	0.0582	0.138	0.508	0.0581	0.138	0.507	0.0574	0.136	0.501
2	2.23	0.100	0.327	0.256	0.0987	0.321	0.254	0.0939	0.306	0.243
3	1.35	0.101	0.318	0.628	0.0991	0.313	0.623	0.0921	0.292	0.584
4	0.82	0.202	0.608	1.17	0.199	0.601	1.17	0.189	0.572	1.12
5	0.50	0.226	0.680	0.852	0.225	0.677	0.850	0.219	0.663	0.832
6	0.30	0.156	0.781	1.43	0.158	0.786	1.44	0.159	0.792	1.45

The use of the fine-group treatment of inelastic scattering is too crude for making accurate reactivity calculations. From Table XXI, one can see that this can lead to a criticality error of about 2% for this particular composition.

3. Cross-section Evaluations

The ETØE code, prepared by the Atomic Power Development Associates, was adapted by them for use on the CDC-3600 computer. This code reads data from an ENDF/B library tape and produces a library tape in the format required by the MC² code. A test MC² library tape containing data for Fe, Na, U²³⁸, Pu²³⁹ and Pu²⁴⁰ materials was made using data from the first ENDF/B distribution. This test tape and the current version of the MC² library tape were used to solve a reactor problem involving the above-mentioned materials. The critical bucklings found using these two tapes differed by an amount corresponding to about 1½% in reactivity, with cross sections in the current version of the MC² library tape being more reactive than those based on ENDF/B library tape.

B. Reactor Fuels and Materials Development

1. Chemistry and Fuels Materials

a. Behavior of Uranium Monophosphide. Tests have shown that uranium monophosphide (UP) and tungsten interact negligibly in times up to 5 hr and at temperatures up to $2593 \pm 10^\circ\text{C}$ if the UP is maintained stoichiometric. If preferential vapor loss of phosphorus from the UP occurs at high temperatures, a limited amount of a ternary U-P-W liquid forms at the interface of the UP and tungsten grains. No solid solubility of UP in tungsten or of tungsten in UP was detected metallographically.

Thermal analysis of a UP-W charge in a tungsten crucible was followed with a Leeds and Northrup recording optical pyrometer. A thermal arrest was observed at $2630 \pm 20^\circ\text{C}$ on cooling. Metallographic examination of the products of this test, of the (partial) reaction of UP in contact with tungsten at $2647 \pm 10^\circ\text{C}$, and of the compatibility test at 2593°C suggest that (1) UP and tungsten form a simple eutectic system, (2) the observed thermal arrest temperature is the eutectic temperature, and (3) the composition of the eutectic is ~20-30 w/o tungsten. The previously reported melting point ($2610 \pm 20^\circ\text{C}$) of UP, observed from its slumping in a tungsten crucible, is probably the eutectic temperature; the true melting point may be significantly higher.

b. Preparation of Fuel Specimens in Helium. Procedures for the preparation of ceramic fuels for metallographic examination are being tested in a glovebox containing a helium atmosphere. The purpose of the test is to develop fuel-handling techniques which can subsequently be applied in a helium atmosphere enclosure of a shielded fuel evaluation facility which is now under construction in the Chemistry Division cave complex.

In the tests, the helium used in the glovebox was continuously purified (by a cryogenic method) to minimize the reaction of impurities with simulated ceramic fuels. Typically, the combined moisture and oxygen impurities were <10 ppm, with the oxygen fraction seldom exceeding 2 ppm.

Materials selected for initial testing were pressed-and-sintered UC and arc-melted UC_x. Sections of the fuel selected for testing were mounted in cylindrical plastic molds with epoxy resin. Grinding of the specimens was carried out on wheels surfaced with SiC papers with 80, 120, 180, 320, 400, and 600 grit sizes, and polishing was carried out on Buehler "Texmet" cloths impregnated with 6- and 1-micron diamond paste. A Buehler "Whirlimet" apparatus was employed in the grinding and polishing steps, and a mixture of hydrogenated terphenyls (a Monsanto product, HB-40) was used as lubricant for the specimens. The specimens were cleaned ultrasonically with "Freon" TF solvent (a du Pont product, CCl₂F-CClF₂) and/or reagent-grade benzene. The benzene-cleaned specimens were vacuum dried. Optical inspection of the prepared specimens was accomplished by remote viewing through a metallurgical microscope which was installed in the glovebox.

Cleaning of the surfaces of pressed-and-sintered UC specimens with benzene produced discolorations of various shades of brown; however, these discolorations did not reveal the structure of the material. On the other hand, benzene cleaning of the arc-melted specimens revealed the presence of large irregular grains approximately 1 mm in cross section. The grains were sharply defined by color variations, predominantly white, green, blue, red, and yellow. Examination of an arc-melted specimen with an electron-probe microanalyzer indicated that the various colored grains were of similar composition. Apparently, impurities in the benzene (major impurity, 550 ppm H₂O) had reacted with the surface of arc-melted carbide to form an interference film which produced colors that varied with orientation of the grains. Accary¹³ had previously noted similar grain-coloring effects when carbide specimens (arc-melted or sintered) were etched in acid solutions.

Simulated uranium carbide fuel specimens prepared by the above procedures were not stained when cleaning after the final diamond polish was accomplished with the use of "Freon" TF (CCl₂F-CClF₂). Some microstructure was revealed in arc-melted UC_x but was not apparent in pressed-and-sintered UC. The polished surfaces showed no apparent change after storage under helium for three weeks.

To determine if air etching would reveal the microstructure of the UC specimens, they were exposed to room atmosphere. Fracturing of the polished surfaces by reaction with air, which occurred within 16 hr, did little to assist in revealing fuel structure.

¹³Accary, A., J. Nucl. Mater. 8(3), 281 (1963).

Cathodic etching of ceramic fuel specimens will be tested as a possible method of defining microstructure. An advantage of this etching technique is that the fuel surfaces will not be contaminated with conventional acid etchants which may interfere with subsequent microprobe or mass analysis.

c. Sampling of Irradiated UO_2 by Microdrilling. Data on the fission product and heavy element isotopic composition changes within irradiated fuel can readily be obtained from fuel samples obtained by drilling. A commercial microdrill (Dumore Co., Racine, Wis.) has been adapted for remote use, and preliminary tests of the unit for sampling irradiated fuel have been completed. The fuel used in the sampling tests was a UO_2 fuel pellet, 3.8% enriched in U^{235} , irradiated to about 1.2 a/o burnup in the General Electric Test Reactor, and cooled for more than one year prior to sampling. To prepare the fuel for sampling, a radial section of the pellet was ground and polished. A macrophoto of the polished specimen showed radial cracking of the UO_2 but no evidence of fuel melting.

Twelve samples, 0.020 in. in diameter and 0.015 in. in depth, were drilled across the 0.494-in. diameter of the fuel so that the centers of the sampling areas were 0.040 in. apart. Analytical samples were taken during drilling by vacuuming the powder through a hypodermic needle (ID of 0.032 in.) and collecting it on filter paper with an average pore size of $0.8\ \mu$. The solid fuel samples were first analyzed gamma spectrometrically, and then dissolved for analysis of uranium and plutonium.

Appreciable density change across the fuel diameter was indicated by the variation in the amount of uranium in the samples. The analyzed weight of uranium in the 12 drillings of equal depth and diameter varied by a factor of four (from 105 to 400 μg). Therefore, in determining the distribution of fission elements in the fuel it was assumed that the amount of fuel in each sample was proportional to the amount of uranium present. Fission element activity per milligram of uranium in the individual samples was normalized to that of the sample containing the lowest level of activity of the element per milligram of uranium.

Gamma photopeaks that could be resolved in each drilled sample were those of $\text{Zr}^{95}\text{-Nb}$, Cs^{137} , $\text{Ce}^{144}\text{-Pr}$, and $\text{Ru}^{106}\text{-Rh}$. The redistribution of $\text{Zr}^{95}\text{-Nb}$, Cs^{137} , and $\text{Ce}^{144}\text{-Pr}$ was relatively small; their changes in concentration across the diameter of the fuel were less than a factor of 2.5. However, $\text{Ru}^{106}\text{-Rh}$ was concentrated in the peripheral area of the fuel. Under the conditions of fuel irradiation, the ruthenium apparently reacted with oxygen to form a highly volatile species which migrated to the cooler outer zone of the fuel.

The redistribution of bred plutonium in the fuel was also small. Ten of the 12 areas sampled were of equal plutonium concentration, within

the limits of analytical accuracy. An apparent 30% decrease in plutonium in two areas sampled at the outer edge of the fuel is attributed to the effects of flux depression in this zone.

d. Laser Sampling Device. A small laser attached to a microscope has been developed for use as a microsampling device. Depending on the selection of optical components and the input power to the laser, craters from 5 to 100 μ in diameter can be vaporized in uranium dioxide. If a microscope cover glass is mounted 0.2 to 0.4 mm above the surface of the specimen and the laser is fired through it, much of the vaporized UO_2 is condensed on the glass. Samples from about three nanograms to one or two micrograms can be obtained in this manner.

Vaporizations have been carried out in both vacuum and air. There seems to be little difference in the appearance of the deposit when the craters are large, i.e., greater than 20 μ . In each case a small mirror-like spot can be seen on the cover glass without the aid of magnification. Most of the material is condensed as a thin film covering an area with a diameter at least three times the diameter of the crater. In addition, some of the material is caught in the form of fine droplets. These are rarely larger than a micron in diameter.

Such samples should be useful for the examination of the composition of very small areas of metallurgical specimens. Three methods are being considered: alpha pulse height analysis, gamma spectroscopy, and neutron activation.

As a test of this technique, samples were taken from a small segment of an irradiated fuel rod. The rod was 3.8% enriched uranium dioxide, irradiated to approximately 1% burnup. A crater 50 μ in diameter was made near the edge of the specimen. The weight was estimated to be about 0.5 μg . One drop of 0.05% collodion solution was dried on the vaporized sample on the cover glass to help hold the sample. The cover glass was then placed directly into the counting equipment. The sample was counted for 16 hr using a silicon-surface-barrier solid-state detector. The maximum resolution obtained with this detector was 25 keV (FWHM) on a monoenergetic alpha source.

Specimens obtained by this technique should also be adequate for gamma spectroscopy and for neutron activation. Further work is planned on all three types of analyses.

2. Fabrication and Evaluation

a. U-S-O System. The extent and nature of the UOS solid-solution area (see Progress Report for February 1967, ANL-7308, pp. 56-57) was investigated in more detail. Uranium disulfide ($\text{US}_{1.97}$) and uranium

dioxide ($\text{UO}_{2.15}$) powders were blended to give nine compositions ranging from 12 m/o US_2 -88 m/o UOS through UOS to UO_2 , pelletized, and fired at 1450, 1550, and 1650°C for 3 hr in vacuum.

