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Argonne National Laboratory

**REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT**

October 1967

MASTER

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REACTOR DEVELOPMENT PROGRAM

Highlights of Project Activities for October 1967

EBR-II

Following operation for physics measurements during Run 26A, the reactor was shut down for loading changes that included replacement of one of the control rods by an oscillator rod. Checkout of this oscillator rod during the first part of Run 26B indicated a repeat of the previous difficulties with rubbing of the oscillator at low frequencies at reactor power levels above 25 MWt. Oscillator data were obtained at frequencies above those where rubbing occurred, and analysis indicated that the feedback is of a type that guarantees operational stability at all feasible power levels over the frequency range studied.

Run 26B continued as a power run at 45 MWt. For this run, three new experimental subassemblies containing structural, thermocouple, and graphite samples were loaded into the reactor grid, making 18 experimental subassemblies now in the reactor.

Fabrication of EBR-II driver fuel remained on a reduced schedule because of the delay in discharge of Run 25 subassemblies and the requirement of accommodating surveillance of those subassemblies that were removed. Twelve subassemblies were fabricated.

ZPR-3

Neutron-spectrum measurements on Assemblies 49 and 50 completed current work on the series of simple, plutonium-fueled assemblies. The entire matrix was then unloaded in preparation for the Fast Flux Test Facility (FFTF) Phase B critical program, which began on October 18. Additional access holes were drilled in the ZPR-3 matrix to accommodate worth and reaction-rate traverses for future FFTF criticals in split-core and conical-core geometries. Detailed plans have been formulated for all experiments with the first assembly in the Phase B program, Assembly 51, which will be a simple, nonsplit cylinder.

ZPPR

Construction progress at ZPPR included completion of earth fill for the mound, attainment of 50% of completion of the sand-and-gravel filter over the reactor cell, and completion of seal welding on 22 of the 24 high-efficiency filter frames in the backup containment structure. Overall construction is about 93% complete.

Seven proposals for installation of the reactor and associated equipment were received and are being evaluated.

AARR

The AEC Advisory Committee on Reactor Safeguards has tentatively approved the AARR Preliminary Safety Analysis Report. The seismic-design criteria must be acceptable to AEC before concrete is poured.

A contract has been signed with Franklin Institute for stress analysis of reactor components.

Assembly of the beam-tube test facility has started, as has evaluation of transient thermomechanical effects in the reactor core.

The present analog model for prediction of power-excursion behavior has proved to be optimistic in its results; future calculations will employ a model similar to the CHIC-KIN code.

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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

October 1967

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Issued November 30, 1967

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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:

April 1967	ANL-7329
May 1967	ANL-7342
June 1967	ANL-7349
July 1967	ANL-7357
August 1967	ANL-7371
September 1967	ANL-7382

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I. PLUTONIUM UTILIZATION--CIVILIAN--EBWR

A. Operations

1. Lay-up Activities

The draining and decontamination of the liquid retention system completed the final "mothball" operation of the EBWR plant. The entire plant is now in a lay-up state, thus completing the EBWR participation in the Plutonium Recycle Program.

2. Fuel-pin Shipment to Battelle-Northwest Laboratory

The third shipment of irradiated fuel pins was loaded into the "Yankee" transfer cask and sent to Battelle-Northwest Laboratory. This shipment consisted of one whole plutonium fuel assembly (^{59}Pu) plus 36 pins from the uranium and plutonium areas of the core. The shipment arrived at Battelle with all pins in good shape. This was the final set of pins and concluded the transfer of irradiated fuel to Battelle.

II. LIQUID-METAL FAST BREEDER REACTORS--CIVILIAN

A. Fuel Development--LMFBR

1. Metallic

a. Fuel-element Performance (W. F. Murphy)

(i) U-Pu Alloy Fuel Performance

Last Reported: ANL-7382, pp. 73-75 (Sept 1967).

Fuel elements irradiated in subassembly XA07 were identified in Table II.D.1. of ANL-7382 (Sept 1967). In that table the composition of the second group of elements was given as U-15 w/o Pu-12 w/o Zr; this should have been U-14 w/o Pu-12 w/o Zr. Fission gas samples from the plenum of eight fuel elements from this group have been analyzed by the Chemistry Division. The results are given in Table II.A.1. The fission gases were normally collected without heating the fuel elements. However, the results from elements ND-28, -32, -35, and -37 include a second small gas sample obtained by heating to about 200°C to melt the sodium. The atom-percent burnup values in Table II.A.1 were calculated from fission rates for the irradiation location in EBR-II. Three samples from element ND-30 have been analyzed for burnup, on the basis of technetium content, with the following results: top of fuel column, 3.0 a/o of heavy elements; middle, 4.4 a/o; bottom, 3.5 a/o. The percent of fission gas released from the elements ranged from 60 to 71% (average 63%) of the theoretical yield. The ratio of Kr/Xe was $0.153 \pm \begin{smallmatrix} 0.004 \\ 0.006 \end{smallmatrix}$ (12 values).