At 1650°C, X-ray and metallographic data indicated that almost 70 m/o UO_2 is soluble in UOS, meaning that up to 70% of the sulfur atoms in the UOS structure can be replaced by oxygen atoms. An estimated 1 to 3 v/o of small US particles were dispersed in equilibrium with the UOS-UO_2 solid solution. With addition of more than 70 m/o UO_2 to the UOS, a cubic phase having almost the same lattice parameter as pure UO_2 occurs in equilibrium with the solid solution, suggesting only slight solubility of sulfur in UO_2 . In the UOS-US_2 direction, other data indicate that no more than a few percent of US_2 is soluble in UOS.

The extensive substitution of oxygen for sulfur atoms was unexpected since O^{2-} ions ($r = 1.32 \text{ \AA}$) are 28.2% smaller than the solvent S^{2-} ions ($r = 1.84 \text{ \AA}$), and familiar substitution rules indicate that solubility should become restricted when the difference reaches 15%. In this case, however, the tetragonal PbFCl structure of UOS ($a_0 = 3.844 \text{ \AA}$; $c_0 = 6.694 \text{ \AA}$) is completely layered, having like atoms arranged in planes perpendicular to the c -axis in a $\text{O-U-S-S-O-U-S-S} \dots$ sequence. The smaller oxygen ions can thus be accommodated by a contraction in the c -direction.

An analysis of the X-ray powder data for the solid solutions reveals a definite relationship between the UOS and UO_2 structures. This relationship involves the effect of increasing oxygen substitution causing the d -spacings of the $\{110\}$ and $\{101\}$ planes of the UOS phase to approach systematically those of the $\{200\}$ and $\{111\}$ planes, respectively, of UO_2 (fluorite structure, $a_0 = 5.470 \text{ \AA}$), i.e., the cation arrangement in the UOS phase approaches identity with the cation lattice of UO_2 . In the case of $d_{110} \rightarrow d_{200}$, only a slight expansion of a_0 ($3.844 \rightarrow 3.868 \text{ \AA}$) is involved, whereas for $d_{101} \rightarrow d_{111}$ a large contraction of c_0 ($6.694 \rightarrow 5.470 \text{ \AA}$) is additionally required. A continuous transition between the two structures is not possible since, even if a single plane of oxygen atoms was formed by complete substitution in the sulfur double layer, it would have to be translated through $a_0/2$ to obtain the fluorite structure.

Room-temperature measurements showed the resistivity of the UOS solid-solution phase to have changed from that of a semiconductor to that of an insulator somewhere between the compositions having 50 and 70 m/o of the solute UO_2 . From UOS through the 50 m/o UO_2 solution, the resistivity varied (although not systematically) from 1.2 to $2.8 \times 10^{-2} \Omega\text{-cm}$, and then jumped to $4.2 \times 10^2 \Omega\text{-cm}$ for the 70 m/o UO_2 solution (as compared to $5.2 \times 10^2 \Omega\text{-cm}$ for the pure UO_2 end member). Intermediate compositions are being prepared to determine whether this is a gradual or an abrupt transition.

Resistivity measurements by the conventional four-point probe method required silvering the ends of the pellets for adequate contact with the current-lead blocks. When a thin coating of silver was baked-on for 10 min at 610°C in vacuum, the block-to-sample interfacial resistance dropped by a factor of about 30, apparently because of the formation of an intermediate Ag_2S phase. This phase was not formed at 450°C. Use of the improved contact increased the reproducibility of independent measurements by reducing the variance from $\pm 20\%$ to $\pm 1\%$.

b. Dissolution Kinetics in Liquid-metal Systems. The validity of applying the internal-friction method of analysis for oxygen of tantalum wire equilibrated with sodium to the determination of oxygen in sodium has been studied (see Progress Report for February 1967, ANL-7308, pp. 60-61). This work has been mainly concerned with accurate determination of the distribution coefficient for oxygen between tantalum and sodium. The specificity of the method (a characteristic as important as the sensitivity) has led to initiation of a study of the behavior of nitrogen in sodium systems.

Development of the proposed method for analysis of low oxygen concentrations in sodium continues to involve the equilibration, with respect to oxygen activity, of tantalum wires in liquid sodium and the subsequent measurement of the oxygen concentration in the tantalum by internal-friction techniques. The oxygen concentration in the sodium is then related to the oxygen in tantalum by the distribution coefficient ($\text{ppm O in Ta/ppm O in Na}$), which has been calculated from the best available free energy and solubility data.

The detection of significant nitrogen pickup by the tantalum wires during exposure to sodium (see ANL-7308, pp. 60-61) has led to an attempt to measure the nitrogen and carbon concentration in sodium by the same method. Since each interstitial element has a characteristic internal-friction peak in a body-centered-cubic metal, it is possible to distinguish the impurity elements and measure them simultaneously with the same wire. Because of a lack of free energy and solubility data, the distribution coefficients for nitrogen and carbon between tantalum and sodium cannot be determined. Therefore, the friction peak heights have not been quantitatively related to the concentrations of these elements in sodium.

Since the diffusivities of nitrogen and carbon in tantalum are relatively low, niobium and vanadium wires are being tested in an attempt to reduce the time required for equilibration of the impurity elements. The diffusivities of nitrogen and carbon are considerably higher in these metals than in tantalum. Based on solid-state diffusion control, the times required for 99% equilibration of the 0.25-mm-dia wires are listed in Table XXII.

TABLE XXII. Equilibration Times

Wire (0.25-mm dia)	Equilibration Time in 700°C Sodium (99%) (hr)		
	Oxygen	Nitrogen	Carbon
Tantalum	3	2000	2500
Niobium	2	280	220
Vanadium	9	170	11

Listed in Table XXIII are the temperatures at which the internal-friction peaks have been identified for 0.25-mm-dia by 5.0-cm-gauge length wires of the three refractory metals.

TABLE XXIII. Internal-friction Peak Temperatures

Refractory Metal	Frequency (cps)	Temperature of Internal- friction Peak (°C)		
		Oxygen	Nitrogen	Carbon
Tantalum	0.45	132	323	332 ^a
Niobium	0.30	131	268	255
Vanadium	0.40	171	252	159 ^a

^aEstimated.

c. Irradiation of Th-U-Pu Alloys. Six Th-U and Th-U-Pu fuel alloys jacketed in V-20 w/o Ti tubing continued under irradiation in the CP-5 reactor. The objectives of the experiment are to determine the relative swelling behavior of the fuels, the restraint characteristics of the jacket, and the maximum attainable burnup before jacket failure. Table XXIV summarizes irradiation variables for these metal fuel experiments.

TABLE XXIV. Status of Metal Fuel Irradiations in CP-5

Specimen Number	Design Parameters ^a		Max (kW/ft)	Max Cladding Temp (°C)	Burnup to Date	
	Fuel Composition (w/o)	Effective Density (%)			a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ ^b
1N16	Th-20 U	74.7	8.7	590	23.1	11.1
4N19	Th-20 U	74.0	8.7	560	23.1	11.1
2N17	Th-10 Pu-10 U	74.1	9.2	590	26.2	12.3
5N20	Th-10 Pu-10 U	73.0	9.2	590	26.2	12.3
3N18	Th-10 Pu-20 U	73.5	8.7	560	15.6	11.4
6N21	Th-10 Pu-20 U	73.5	8.7	560	15.6	11.4

^aAll specimens are sodium-bonded to V-20 w/o Ti jackets, of 0.198-in. OD with a wall thickness of 0.015 in.

^bBased on effective density.

d. Thermal Stability of Plutonium Ceramics

(i) PuO₂ Evaporation. The results of experiments on the determination of oxygen-to-metal (O/M) atomic ratios in hypostoichiometric (U,Pu)O_{2-x} compositions, have been reported (see Progress Report for February 1967, ANL-7308, pp. 55-56). Heat treatment at 1100°C in helium-1 v/o O₂ followed by equilibration at 850°C in a 10 to 1 CO/CO₂ volume mixture apparently produced a final O/M ratio of 2.00. Measurements of the vapor pressure of hypostoichiometric PuO_{2-x} have now been completed. Although it has been indicated (see Progress Report for January 1967, ANL-7302, p. 65) that rhenium Knudsen cells might be used for subsequent measurements of vapor pressure, all measurements have so far been made with a tungsten cell; iridium, although previously demonstrated to be compatible with PuO₂, has such a high vapor pressure above 2100°K that the use of iridium cells is impracticable at the temperatures required for the PuO₂ work.

Fig. 17

Weight Losses per Second of PuO_{1.825} and PuO_{1.875} in Knudsen-cell Runs. (The respective final O/M ratios are 1.82 and 1.79.)

Figure 17 shows the results of two series of Knudsen-cell runs on specimens with initial oxygen-to-metal (O/M) ratios of 1.825 (Series 1) and 1.875 (Series 2), respectively. The O/M ratio after evaporation of the former was 1.820 and that of the latter was 1.794. Linear evaporation rates obtained for all of the runs in Series 1. In the first few high-temperature runs in Series 2, however, nonlinearity of the evaporation rates indicated that the composition of this specimen was changing. The data given for Series 2 represent runs made after the rates became linear, and presumably represent evaporation of a composition corresponding to the final O/M ratio of 1.794.

The small composition difference between the two specimens does not cause a significant difference in the rates of evaporation of each. Furthermore, the measured weight-loss rates were linear and the final compositions almost the same. The evidence thus indicates that the congruently evaporating composition has an O/M ratio close to 1.80 in the temperature range from 2070 to 2360°K.

3. Techniques of Fabrication and Testing

a. Ultrasonic Instrument and Transducer Development. Fabrication of two transducer probes with a PZT-4 piezoelectric element was completed. The two elements are identical except that the tungsten backing of one is 62% dense and of the second only 52% dense. Evaluation of these probes has started.

b. Development of a Neutron Image Intensification System. The neutron television system incorporating a neutron-image intensifier tube (see Progress Report for April 1966, ANL-7204, p. 52) is now being evaluated for use in studying the process of casting heavy metals. X-ray motion-study techniques can be used¹⁴ for casting studies, but X-radiation transmission for reasonable thicknesses of reactor fuels of interest, such as uranium or plutonium, would be relatively low. The neutron motion study of castings essentially complements the X-ray work by extending the range of metals for which this technique may be useful.

Prototype casting studies were made at Juggernaut reactor with the neutron television system. A low-melting point alloy, Wood's metal (50% Bi, 25% Pb, 12.5% Sn, 12.5% Cd), was chosen for the initial tests because the thermal-neutron transmission through that material approximates that through U^{235} .

Figure 18 shows a result of one of the casting study trials. Wood's metal was poured into an aluminum block in which a 0.63-cm-dia mold pattern was formed. Every other frame from a 16-mm motion-picture recording of the neutron television monitor is shown. The metal begins to enter the mold from the top in frame 1, runs horizontally in frames 2 and 3, begins to drop again into the vertical member of the mold in frame 3, and

Fig. 18. Individual Frames of a Motion-picture Recording Showing a Televised Neutron Transmission Image of the Casting of Wood's Metal into an Aluminum Mold. The sequence begins in frame 1, with the molten metal entering from the top. The total time represented by the ten frames shown is less than one second.

¹⁴Rose, H., Dyke, W. P., Grundhauser, F. J., and Barbour, J. P., X-ray Movies of Metal Casting, presented at the 1966 Spring Convention of the Society for Nondestructive Testing, Los Angeles, California, March 1966; see Materials Evaluation 24, 92 (February 1966).

then extends horizontally again beginning in frame 4. Note how the vertical section fills initially just on one side (frames 4-6) and then begins to fill from the bottom in frames 7-10. Further analysis of these casting pictures is now in progress.

c. Determination of Elastic Moduli of High-temperature Materials by Ultrasonics. Ultrasonic measurements were made with a specially machined bar of Type 304 stainless steel at temperatures up to 1000°C in order to determine its elastic properties as a function of temperature (see Progress Report for November 1966, ANL-7279, p. 58). The difference in test temperature between the ends of the index region was kept less than 10°C. Since then some preliminary work was undertaken to develop a means of using shorter measurement samples than had been initially required. A buffer-rod technique is now being used.

Because common ultrasonic couplants will not withstand the high temperatures, they cannot be used. A forced mechanical bond approach was selected. The bonding surfaces have to be flat and polished to a 3 μ finish. Threads placed on the buffer and sample, to clean up the reflections, were used to force the two surfaces together. A coupling was screwed on the buffer and the sample screwed into it. A good mechanical bond was obtained.

Initial tests with this technique were made with an ASTM 6061 aluminum buffer and sample. Temperatures close to the melting point (660°C) were obtained from thermocouple readings on the top of the sample. Sample length was only 7.5 cm, instead of about 46 cm as formerly required. The top configuration did not change. Trailing pulse echoes were measurable to temperatures above 600°C. Work on stainless steel is now in progress.

d. Measurement of Thermal Conductivity of Irradiated Fuel as a Function of Burnup and Temperature. Following completion of the calibration of the system for measuring the thermal properties of fuel elements (see Progress Report for March 1967, ANL-7317, p. 28) the thermal properties of unirradiated EBR-II driver fuel are being measured.

C. Engineering Development

1. Development of Master-Slave Manipulator Systems

a. Electric Master-Slave Manipulator, Mark E4A. An effort is being made to improve the radiation resistance of the Mark E4A slave arm. At the present time, the first gear and synchro in the servo drive unit appear to be the major items limiting the resistance. The motors, tachometers, and the lubricants used throughout the slave arm are good to dosages of 5×10^8 R or better. All other components, except the external wiring, are resistant to much higher dosages.

A low-inertia, hardened-steel (R_c 55) first gear has been designed to replace the formica gear now in use. This gear will increase the effective inertia of the manipulator, but only by about 2%. An aluminum gear had been tried previously, but, due to oxide formation and servo-motor jitter, these wore out very quickly. A few of the new gears and hardened pinions are on order. After these are received and installed, a life test will be run to determine their suitability.

A few synchros will be sent out next month for rewiring with Class H insulation. All components of a complete servo drive unit will then be irradiated to determine the maximum dosage they can withstand.

b. Low-inertia Servo Motor for Manipulators. A special, high torque-to-inertia servo motor has been designed, detailed, and parts are on order. This 2-pole, 2-phase motor uses a rotor with an annealed-steel core and a copper sleeve. The motor was designed to have the following characteristics:

Synchronous speed	3600 rpm
Stall torque (max)	9.6 in.-lb
Rotor inertia	0.036 lb-in. ²
Max input power	560 W
Mechanical time constant	3.63 msec
Cooling required (air)	17 cfm
Pressure drop in motor	0.8 psi

With these characteristics, the motor would be suitable for a 100-lb capacity manipulator such as the Mark E4B, with a synchronous speed at the wrist joint of 36 in./sec. The rotor inertia, referred to the manipulator wrist joint, is equivalent to a 4-lb mass at the wrist joint. This is about the same as the inertia of the motors used on the 50-lb capacity Mark-E4A manipulator.

2. Heat Transfer and Fluid Flow

a. Niobium-1% Zirconium Loop. A facility has been built 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 critical-flux occurrence, boiling and adiabatic two-phase pressure losses, vapor volume fraction, boiling stability parameters, and ultimately the transient behavior of some of these same quantities.

Flowmeter calibrations have been completed for sodium temperatures up to 1200°F. Several minor additions to the Nb-1%Zr loop have been made in preparation for runs above 1200°F.

b. Heater Experiments

(i) Electron-bombardment Heater (EBH) Experiment. As mentioned in previous progress reports, the original tests have been terminated following satisfactory operation; further EBH tests will be conducted in the new EBH test facility that is being constructed (see Progress Report for January 1967, ANL-7302, p. 71). This facility will be used to test heaters to their maximum heat-flux capabilities before they are installed in the Nb-1%Zr loop.

c. Heat Transfer in Double-pipe Heat Exchangers

(i) Countercurrent Turbulent Liquid-metal Flow. Experimentation with the second heat-exchanger test section was completed. Plans were completed for experiments with the third test section.

Computations for the mathematically degenerate case of equal-heat-capacity flowrates (see Progress Report for February 1967, ANL-7308, p. 68) have been extended for the purpose of studying wall-temperature distributions along the heat-exchanger length.

(ii) Nonsymmetrical Geometries. Construction of the small heat-transfer loop for the purpose of performing experiments with sodium in model heat exchangers is about 50% complete. Fabrication of the first test section, a double-pipe heat exchanger designed to obtain data relating eccentricity of the annular phase on overall heat-transfer rates, is about 60% complete.

(iii) Liquid-metal-heated Steam Generators. An analysis of a simple model of a liquid-metal-heated steam generator (see Progress Report for January 1967, ANL-7302, p. 72) has been completed. Computations are being made for comparing the predictions of the analysis with those of current engineering-design methods.

3. Mechanics of Materials

a. Core Structural Dynamics. To study experimentally the axial-flow-induced vibration of simulated fast-reactor fuel elements, methods of measuring fuel-rod motion are needed. The initial effort to build a device which measures the motion of a peripheral rod has been successful. The inductive device (sensitive to 1×10^{-4} -in. displacement) discussed previously (see Progress Report for January 1967, ANL-7302, p. 73) was used to determine the response of a single rod. Water flowing axially at 10 ft/sec over a 36-in.-long, 1/2-in.-dia polystyrene rod with pinned-pinned ends produced random peak-to-peak displacements of as much as

45×10^{-3} in. The displacement apparently results from repetitive excitation with varying amplitude and duration. In other words, random transverse-pressure fluctuations caused by turbulent flow could be exciting the rod. Two transducers perpendicular to each other indicate rod motion of the same order of magnitude. Thus it can be deduced that the transverse-pressure fluctuations (although applied at differing times) have rms magnitudes which are independent of radial orientation.

Spectrum analysis of the displacement data indicates three frequency bands of rod motion. The major amount of motion occurs at a frequency which corresponds to the first natural resonant frequency of the rod in water. The other two bands of significant amplitude could correspond to second and third mode. However, the second-mode frequency should be lower than that where the experimental peak occurs and the third-mode frequency should be higher. This deviation may be explained by the effect of the axial component of water flow, by the pump-induced vibration of the present test section, or by nonuniformities of the present specimen and specimen-mounting method.

As well as clarification of the peripheral rod motion, the following subjects are being studied: (a) reduction of the pump-induced vibration of the test section; application of vibration-isolation mountings and flexible piping components should be adequate, (b) methods and instrumentation for measuring and analyzing the random transverse pressure field due to turbulent flow and the loop characteristics; cross- and auto-correlation techniques applied to data procured with miniature-crystal pressure transducers appear promising, and (c) methods of monitoring the motion of a rod interior to a bundle in axial water flow; semiconductor strain gauges and miniature accelerometers are being investigated.

D. Chemistry and Chemical Separations

1. Fluoride Volatility Processes

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

(i) Fluorination of UO_2 - PuO_2 -Fission Product Pellets. Development studies are being performed in a 2-in.-dia fluid-bed reactor to establish optimum conditions for the recovery of uranium and plutonium from UO_2 - PuO_2 pellets containing fission products. In the process currently under study, BrF_5 is used 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-17) was performed to evaluate the use of 50 v/o fluorine in nitrogen in the plutonium recovery step instead of 90 v/o fluorine as used previously (see, for example, Progress Report for February 1967, ANL-7308, pp. 71-73).

The charge to the fluid-bed reactor in run Purse-17 consisted of 650 g of 0.5 w/o $\text{PuO}_2\text{-UO}_2$ -fission product pellets and 1100 g of alumina containing 0.6 g CsF , 0.15 g RbF , and 0.5 g NpO_2 . The processing sequence consisted of the following: oxidation of the pellets for 4 hr at 450°C with 20 v/o oxygen, fluorination of the uranium to UF_6 with 10 v/o BrF_5 at 300°C for 2 hr, and recycle-fluorination of the plutonium to PuF_6 with 50 v/o fluorine for 5 hr at 300°C , 5 hr while the reactor temperatures was increased from 300 to 550°C , and 5 hr at 550°C .

The final alumina bed contained <0.001 w/o uranium, 0.009 w/o plutonium, and 0.003 w/o neptunium. These results are nearly identical to those obtained in previous experiments using 90 v/o fluorine in nitrogen for the plutonium recovery step (see ANL-7308, pp. 71-73). Therefore, it appears that the concentration of fluorine can be varied within the range 50 to 90 v/o in nitrogen without affecting the removal of plutonium from the alumina bed by fluorination.

(ii) Purification of Plutonium Streams. In the flowsheet being studied for the fluoride volatility processing of oxide fuels, separation of plutonium is achieved by fluorination of PuF_4 to PuF_6 with fluorine. This PuF_6 stream contains small quantities of uranium, neptunium, and certain fission products which form volatile fluorides, and therefore the stream must be processed further for the preparation of a purified plutonium product. A laboratory-scale experimental program is under way to explore several schemes for the purification of the PuF_6 process stream. Two purification schemes are being explored. One method involves the selective reduction of PuF_6 to PuF_4 by BrF_3 . The second method (not reported this period) employs separation of PuF_6 from fission product compounds of low volatility by sublimation of PuF_6 .