TABLE II.A.1. Fission-gas Release for U-Pu-Zr Fuel Elements

Fuel Element	Burnup (a/o)		Volume of Fission Gas ^a (cm ³ at STP)		% Released
	Max	Avg	Calculated	Measured	
ND-28	4.5	3.9	49.1	30.0	61
ND-32	4.2	3.7	46.6	31.0	67
ND-43	4.3	3.8	47.6	29.6	62
ND-25	4.2	3.7	43.0	25.7	60
ND-26	4.1	3.6	42.3	26.5	63
ND-30	4.7	4.1	47.0	28.3	60
ND-35	4.5	4.0	47.0	29.6	63
ND-37	4.6	4.0	47.1	33.5	71

^aKrypton and xenon.

Note: The apparent discrepancy between average burnup values and calculated fission-gas volume results from compositional differences in the fuel.

A small increment of burnup has accrued to the two fuel elements in subassembly XG05 in the EBR-II. About 70% of the target burnup of 7.5 a/o has been accumulated. These fuel elements are identified in Table II.A.2.

TABLE II.A.2. Status of Metal-fuel Irradiations in Progress--Fast-reactor Development

S/A Number	Specimen Number	Design Parameters					Operating Conditions		
		Fuel Composition (w/o)	Effective Density (%)	Cladding Composition (w/o)	Cladding OD (in.)	Cladding Thickness (in.)	Maximum kW/ft	Maximum Cladding Temp (°C)	Burnup to Date a/o fission/cc x 10 ⁻²⁰ ^a
XG05	ND24	U-15 Pu-10 Zr	63.6	V-20 Ti	0.208	0.015	10.0	535	5.5 12.5
XG05	NC17	U-15 Pu-10 Ti	66.7	V-20 Ti	0.204	0.016	10.0	540	5.7 13.1

^aBased on effective density.

Group M-3, consisting of fifteen U-15 w/o Pu-10 w/o Zr fuel elements, has been loaded into subassembly XO28 at the EBR-II. Insertion into the reactor is scheduled for late November.

Injection casting of 40 fuel pins (Group M-4) of U-15 w/o Pu-12 w/o Zr with 93% enriched uranium has been practically completed. The fuel pins are being examined for density, dimensions, and composition.

b. Fabrication Development (F. D. McCuaig)

(i) Crucible and Mold Development

Last Reported: ANL-7317, p. 50 (March 1967).

A crucible study was initiated in conjunction with the U-Pu-Zr alloy development program and in cooperation with the Engineering Ceramics Group. The purpose of the study was to seek a container material to be used for alloying and casting the reactive alloys of U-Zr and U-Pu-Zr, which experience indicates are highly reactive with containers of Al₂O₃, MgO, and graphite. Some degree of success was observed with zirconia crucibles, although oxygen contamination was evident.

A literature search to find a suitable crucible, or a crucible coating material, indicates that zirconia-zirconium and gadolinia offer the greatest promise. Other possible candidates, according to available thermodynamic data, are dysprosia and yttria. Ceria and stabilized urania were added as possible candidates. Stabilized zirconia is of interest as a comparison standard.

Tests intended to obtain quantitative information on the effects of alloying U-Zr and U-Pu-Zr in candidate crucible materials were conducted. The first tests were made with zirconia, zirconia-zirconium,

urania-gadolinia, and gadolinia test crucibles. Ten pressed crucibles were fabricated and sintered, these including three $\text{ZrO}_2 \cdot 15 \text{ w/o Zr}$, three $\text{UO}_2 \cdot 10 \text{ w/o Gd}_2\text{O}_3$, and four Gd_2O_3 crucibles. Lime-stabilized zirconia crucibles, machined from firebrick, were also used. A uranium-12 w/o zirconium master alloy was used for the first test melts. The alloy was prepared from electrolytically cleaned material that had been arc-melted several times to achieve homogeneity. Analytical results showed the zirconium content to be 11.7% and the oxygen level 262 ppm by weight. A tantalum resistance furnace was used to melt the alloy in the selected crucibles. The material was heated to 1700°C in a vacuum of 10^{-5} to 10^{-6} mm Hg. The total time for the run was 1 hr.