The results of initial experiments evaluating the use of BrF_3 as a selective reductant for PuF_6 have been reported (see Progress Report for March 1967, ANL-7317, p. 85). These experiments, performed with mixtures of fluorides containing gram quantities of UF_6 , NpF_6 , SbF_5 , NbF_5 , and RuF_5 , indicated that separation of uranium, antimony, and niobium from plutonium was essentially complete, while separation of ruthenium was only achieved in a subsequent step involving fluorination of the solid PuF_4 with BrF_5 at 300°C . Recent analytical data have shown that neptunium tends to accompany the PuF_4 and that no significant separation is achieved in either the reduction or fluorination steps. Additional experiments with tracer quantities of niobium (Nb^{95}) and ruthenium (Ru^{106}) showed very poor separation of these elements from plutonium. On the basis of these results, it appears that this technique is not effective in removing fission product elements from plutonium and no further development work on this method is planned.

In other work, the possibility of removing ruthenium from the PuF_6 process stream by selective sorption of RuF_5 on alkaline earth

fluorides is being investigated. A series of three experiments was performed to determine the effectiveness of BaF_2 , CaF_2 , and MgF_2 as sorbents for RuF_5 . In these experiments a mixture of UF_6 (a stand-in for PuF_6) and RuF_5 (containing Ru^{106} as a tracer) in nitrogen was passed through a trap containing 10 g of the alkaline earth fluoride at 100 to 150°C, then through a NaF trap at 100°C to remove the UF_6 , and finally through a cold trap at -78°C. It was observed in these experiments that the effectiveness of the fluorides for RuF_5 sorption decreases in the order BaF_2 , CaF_2 , MgF_2 . The respective decontamination factors were 374, 55, and 9. The evaluation of plutonium-purification methods is being continued.

(iii) Neptunium Fluoride Chemistry. Laboratory-scale studies are being carried out to determine the behavior of neptunium hexafluoride in chemical reactions of interest to its recovery. Current studies concern an investigation of the reaction between NaF and NpF_6 with particular emphasis on identifying the nature and properties of the reaction product. An experiment was performed in which NaF was contacted with NpF_6 (250-300 mm Hg) at 150°C. The product appeared as loose, fluffy particles, pale-violet in color. The weight increase of the solid phase in the reaction of NpF_6 with NaF indicated that three moles of NaF are associated with one mole of neptunium in the complex. It was not possible to establish the valence state of neptunium in the product. Further experiments to obtain this information through absorption spectra of the product are planned.

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

(i) Process Development Studies for Uranium Dioxide Fuels. Engineering-scale studies are being performed in a 3-in.-dia fluid-bed reactor facility to determine the effects of important process variables on the fluorination of UO_2 fuels with BrF_5 . The current program involves a series of eight statistically designed experiments to measure the effects of six independent variables on UF_6 production rate, BrF_5 utilization, off-gas composition, and uranium removal from the fluid bed. The fourth experiment (run BRF5-6) in this series has been completed. In this experiment 2.2 kg of UO_2 in an alumina fluid bed was oxidized to U_3O_8 fines by reaction with 19 v/o oxygen in nitrogen for 6 hr at 450°C. Fluorination of the U_3O_8 fines was carried out using 21 v/o BrF_5 in nitrogen for 2 hr at 300°C.

The course of the fluorination reaction was followed by continuous analysis of the oxygen content of the off-gas stream. On the basis of the oxygen analysis it was observed that the UF_6 production rate remained constant at about 130 lb UF_6 /(hr)(sq ft) until about 80% of the oxide was fluorinated. A BrF_5 utilization efficiency of 40% was observed during the constant rate period. The final alumina bed at the end of the fluorination step contained no agglomerates or unreacted oxide, and was a free-flowing material.

(ii) Fluid-bed Studies with Irradiated Fuels. During the past twelve months a series of experiments were performed in a $1\frac{1}{2}$ -in.-dia fluid-bed reactor to determine the distribution of fission products during the processing of highly irradiated UO_2 fuel by fluoride volatility techniques. Two processing schemes were used: (1) two-zone oxidation-fluorination at 460°C , followed by direct fluorination with 90 v/o fluorine at 450 to 550°C (see Progress Report for May 1966, ANL-7219, p. 52); (2) oxidation of the fuel with 20 v/o oxygen at 400°C , fluorination of the oxidized fuel with 10 v/o BrF_5 at 300°C , and fluorination with 90 v/o fluorine for 10 hr as the reactor temperature was increased from 300 to 550°C (see Progress Report for February 1967, ANL-7308, pp. 75-76). The results of the experimental runs which have been completed thus far in this program have been analyzed to provide an insight into the behavior of ruthenium in the process:

The results of the comprehensive analysis suggest that:

1. Treatment of irradiated UO_2 by a process involving two-zone oxidation-fluorination followed by recycle fluorination produces RuF_6 and either RuF_5 or RuOF_4 , or both. The ruthenium fluoride species are relatively stable as indicated by the virtual absence of ruthenium deposit in a nickel-ball trap maintained at 150°C .
2. Only a trace amount of ruthenium is volatilized during oxidation of UO_2 fuel at 450°C .
3. In the process based on the use of BrF_5 , the bulk of the ruthenium is volatilized during the BrF_5 fluorination step. The volatile ruthenium compound is not completely removed from the process gas stream by sorption on NaF pellets at 400°C . The ruthenium compound is effectively removed from the gas stream by activated alumina and soda lime.
4. The volatile ruthenium compounds which are formed in the PuF_6 volatilization step are probably RuF_5 , RuOF_6 , and RuF_6 .

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. General

The primary-heat-exchanger bid package was sent to prospective vendors; bids are due in June. The primary-pump proposals have been received and are being reviewed. The permanent-beryllium-reflector bid package has been sent out for bid. The control-rod guide-assembly proposals were received and are being reviewed. The reactor-vessel subcontract was negotiated and is awaiting AEC approval.

Answers to questions by DRL on the PSAR Supplement 2 were transmitted to the AEC.

2. Containment Potential of the Primary Coolant System

On the basis of the preliminary design, the containment potential of major components of the primary coolant system has been investigated. Table XXV shows the dimensions, mechanical properties, bursting pressures, and strain energies of the major components. The bursting pressure and the sum of the strain energies of the major components of the primary coolant system are 5000 psi and 524 MW-sec, respectively. The primary coolant system will rupture at a lower pressure than the pressure vessel, but will require more energy.

TABLE XXV. Properties of the AARR Primary Coolant System

Part of System	Size			Material	Ultimate Strain	Bursting Pressure (psi)	Strain Energy at Bursting Pressure	Incremental Volume at 5000 psi	Strain Energy at 5000 psi
	Inside Diameter (in.)	Wall (in.)	Volume (ft ³)				(MW-sec)	(ft ³)	(MW-sec)
Heat-exchanger shell	68	2.00	860	Carbon steel	0.22	5000	214	272	214
Pipe	22.064	0.968	1040	304 SS	0.65	5950	1400	346	302
Pipe	16.5	0.750	35	304 SS	0.65	6200	49	8.76	7.7
Total							1663	626.76	523.7

As indicated in the Appendix E of the PSAR¹⁵ (see also Progress Report for January 1967, ANL-7302, pp. 79-82), for an accidental nuclear excursion of 163 MW-sec followed by a rapid metal-water reaction (core only) of 1300 MW-sec, the energy imparted to the vessel would be about 190 MW-sec, and the pressure in the reactor vessel would be 4270 psi. Calculation shows

¹⁵Appendices to the Preliminary Safety Analysis Report on the AARR, Approval Copy, Appendix E, "Reactor Vessel Containment Potential," Vol. II (March 31, 1966, Revised December 9, 1966).

that this excursion would deposit an additional 34 MW-sec of strain energy in the primary coolant system as the pressure is equalized; the pressure after equalization would be 3300 psi. The analysis is conservative, since no credit is taken for any pressure reduction caused by the operation of the pressure-relief system.

If the metal-water reaction occurs slowly or is delayed, then the pressure vessel and the primary coolant system will deform simultaneously.

Fig. 19. Maximum Energy that Could Be Imparted to AARR Vessel and Primary Coolant System after Accidental Nuclear Excursion of 163-MW-sec Followed by a Slow 1300-MW-sec Metal-Water Reaction

Calculation shows that for this type of reaction the maximum energy that could possibly be imparted to the reactor vessel is about 120 MW-sec, and to the primary coolant system about 71 MW-sec. The maximum pressure is about 3740 psi and the steam temperature is 2000°K. Figure 19 shows the strain energy deposited on the pressure vessel and the primary coolant system at different steam temperatures. Again, no credit is taken for pressure relief.

a. Computer Program.

The computer program for the metal-water reaction is still being programmed. When the original version of the program was programmed and tested, the results did not converge.

The same type of difficulty also occurred in the manual computation. Because the iteration method is not satisfactory, a different technique is being used.

3. Instrumentation and Control--Flow-coastdown Dynamics

To determine plant safety if utility electricity is interrupted, the behavior of AARR after loss of coolant pumping power has been studied. On the basis of the flow coastdown estimated previously (see Progress Report for March 1967, ANL-7317, pp. 94-95), the core kinetic and thermal responses to flow coastdown were evaluated.

a. Core Response to Flow Coastdown. The prime concern is the degree to which the core is protected if only the safety system responds to an excessive ratio of flux to flow (ϕ/F), omitting any prior action by the system for control of ϕ/F .

(i) Description of Mathematical Model. The core kinetic and thermal responses to flow coastdown were evaluated by using flow versus time data (see Progress Report for March 1967, ANL-7317, pp. 95-98) as

input to the analog model of the High Flux Isotope Reactor (HFIR). Because the HFIR analog model could be set up readily, these first studies were conducted at ORNL. Slight changes were made in the HFIR model to reflect the differences between AARR and the HFIR cores. The changes made, which were basically in the form of new constants in some of the describing equations rather than changes in the equations themselves, were: (a) hot-channel coolant velocity was changed from 42.4 to 50.0 ft/sec, (b) the "target annulus" and "target section" simulation in the HFIR model were changed to simulate the single bulk-coolant region in the AARR internal thermal column (bulk coolant velocity also was reduced to 4 ft/sec), and (c) the incipient boiling heat flux (ϕ_B) at the hot spot was effectively not simulated in these coastdown studies.

Table C-3 in the PSAR¹⁶ cites the pertinent core-dynamics parameters that change significantly during the core cycle. The parameters changed in this study from "beginning of life" (B.L.) to "end of life" (E.L.) were: (a) fuel-metal temperature coefficient of reactivity (value at E.L. = $1/2$ value at B.L.), (b) core-coolant (moderator) temperature coefficient (value at E.L. = $1/2$ value at B.L.), (c) effective film coefficient at hot spot (reduces at E.L. due to oxide-scale buildup), (d) prompt-neutron lifetime (value at E.L. = 2 times value at B.L.), and (e) shim-safety plate differential worth. The variations of the first four items were used as described in the PSAR. The shim-safety plate differential worth (for initial insertion) decreases monotonically during core life because the plates are continually being withdrawn to maintain a 100-MW power level. In contrast, however, the amount of reactivity available for normal shutdown gradually increases during the core cycle. The first effect was simulated in this study, but the latter was not. Differential shim-safety plate worths used in this study were \$1.07/in. (assumed constant during stroke) at the beginning of life, and worth increases from \$0.11/in. to \$1.07/in. at the end of life.

The total reactivity insertion upon scram was considered constant, with a value of $-8.3\% \Delta k/k$ (\$11.6) at both beginning and end of life. This value of reactivity insertion approximates that available after operating a fresh core 1 day at 100 MW. The total safety-plate delatching time, i.e., the time required to initiate shim-safety plate motion upon reaching the ϕ/F trip was assumed to be 15 msec. The time simulated for plate insertion was ~ 100 msec.

Two cases of flow versus time (see Progress Report for March 1967, ANL-7317, pp. 98-99) were used in this study: (1) coastdown with no pony motor(s) and (2) coastdown with one pony motor. These curves were fitted with a multisegment function generator, and each was fed into the analog model as an external input. It was assumed that the core response did not affect the flow during coastdown.

¹⁶Preliminary Safety Analysis Report on the AARR, Approval Copy, Vol. II, Appendix C: March 31, 1966 (revised December 9, 1966).

An effective flow-sensing lag (time constant) of 0.75 sec was used in all cases considered in this study. This delay was assumed to be a maximum expected value for the total delay in the flow-detection channel between actual flowrate and the measured value used in ϕ/F safety channels. A time constant of 0.75 sec is, however, expected to be far too conservative (high) for the AARR, whose ϕ/F safety channel will have a much shorter effective time delay; a preliminary investigation of this problem will be described in a future report.

Fig. 20. Core Response to Flow Coastdown with One Pony Motor Operating after a ϕ/F Scram; Beginning-of-life Core Parameters

effective time delay; a preliminary investigation of this problem will be described in a future report.

The ϕ/F criterion for scram and the flow-sensing delay used in this study is that "scram" occurs for $\phi/F \geq 1.3$, where ϕ is normalized flux or power, $F = \mathcal{L}^{-1}[F'(s)/(1 + \tau s)]$ = measured normalized flow, F' = actual flow, $\tau = 750$ msec, s = Laplace variable, and $\mathcal{L}^{-1}$ = inverse Laplace Transform.

(ii) Calculated Results.

The analog calculations made can be grouped according to the following effects: (a) effect of coast-down with no pony motor versus one pony motor, (b) effects of parameter values at beginning of life (B.L.) versus end of life (E.L.), where the parameters assumed to change are listed above, and (c) effect of reactor scram versus no scram. Thus a total of eight calculations were made, and for each the following eight variables were recorded graphically: (1) reactor flowrate in %, (2) scram reactivity insertion in $\% \Delta k/k$, (3) reactor power in MW, (4) hot-spot heat flux in Btu/hr/ft², (5) hot-spot fuel metal temperature in °F, (6) average fuel metal temperature in °F, (7) average coolant temperature in core in °F, and (8) hot-channel coolant outlet temperature in °F.

A typical set of response curves are depicted in Fig. 20.

Table XXVI summarizes the major results obtained from the eight analog runs, in order of increasing severity of the thermal transient (maximum fuel temperature). It must be emphasized that this study was performed to analyze the coastdown response, both including and excluding ϕ/F safety channel protection but never "taking credit for" control-channel response. Thus, the analog model did not simulate the "Setback" and "Reverse (Fast)" control functions which will normally take place during this transient. "Setback" occurs for ϕ (DEMAND)/ $F > 1.0$; consequently, power is reduced via the shim-regulating cylinder because flow then becomes the demand set point for the flux-control system. "Fast Reverse" occurs for $\phi/F \geq 1.1$, resulting in rapid reduction of power via rapid shim-safety plate insertion. These control functions are designed to preclude the need for reactor scram upon flow coastdown.

TABLE XXVI. AARR Response to Flow Coastdown^a

Pony Motor	Core Life	Power (MW or %)	Flow F' (%)	Max ϕ/F'	T _{hs} (°F)	T _c (°F)	T _s (°F)	t ₁ (sec)	t ₂ (sec)	t ₃ (sec)
<u>Scram</u>										
One	Begin	76	44	1.73	535	347	520	0.8	1.2	2.2
None	Begin	75.5	43.5	1.74	538	349	523	0.9	1.25	2.2
One	End	89	51	1.74	735	343	511	0.75	1.0	1.75
None	End	89	51	1.74	737	343	511	0.8	1.0	1.7
<u>No Scram</u>										
One	Begin	35	10	3.5	772	600	765	0.8	1.2	-
One	End	46	10	4.6	1080	740	761	0.75	1.0	-
None	Begin	(30)	(6)	(5.0)	(880)	(690)	(874)	0.9	1.25	-
None	End	(40)	(6)	(6.7)	(1220)	(850)	(872)	0.8	1.0	-

^aThe power, flow, and temperature columns show values when maximum ϕ/F occurs. The values in parentheses were chosen arbitrarily at $t = 20$ sec.

Symbol definitions:

T_{hs} = Hot-spot fuel (meat) temperature.

T_c = Hot-channel coolant outlet temperature.

T_s = Surface (oxide-water interface) temperature (deduced from analog results via quasi-steady-state analysis).

t₁ = Time at which actual ϕ/F exceeds 1.30.

t₂ = Time at which hot-spot heat flux exceeds incipient boiling heat flux.

t₃ = Time at which ϕ/F scram occurred.

The flow-sensing lag of 0.75 sec is quite long compared with the calculated 50%-flow time delay (1.8 sec) and was found to be very important in this study. Although scram was initiated when ϕ/F measured 1.3, it was found that the actual ϕ/F exceeded 1.7 just prior to scram. Smaller values of measuring lag were not tried in this study but, of course, would greatly reduce the maximum actual ϕ/F during the transient.

It was found that the core-averaged coolant temperature did not reach boiling temperatures (remained $< 300^{\circ}\text{F}$) except for the two cases where no pony-motor coastdown was combined with no scram. However, the hot-spot surface (oxide-water interface) temperature exceeded the local boiling threshold for all cases primarily because of the long flow-sensing delay. The hot-spot fuel (meat) temperature did not exceed 1100°F except for the two no pony motor-no scram cases. Table XXVI shows peak values of the pertinent variables for all the cases.

The initial thermal transient caused by coastdown is essentially not affected by the absence of a pony motor. Later in the transient, however, temperatures reach very high values.

The primary effects of cycle time on core response to coastdown are (a) the 188°F (assumed) increase in fuel-to-coolant ΔT at end of life due to scale formation and (b) lower reactivity feedback coefficients at the end of life. As a result, the end-of-life conditions appear to be more critical than for beginning of life.

Finally, the results show that safety action upon excessive ϕ/F values is absolutely essential for maintaining core integrity. The criterion for "excessive ϕ/F " is herein assumed as actual $\phi/F \geq 1.40$, corresponding approximately to Fig. 8.1.1 in the HFIR description report.¹⁷ Although the maximum safe value of ϕ/F is expected to increase both during the core cycle and with decreasing flow, the conclusions of this study assume that that value remains constant. The "no scram" cases reported here were included primarily to evaluate the effects of scram delayed by an arbitrary time interval. However, flow coastdown without a ϕ/F safety or control action is considered incredible in AARR.

(iii) Quasi-steady-state Analysis. Since the flow-coastdown transient is slow relative to the response of the core,¹⁸ it had been assumed earlier that the thermal response of the core to coastdown could be considered a quasi-steady-state process. A comparison was thus made (for the first case in Table XXVI) between the actual analog output and steady-state calculations using instantaneous values of power and flow at various times throughout the transient. This comparison is given in Table XXVII, which shows that the thermal lags and heat-transport effects are indeed negligible for the coastdown transient. In Table XXVII, the hot-spot film coefficient is assumed to vary as $(\text{Flow})^{0.5}$, and the quasi-steady-state analysis is made as follows:

$$\frac{q''_{\text{H.S.}}}{(q''_{\text{H.S.}})_0} = \frac{P}{P_0};$$

¹⁷The High Flux Isotope Reactor (A Functional Description), Vol. I, ORNL-3572 (May 1964).

¹⁸The effective fuel-plate time constant and coolant core transit times are both less than 35 msec at rated conditions.

$$\Delta T_{H.C.} = 136 \left(\frac{P}{P_0} \right) \left(\frac{Q_0}{Q} \right);$$

$$\Delta T_{H.S.} = 21 \left(\frac{P}{P_0} \right) + 150 \frac{(P/P_0)}{(Q/Q_0)^{0.5}},$$

where P is power and Q flow.

TABLE XXVII. Comparison of Analog and Quasi-steady-state Calculations

Time (sec)	Flow (%)	Power (%)	Analog Results			Quasi-steady State		
			Bulk $\Delta T_{H.C.}$ (°F)	Plate to Coolant $\Delta T_{H.S.}$ (°F)	$q''_{H.S.}$ (10^6 Btu/ ft ² /hr)	Bulk $\Delta T_{H.C.}$ (°F)	Plate to Coolant $\Delta T_{H.S.}$ (°F)	$q''_{H.S.}$ (10^6 Btu/ ft ² /hr)
0.0	100	100	136	171	2.1	136	171	2.1
0.25	91	98	144	175	2.0	146	174	2.05
0.75	73	92	164	180	1.88	171	181	1.93
1.25	60	85	189	182	1.75	193	184	1.79
1.75	51	80	209	183	1.64	214	186	1.68
2.15	44	76	226	188	1.57	235	189	1.60
5.0	25	10	53	33	0.20	54	32	0.21

(iv) Conclusions. In summary, the five essential conclusions of this study of core response to flow coastdown are:

1. Safety action following loss of flow to the reactor is essential from a reactor safeguards point of view; however, ORNL has demonstrated that Setback and Reverse actions on HFIR actually preclude the need for scram.

2. Higher values of actual ϕ/F are reached at the end of life than at the beginning of life; thus, although the ϕ/F threshold for local boiling tends to increase during the core cycle, the core may be more vulnerable to coastdown at the end of life.

3. Core response to coastdown with none or one pony motor is essentially identical for the "initial" transient; later, however, temperatures rise to unacceptably high values without pony motor(s).

4. The value of the flow-sensing delay is apparently very important in preventing high values of actual ϕ/F prior to scram.

5. The coastdown transient is sufficiently slow that the temperatures and heat fluxes can be computed assuming quasi-steady-state thermal behavior with reasonable accuracy; comparison of this calculation with analog results shows that the relative errors in bulk ΔT and heat flux are no more than 5 and 3%, respectively.

4. Component Development

a. Beam Tubes

(i) Beam-tube Piston-ring Seal Test. A fixture (see Fig. 21) has been designed and fabricated for leakrate testing the piston seal rings in the prototype beam tubes. The fixture consists of a test vessel with a gasketed and bolted cover plate; within the vessel is a piston equipped with two circular seal rings that are retained within the grooves of the piston. The hooked-end seal rings are compressed when inserted into the vessel with the piston. The energy stored in the rings forces the seal rings to contact the inside diameter of the test vessel. The test vessel, 8.500-8.502 in. in inside diameter, was machined to an rms-16 surface finish and will correspond to the reactor-vessel beam-tube nozzle bore. The piston material is Type 304 stainless steel; the outside diameter, ring groove dimensions, and the surface finishes correspond to those of the prototype beam tube. The two commercially available hooked-end piston seal rings are designed to operate in demineralized water at 250°F and 875 psi, with a maximum Δp of 55 psi; the material is 17-4 AMS5643 heat-treated to condition H1025.

Fig. 21. Fixture for Testing Piston-ring Seals for Beam Tube

Water is supplied to the test vessel from a water tank pressurized with air to the desired pressure. Water that leaks past the seal rings was collected and weighed. The leakrate test indicated that the piston seal rings are acceptable for AARR beam-tube application.

(ii) Beam-tube Shield-plug Test Sleeve. The test sleeve described previously (see Progress Report for March 1967, ANL-7317, p. 94) was modified to demonstrate the method of mixing and pumping high-density grout into the test sleeve. The modifications included the addition of gasketed

and bolted end flanges to the sleeve. The end flanges are fitted with pipe nozzles and valves to permit hose connections for pumping the grout into the sleeve cavity. A pipe nozzle with a valve is fitted to the top of the sleeve to permit venting and overflow of the grout during filling.

An appropriate mix consisting of ferrophosphorous sand (maximum particles passing No. 4 screen), cement, water and a shrinkage-limiting additive was prepared. The high-density grout was pumped into the cavity. After 72 hr of curing time, the test sleeves were removed. Visual inspection revealed a very good casting as compared with a casting made by the hand-packing method. Radiographs made with a 40-Ci Ir^{192} source showed a void-free and uniformly dispersed aggregate throughout the concrete casting. The weight of the concrete casting was 3220 lb; average density was $\sim 225 \text{ lb/ft}^3$. This test shows that the pumping method can be applied to fill the annulus in the AARR biological shield and beam-tube adapter with high-density grout with good control of voids and density of the concrete.

5. Core Development

a. Steady-state Heat-transfer Experiments. The effects of channel spacing, flow, temperature of inlet water, pressure, and power on pressure differential along the channel, void distribution, and critical heat flux are being measured in electrically heated rectangular test sections that simulate the flat- or involute-plate geometry of the AARR core.

Single-channel tests were run at an exit pressure of ~ 655 psia and 135°F inlet with a nominal 0.040-in. test section. Channel spacing was measured under an internal pressure of ~ 650 psig, which confirmed a 0.040-in. channel (see Progress Reports for May 1966, ANL-7219, pp. 58-62, and December 1966, ANL-7286, pp. 58-64). Powers of 100, 125, 150 and 175 kW were used.

The test results compared well with results calculated by the STDY-3 Code. An example is shown in Fig. 22 (the circles represent the calculated points; the triangles represent actual data). The flowrates for the inception of local boiling, as found by using the HFIR design approach (denoted by the arrow for the Bergles-Rohsenow correlation¹⁹) and as predicted by the STDY-3 Code,²⁰ are also shown.

For the 100-, 125-, and 150-kW powers, the condition for flow instability in parallel multichannels occurs before critical heat flux is

¹⁹Bergles, A. E., and Rohsenow, W. M., Trans. ASME J. Heat Transfer, pp. 365-372 (Aug 1964).

²⁰STDY-3, A Program for the Thermal Analysis of a Pressurized Water Nuclear Reactor during Steady-state Operation, WAPD-TM-213 (June 1960).

reached. (The condition for flow instability in parallel multichannels corresponds to the flow at which the pressure drop is a minimum.) Operation with the single-channel test section at powers of 100, 125 and 150 kW and at lower flows than that corresponding to the minimum in the pressure-drop curve was maintainable, and a critical-heat-flux condition was not noted. However, with 175-kW power, a minimum in the pressure-drop versus flow curve was not attained. Also, while conducting the tests at this power, the test section experienced an actual burnout. Unfortunately, good data for the actual burnout conditions were not obtained.

Fig. 22. Effect of Flow on Pressure Drop in 40-mil Channel
Operating at 100 kW (1.391×10^6 Btu/ft²/hr)

For parallel-multichannel systems, the minimum of the curve of pressure drop versus flow is important for 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. Therefore, the experimental pressure-drop minima were compared with the flow for incipient nucleate boiling as given by this correlation. Table XXVIII 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 drop at incipient nucleate boiling is greater than that for the onset of flow instability. Notice that the ratios of flows for incipient nucleate boiling and the minimum in the pressure-drop curve, W_{inb}/W_{mpd} , are fairly close to each other, ranging from 1.45 to 1.53 for the power range for 100 to 150-kW, where flow instability is a problem. It is interesting also to note that a ratio of 1.41 existed for the 175-kW data obtained just previous to burnout, even though the flow-instability criterion does not apply. These values of the ratio of W_{inb}/W_{mpd} are lower than the same ratios obtained with the 51-mil test section, namely 1.84 to 1.99; however, compared with the ratios of 1.33 to 1.53 for the 30-mil distorted test section, the 40-mil test section shows little difference. The same observation is noted with regard to the ratio of $\Delta P_{inb}/\Delta P_{mpd}$.

TABLE XXVIII. 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

Power, kW	100	125	150	175
W_{inb} , lb/min ^(a)	23.50	30.0	37.0	45.0
W_{mpd} , lb/min ^(a)	16.25	20.25	24.25	31.87 ^(b)
Ratio W_{inb}/W_{mpd}	1.45	1.48	1.53	1.41 ^(b)
ΔP_{inb} , lb/in. ² ^(a)	22.0	30.50	44.0	62.0
ΔP_{mpd} , lb/in. ²	14.0	21.0	28.0	37.0 ^(b)
Ratio $\Delta P_{inb}/\Delta P_{mpd}$	1.57	1.45	1.57	1.68 ^(b)

(a) W_{inb} = flowrate for incipient nucleate boiling,

W_{mpd} = flowrate for minimum pressure drop,

ΔP_{inb} = pressure drop for incipient nucleate boiling,

ΔP_{mpd} = pressure drop at minimum.

(b) Related to the most severe single-channel operating conditions at which reliable data were obtained.

The relationship between flow instability and 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 was analyzed. Table XXIX gives the results. The values for the

40-mil channel range from 0.84 to 0.94 and appear reasonable, based on previous studies using the computer code STDY-3 and with the test results from 30- and 50-mil test sections (see Progress Report for December 1966, ANL-7286, pp. 58-64).

TABLE XXIX. The Quantity r Corresponding to the Minimum of the Curve of Pressure Drop versus Flow

$$\left(r = \frac{\text{Exit temperature minus inlet temperature}}{\text{Saturation temperature minus inlet temperature}} \right)$$

Power, kW:	100	125	150	175
r :	0.940	0.943	0.945	0.839 ^a

^aRelated to the most severe single-channel operating conditions at which reliable data were obtained.

V. NUCLEAR SAFETY

A. Coolant Dynamics

1. Sodium Expulsion

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

Pending delivery of the programmed electron-bombardment heater within the next several months, plans have been made for investigating related analytical problems, such as temperature profiles in sodium prior to boiling.

2. Superheat

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

Installation of the thermal-radiation heater is continuing. This heater is expected to extend the previously attained range of heat flux significantly. Reduction of the superheat data also is continuing.

3. Critical Flow

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

To investigate such occurrences, identical loops have been built for measuring sodium liquid-vapor and steam-water critical flowrates; measurements include critical pressure, critical flowrate, stagnation temperature and pressure, and axial temperature profile in the test sections. Critical-flow data for sodium have been collected, including exit critical pressure ranging from 0.5 to 6 psia, flowrates up to 1200 lb/ft²/sec, and stagnation temperatures up to 1600°F. Similar data are being collected for steam-water mixtures. In Figs. 23 and 24, sodium and water data for stagnation temperatures of 1380 and 154°F, respectively, are compared. Large deviations from the Homogeneous Thermal Equilibrium Model (HEM) are demonstrated for water as well as sodium, the ratio G_{exp}/G_{HEM} being higher for the sodium data.

Fig. 23. Critical Flow for Sodium Liquid: Vapor and Water: Steam

Fig. 24. Comparison of Measured (G_{exp}) and Calculated (G_{HEM}) Critical Flowrates for Sodium and Water

The larger deviation between calculated (HEM) and measured maximum expulsion rates for sodium is attributed primarily to much higher initial superheats required before flashing takes place than is required for water; because of much greater contents of dissolved gases, water experiences little or no superheat. The initial superheats for the sodium data in

Figs. 23 and 24 range from ~ 40 to 60°F . On the other hand, both sets of data experience large superheats in the two-phase region during the flow expansion upstream of the point of choking. For the water data in Figs. 23 and 24, superheat ranges from 17 to 34°F , whereas for the sodium data the range is from ~ 45 to 90°F . Although the large two-phase superheats and correspondingly high expulsion rates obtained appear favorable in light of fast-reactor safety, they are, unfortunately, severely detrimental to analytical treatments based on stable equilibrium conditions. Reduction and analysis of the data are continuing.

A new test section is being designed; it will have a constant-area section upstream and a slightly diverging section downstream of the point of choking. Axial temperature as well as pressure-gradient measurements will be attempted. The test section now being tested has an abrupt-expansion section downstream of the point of choking and is not instrumented for measurements of axial pressure gradient. The test section being designed should yield more accurate measurements of critical pressure, of initial superheat sustained before forced-circulation boiling takes place, and of the superheat in the two-phase region.

4. Electron-bombardment Heater Tests

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

Fabrication of the first EBH test section is $\sim 50\%$ complete. Three banks of powerstats are being installed in the high-voltage power supply to enable positive control over the EBH power ranging up to 200 kW . Also, several minor modifications to the high-voltage power supply are being made to improve the utility of the EBH test experiments.

5. Convective Instability

The experimental apparatus is in the final stages of construction after minor delays caused by slow delivery of ordered items. All mechanical parts have been operated independently; they appear to function well. Electrical-power connections to supply and control 300 A at 480 V are essentially finished. An interlock system is being designed so that there can be no error in startup of the unit.

Analytical solutions were attempted in a few runs using the third approximation. These met with little success. Apparently, an essential singularity may exist in the higher-order solutions, which could make the solutions impossible to acquire.

6. Thermophysical Properties of Sodium

The literature on sodium thermophysical properties has been evaluated to form a basis for recommending "best values" of these properties. Comparisons of safety studies of sodium-cooled fast breeder reactors will be more meaningful if the various groups doing such studies use standardized data. Recent related experimental work serves as the impetus for this evaluation. The sodium thermophysical property survey and report (see Progress Report for February 1967, ANL-7308, p. 84) has been drafted and will be reviewed before being published as report ANL-7323. The report covers the liquid density, vapor pressure, thermodynamic and P-V-T properties, transport properties, electrical resistivity, and surface tension.

B. Fuel Meltdown Studies with TREAT

1. Reactivity Measurements for Large Loop and Test-section Mockup

a. Physics Measurements on Large TREAT Loop Test-section Mockup. A detailed mockup was prepared of the large TREAT sodium-loop test section for reactor physics measurements at the TREAT reactor. The mockup and test clusters of sample pins and of sample blanket sections were irradiated in order to:

- (i) measure the reactivity worth of the loop test section loaded inside the reactor loop test hole liner, with full reactor viewing slots;
- (ii) measure the reactivity worth of two reference sample clusters, including sample blanket pins;
- (iii) check the changes in control-rod worth;
- (iv) measure the power distributions through the reference clusters;
- (v) obtain experimental ratios of sample power to reactor power for the reference exposed clusters in the TREAT reactor with this full slot and large core loading to accommodate the loop.

The first cluster consisted of 37 EBR-II pins, all enriched to 8%, surrounded by 54 steel dummy pins. The second cluster contains pins of graded enrichments to check compensation of cluster power depression by varying the enrichments over the cluster. The central pin was enriched to 7% and was surrounded by three concentric rings of a) six pins enriched to 6%, b) twelve pins enriched to 4%, and c) 72 steel dummy pins, to complete the standard 91-pin EBR-II subassembly. Fully enriched uranium foils were attached to the cladding surfaces of pins in the cluster. In some

cases, three foils were attached around the cladding periphery at a given axial position in order to measure the azimuthal power variations. Both sample clusters have been returned to Illinois and have been disassembled. Foils and selected segments of fuel alloy have been submitted for radio-chemical analyses.

b. Reactivity Measurements on Large Loop Assembly. The reactivity change resulting from the removal of four full and two slotted elements and the addition of the zirconium liner was measured and found to be $-2\% \Delta k$. The change of reactivity in the case where all six of the removed fuel elements are full elements is estimated to be about $-3\% \Delta k$. The addition of Schedule 40 stainless steel pipes of 4- and 3-in. dia inside the zirconium liner (as simulation of the actual steel test section components) had been measured earlier²¹ to result in ~ 2.7 and $0.6\% \Delta k$ loss of reactivity, respectively. In the experiments performed during the report period, the addition of the actual test section and a 37-pin cluster (inside the 3-in. pipe) produced a 3% loss of reactivity.

By comparison with the previously mentioned experiment, one could deduce that the 37-pin cluster has a worth of about $+0.3\% \Delta k$. Direct measurement of the worth of the 37-pin cluster yielded $+0.17\% \Delta k$.

The calculations performed previously predicted a $7\% \Delta k$ loss upon the addition of the test section compared to the measured value of $6\% \Delta k$:

i. Substitution of zirconium liner for six TREAT elements	-2%
ii. Correction for all elements being full	-1%
iii. Addition of pipes and subassembly	-3%
Total	-6%

There are some differences between the calculational model and the physical arrangement of TREAT during the experiment.

(1) The calculations assumed the TREAT was fully loaded (361 elements), whereas the experiment required 290 elements. If a smaller core were used in the calculations, the predicted value of the reactivity loss would be greater.

(2) The calculational model did not include the slotted fuel element present.

²¹Madell, J. T., Physics Analysis of TREAT with an EBR-II Subassembly, in Reactor Physics Division Annual Report: July 1, 1965 to June 30, 1965, ANL-7110, pp. 154-157.

The calculated values of the reactivity changes would be less in a model which included the slotted elements.

The experiment measured that the control-rod worth for the critical TREAT reactor with the test section is about the same as for the critical TREAT reactor without the test sections. The calculations indicated that the control rods in a fully loaded TREAT are worth about 7% Δk more with the test section than without it. Experiment and calculations are not in disagreement but rather are considering two different situations. Insertion of the test section increases the required size of the core and also shifts the flux so the control rods are in a more important region. These two factors cancel themselves. The overall reactivity worth of a certain amount of material (control rod) decreases as the size of the core increases, while an increase in the importance in the region of the material will increase its reactivity worth. The measurements show that addition of the test section does not decrease the shutdown capacity of the reactor control rods below that of the reference, solid core loading.

2. Unbonded EBR-II Pin in Flowing Sodium

An argon-bonded EBR-II pin was exposed in flowing sodium in a Mark-I integral sodium loop to a TREAT transient. The experiment was planned to provide information for use in assessing the results of a possible cladding failure of a pin in EBR-II (see Progress Report for January 1967, ANL-7302, pp. 92-93). The loop was returned to the Illinois site for study of the sample. Calculations of the transient temperature made, assuming intact geometry, with the ARGUS code indicated that complete sample melting took place. However, there are large uncertainties in the heat transfer across the gas gap from fuel to cladding, the timing and speed of hot fuel slumping against the cladding, and the three-dimensional transient thermal stresses inside the cladding. All of these uncertainties can affect the severity of the transient, and experimental data are needed.

3. Power Supply for High-temperature Furnace Operation

A 300-kW silicon diode, saturable-reactor-controlled, power supply has been delivered and installed in the hot laboratory building D301. The power supply will be used with a high-temperature furnace to provide data for the equation-of-state studies being performed as a part of the Fast Reactor Safety Program.

The supply has been tested successfully at D301 under a dummy load at levels up to 9000 A at 28 V. Design maximum is 10,000 A at 30 V.

4. Glovebox for Low-level Beta-Gamma TREAT Samples Containing Plutonium

Facilities are necessary to handle plutonium-bearing TREAT specimens having low levels of beta-gamma activity. Specifications have been established for a glovebox to be installed in Building 301 which will accommodate the capsules, loops, and autoclaves used in the TREAT meltdown programs. Part of the partition between rooms A106 and A114 is to be removed, permitting the space in both rooms to be used for the handling operations. In addition, cave-manipulator repair and a high-temperature furnace (see Sect. V.B.4) will share the area.

The glovebox will be of a modular design. It will be two modules high and three modules long, in order to accommodate test-section components of the loop and permit work to be performed on two experiments at the same time. The glovebox will have an auxiliary loading hood which provides space for loops or capsules while they are attached to the glovebox. Loops and capsules will be fastened to the glovebox using the split-flange technique to minimize the possibility of alpha contamination outside the glovebox; the auxiliary hood provides a backup to restrict spread of contamination and to permit a cleanup of any loop or capsule contamination occurring during glovebox operations. In addition, there will be a vacuum transfer lock large enough to admit fuel samples. The high-pressure autoclaves of the violent meltdown experimental program in TREAT can also be loaded into the glovebox through the vacuum lock. A system for purification of recirculating inert gas will be used to supply and maintain an inert cover gas inside the glovebox. The auxiliary hood will be attached to the existing radioactive exhaust system serving room A106. However, the new installation will require an expansion of the capability of the exhaust system.

The glovebox installation is to accommodate samples which have only the relatively low beta-gamma activity resulting from TREAT irradiations. Alpha-containment caves for Fast Reactor Safety experimentation on specimens preirradiated in a steady-state reactor are being designed also.

C. TREAT Operations

An unbonded, 6% enriched EBR-II-type fuel pin contained in a package sodium loop was subjected to a flat-top-type transient designed to simulate the power density in an EBR-II fuel pin during normal EBR-II operation. The reactor was shut down after 7 sec when the power level exceeded the maximum value established for the experiment. Fast-neutron-hodoscope data obtained following the transient indicate that gross failure did not occur during the transient. Small, localized cladding failures might

not be detected by the hodoscope. The sample was returned to Argonne, Illinois, for examination. The fast-neutron hodoscope and associated new digital readout equipment were operated during this test, and the film has been sent to Argonne, Illinois, for analysis.

Two samples of prototype Fast Ceramic Reactor-type fuel (C-4-D and C-4-A) were irradiated for the General Electric Company. The energy released during the transient on sample C-4-D may not have caused fuel melting, and it will be stored at TREAT until experimental data are compared with computer calculations and a decision is made on whether to subject this sample to another transient or return it for examination. Sample C-4-A will be subjected to another transient after data from the calibration transient are analyzed and specifications for the next transient are received from the General Electric Company.

A 37-pin and a 19-pin cluster of EBR-II-type fuel elements were irradiated under steady-state conditions in a mockup of the test section of the large TREAT sodium loop. Radiochemical analysis of these pins and attached foils will be used to determine the relationship between reactor energy release and pin energy release. These data will be used to specify the reactor energy release required to obtain the desired test conditions during future transient tests using the large loop.

The cold trap and plugging metal systems for the large TREAT sodium loop were operated until the plugging temperature was reduced to less than 110°C.

Investigation of the bolt failures in the safety head flange (see Progress Report for March 1967, ANL-7317, pp. 109-110) is continuing. Testing of the loop at design temperature is being delayed pending completion and action necessary to prevent similar bolt failures in the future.

D. Chemical and Associated Energy Problems (Thermal)

1. Laboratory Studies with Zircaloy-2-clad, UO₂-core Fuel Rods Simulating the Conditions of a Loss-of-coolant Accident

Experiments have been continued to simulate the environment of the fuel in a water-cooled power reactor following a loss-of-coolant accident. In such a situation, the fuel would be subjected to a slowly rising temperature (compared to that of a transient) because of heating due to fission product decay and the exothermic cladding-steam reaction.

The experimental simulations of the metal-water reaction and melt-down conditions that will exist in a loss-of-coolant type of reactor accident are being performed by the following methods:

(1) Fuel-meltdown Experiments. These simulate the chemical and fission-product-decay heating conditions in a reactor core upon loss of the primary coolant.

(2) Emergency-coolant-spray Experiments. These simulate the conditions in a reactor core when a drop in the primary-coolant-water pressure due to a loss of coolant activates a secondary-coolant spray.

a. Fuel-meltdown Experiments by an Induction-heating Method.

Key unknowns in the analysis of the loss-of-coolant accident for power reactors which use Zircaloy-clad, UO_2 -core fuel rods are the effective failure temperature and the mode of failure of the fuel rods. Failure models range from melting and dripping of Zircaloy at 1850°C (melting point of Zircaloy) to the total holdup of the UO_2 by a shell of oxidized zirconium until temperatures reached 2600 to 2800°C , where the oxidized cladding would be expected to melt. Uncertainty centers about the ability of partly oxidized cladding to maintain integrity at temperatures above the melting point of unoxidized cladding. Studies in the high-pressure furnace of the behavior of fuel rods under conditions simulating a loss-of-coolant accident were limited to maximum furnace temperatures of 1600°C (see Progress Reports for July 1965, ANL-7082, p. 67, and May 1966, ANL-7219, p. 86). Thus, it was not possible in these experiments to effect melting of the Zircaloy cladding because of the temperature limitation of the furnace. Experiments were performed, therefore, using induction heating to achieve temperatures $>2000^\circ\text{C}$ in order to observe some of the features of the failure and collapse of Zircaloy-2-clad, UO_2 -core fuel rods. Results utilizing single rods showed that there was no tendency for the Zircaloy to melt and drip from the fuel rod upon reaching a maximum temperature of 2140°C . However, because a true simulation of the conditions expected during a loss-of-coolant accident requires a large number of fuel rods to achieve a realistic thermal environment for each rod, a four-rod experiment was performed.

Figure 25 shows the arrangement of the four simulated fuel rods made up of tubes (27-mil wall thickness) of Zircaloy-2 cladding filled with UO_2 pellets. The rods, enclosed in a $1\frac{9}{16}$ -in. quartz-tube steam channel, were placed with 0.6-in. center-to-center spacing in a square lattice by means of Teflon end-plugs. An induction coil, 3 in. long, surrounded the quartz tube near the center. Two tungsten-rhenium thermocouples (placed in channels between the inner edge of the Zircaloy cladding and the outer edge of the UO_2 pellets) were used to measure the temperature of the sample; one thermocouple was positioned facing toward the center of the bundle, while the other was positioned facing toward the bundle perimeter. The input steam flowrate was established and maintained at 4 g/min (1 g/min/rod).

Fig. 25. Fuel-meltdown Assembly using Four Zircaloy-2-clad, UO_2 -core Simulated Fuel Rods

eters of 5.28 to 5.31 Å (UO_2 , 5.470 Å; ZrO_2 , 5.10 Å), thus indicating a solid solution between ZrO_2 and UO_2 . Electron-microprobe analysis of a similar particle showed the presence of mixed uranium-zirconium oxides which varied from 34 w/o uranium and 33 w/o zirconium to 24 w/o uranium and 43 w/o zirconium.

The temperature gradient observed between the inner and outer edges of each rod, and the subsequent longitudinal rupture of the cladding along the inner walls of the 4-rod bundle, indicate the need for additional experiments utilizing more rods per bundle: a center rod with at least one, and preferably two, outer rows of rods.

b. Emergency-coolant-spray Experiments. In the event of a loss-of-coolant accident, one design of a water-cooled

Figure 26 shows the sample temperature peaking after an 80-sec heating period; shortly thereafter the thermocouple facing the center of the bundle indicated a sharp rise to 2950°C. This sharp rise in temperature may have been caused by molten UO_2 contacting the thermocouple and then falling away. Color films taken of the experiment confirm the uneven temperature distribution registered by the thermocouples. The films show that hot zones occurred in areas adjacent to another rod, i.e., there was a large temperature gradient from the inner side of the rods to the outer side (the inner thermocouple was located in one of these hot zones).

Examination of the residue revealed extensive fragmentation, cladding rupture (in the areas adjacent to another rod), and evidence of molten UO_2 . Visual observation indicated that fragmentation occurred after the experiment had been terminated. X-ray diffraction examination of a 1/4-in.-dia droplet from one of the ruptured areas revealed a face-centered-cubic phase with lattice param-

Fig. 26. Thermocouple Records of Fuel Meltdown by Induction Heating (Four Zircaloy-2-clad, UO_2 -core simulated fuel rods)

reactor provides for activation of emergency-coolant spray by a drop in the primary-coolant-water pressure (below approximately 150 psi). The question to be answered is: Will the hot fuel rods be effectively quenched and maintain their integrity when the emergency-coolant spray is activated?

In laboratory simulation of these conditions, single Zircaloy-2-clad, UO_2 -core simulated fuel rods (as described above in Sect. V.B.1.a), enclosed in a quartz-tube steam channel, are being used. Two thermocouples are employed to measure sample temperature, one placed in a channel between the UO_2 and the Zircaloy, and the other spot welded to the outside of the cladding. An input steam flow of 1 g/min is used to simulate a loss-of-coolant condition. The sample is inductively heated to a temperature above the melting point of Zircaloy-2 (approximately 1850°C), at which time water is sprayed into the top of the chamber at a rate of 190 g/min [corresponding to 0.05 gal/(min)(rod) for a reactor core]. Inductive heating is continued through the spray-cooling period.

In experiment 1Z-4, the fuel rod was successfully heated (at $\sim 23^\circ\text{C}/\text{sec}$) above the melting point of Zircaloy-2 and water was sprayed on the hot rod. The rod cooled from a maximum temperature of 2045 to 1380°C during the 30-sec spray period. Figure 27 illustrates the sharp drop in rod temperature during the spray period, and the subsequent increase in temperature after the spray was discontinued. Crumbling of the fuel rod after cooldown was noted in this experiment. Figure 28 shows the heating and cooling curves of two other experiments designed to simulate nearly total cooldown. In experiment 1Z-5, a fuel rod simulating a reactor core at a temperature below the melting point of Zircaloy-2, cooled from 1750 to 110°C in 185 sec. In experiment 1Z-6, a fuel rod simulating a reactor core at a temperature well above the melting point of Zircaloy-2, cooled from ~ 2820 to 95°C in 180 sec. The rods remained intact throughout the experiments but broke during dismantling of the cell. In these simulation experiments, the degree of fragmentation does not necessarily depend upon the maximum temperature to which the fuel rod is exposed; the temperature-time history and embrittlement caused by cladding oxidation are the determining factors (the two factors, of course, are not independent).

Fig. 27. Preliminary Emergency-coolant-spray Experiment with a Zircaloy-2-clad, UO_2 -core Simulated Fuel Rod. Input Water Spray: 190 g/min.

Fig. 28. Emergency-coolant-spray Experiments with Zircaloy-2-clad, UO_2 -core Simulated Fuel Rods Undergoing Nearly Total Cooldown. Coolant Spray Rate: 190 g/min.

In noting the effects of an emergency-coolant-spray simulation, no attempts have been made to correlate heating and cooling rates to an actual reactor core; rather, the attempt has been only to demonstrate the feasibility of cooling a partially molten Zircaloy-2-clad, UO_2 -core fuel rod (undergoing heating) by means of a water spray. Factors such as an insufficient number of rods in the simulation, and changing inductive coupling (as the cladding oxidizes) between the induction coil and the fuel rod, make a direct correlation of the efficiency of an emergency core-cooling system to an actual reactor core impossible.

E. Safeguard Technology--Containment

1. Primary Containment by Energy Absorption for Water Reactors

To determine the applicability of this containment concept to water reactors, the possible effects of a hypothetical hydrogen-air-steam explosion in the primary cavity were studied. The term "explosion" covers combustion processes and therefore can be misleading when studying reactor accidents. To clarify the pertinent reactions, Moore and Gilbey²² define the processes as:

(a) Explosion--a combustion process controlled by chemical reaction rate and mixing processes; it generates pressure waves.

(b) Detonation--an advancing shock wave in which exothermic chemical changes take place in such a manner as to make the shock wave self-propagating.

To set off a combustion process, an ignition source is needed. This is possible through self-ignition of the gases or due to external sources of ignition. All studies on spontaneous ignition of hydrogen-air-steam mixtures show that such an occurrence is most unlikely; for external sources of ignition, energies of the order of millijoules are required for an explosion in hydrogen-air-steam mixtures, whereas the initiation level for detonation is 10^4 J or more. Hence, generation of detonation waves is considered to be a most unlikely accident. A credible accident is an explosion from a spark source.

Calculations for such an accident show that the pressure wave generated will be less than 500 psi under the most optimum conditions. Thus, since pressures of this type present no damage potential to the primary containment, a hydrogen explosion would not invalidate the feasibility

²²Moore, J. G. and Gilbey, R. V., A Review of the Problems Arising from the Combustion of Hydrogen in a Water Reactor Containment, AHSB-S-R-101, United Kingdom Energy Authority, Authority Health and Safety Branch, 1966.

of the concept. The inertial aspects of the primary-containment concept provide sufficient safety even against a hypothetical detonation. The response of the primary containment to such a loading has been treated in detail.²³

F. Plutonium Volatility Safety

1. Chemistry of Tellurium Fluorides

Studies of the reactions of volatile tellurium fluorides are under way to determine the conditions under which tellurium is fixed on solid materials and to investigate the kinetics of the reaction. A series of nine experiments has been completed to determine the effects of process variables on the sorption of TeF_6 on activated alumina in a flow system. In the course of this study, an isotopic dilution technique employing $\text{Te}^{125\text{m}}$ tracer was developed which allowed direct measurement of the decontamination factor obtained across the bed of activated alumina. The process variables studied were: temperature of activated alumina, 25 and 100°C; bed height, 1 and 2 in.; velocity of gas stream through the activated alumina bed, 20 and 40 ft/min; and concentration of TeF_6 in the gas stream, 250 and 500 ppm. The results indicated that only the bed height had a significant effect on the tellurium decontamination factor (DF). Gas flowrate and TeF_6 concentration showed only a slight effect on tellurium removal from the gas stream. In addition, the results showed that the required tellurium decontamination factor ($\text{DF} > 700$) for the removal of TeF_6 from air streams can be readily achieved using activated alumina as a sorbent.

²³Sorensen, H. and Fistedis, S., Hydrodynamics of a New Containment Concept by Energy Absorption, ANL-7214 (Dec 1966).

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ANL-6841 SAFETY ANALYSIS ASSOCIATED WITH THE PLUTONIUM RECYCLE EXPERIMENT IN EBWR

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ANL-6994 ENGINEERING DEVELOPMENT OF FLUID-BED FLUORIDE VOLATILITY PROCESSES. Part 10. Bench-scale Studies on Irradiated, Highly Enriched, Uranium-alloy Fuels

A. A. Chilenskas, K. S. Turner, J. E. Kincinas, and G. L. Potts

ANL-7183 FEASIBILITY STUDY OF NUCLEAR STEAM SUPPLY SYSTEM USING 10,000-MW, SODIUM-COOLED BREEDER REACTOR

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ANL-7202 THE ELECTRICAL DESIGN OF THE EBR-II

F. Verber and H. L. Schmidt

ANL-7277 TWO AXISYMMETRIC STEADY-STATE THERMO-
ELASTICITY PROBLEMS FOR TRANSVERSELY ISOTROPIC
SOLIDS

Richard A. Valentin

ANL-7281 MEASURED PHYSICS PARAMETERS IN A ZONED FAST
URANIUM CARBIDE CORE, ASSEMBLY 4Z

R. A. Karam, A. Ancarani, F. H. Helm, W. Y. Kato,
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