The U-Zr alloy did not wet the UO_2 and ZrO_2 crucibles. Partial wetting occurred in the Zr- ZrO_2 crucible, and complete wetting was indicated in the Gd_2O_3 crucible. The alloy in the UO_2 and ZrO_2 crucibles, however, did not exhibit the bright metallic luster observed in the alloy from the Zr- ZrO_2 and Gd_2O_3 crucibles. The crucibles and alloys were sectioned for metallographic examination and chemical analyses. Metallographic examination was used to determine the presence and location of alpha-zirconium in the specimen.

Most of the alpha-zirconium was found in the upper portion of the sample, because this phase is less dense than the uranium-zirconium phases. The only two samples that did not wet the crucible contained the most alpha-zirconium. Results from initial tests indicated the need for larger charges in the crucible so that more representative chemical samples could be taken. The results from the initial tests plus a second run with larger quantities of alloy are given in Table II.A.3.

TABLE II.A.3. Crucible Interaction with U-Zr Alloy; Initial Tests

Alloy Source	Oxygen Analysis (ppm by wt)		Alloy Source	Oxygen Analysis (ppm by wt)	
	Run No. 1	Run No. 2		Run No. 1	Run No. 2
ZrO_2	1800	630	Gd_2O_3	1700	320
$\text{UO}_2 \cdot \text{Gd}_2\text{O}_3$	1300	550	$\text{ZrO}_2 \cdot \text{Zr}$	790	660

In the second test the alloy melted in the ZrO_2 and UO_2 did not wet the crucible, but the oxygen analyses and metallographic examination revealed they were more contaminated with oxygen than the other two samples. In both tests, a reaction zone was observed between the alloy and the ZrO_2 -Zr crucible material. Preliminary results indicate the composition of the layer is UO_2 , with a small amount of ZrO_2 . Between this layer

and the crucible is another narrow region that contains free zirconium. Further evaluations are being made.

Crucible materials of UO_2 -10 w/o Gd_2O_3 and ZrO_2 -10 w/o Zr were eliminated from the next tests and various mixtures of Gd_2O_3 and Y_2O_3 were concentrated upon. The results of on-going runs are summarized in Table II.A.4.

TABLE II.A.4. Crucible Interaction with
U-Zr Alloy; Continuing Tests

Material	Crucible		Oxygen Analysis of Melt (ppm by wt)
	Type	Firing Atm	
ZrO_2 -10 w/o Gd_2O_3	Pressed	Air	1200
		H_2	1000
		Vac	970
ZrO_2 -10 w/o Y_2O_3	Pressed	Air	990
		H_2	920
		Vac	900
Code 920 ^a	Pressed	Air	830
		H_2	1100
		Vac	840
Gd_2O_3	Pressed	Air	630
		H_2	320
		Vac	400
Master Alloy			245/251

^aOxide mixture composed of Sm_2O_3 , Gd_2O_3 , and Y_2O_3 .

To date, the U-10 w/o Zr alloy melted in Gd_2O_3 crucibles have produced melts with the least oxygen contamination.

2. Oxide

a. Fuel-element Performance (F. L. Brown)

(i) Mixed-oxide Fuel Performance

Last Reported: ANL-7382, pp. 75-77 (Sept 1967)

Element numbers HOV-4 and HOV-10, disclosed by neutron radiography to have developed cladding failures under irradiation in EBR-II, were returned to the Metallurgy Division for examination. Nondestructive

leak tests will be made to locate possible penetrations in the capsule walls and end plugs which would help to account for the escape of fission products from experimental subassembly XO11 in which the failed elements were irradiated.

3. Carbide--Fabrication and Evaluation

a. Fuel-element Performance (J. H. Kittel)

(i) Uranium-Plutonium Carbide

Last Reported: ANL-7382, pp. 78-80 (Sept 1967).

Elements undergoing irradiation in EBR-II are shown in Table II.A.5. The three elements that have completed irradiation in EBR-II to burnups of 1.91 to 3.02 a/o are described in Table II.A.6. These elements are being examined in the Metallurgy Division's Alpha-Gamma Hot Cell. Metallographic samples have been prepared and evaluations of microstructures are now being made.

TABLE II.A.5. Status of Mixed-carbide Fuel Irradiations in Progress in EBR-II

S/A No.	Specimen Number	Design Parameters						Operating Conditions			
		Fuel Composition (w/o)	Fuel Form ^a	Effective Density (%)	Jacket Composition (w/o)	Cladding OD (in.)	Jacket Thickness (in.)	Max (kW/ft)	Max Jacket Temp (°C)	Burnup to Date	
										a/o	fiss/cc x 10 ^{-20b}
XG05	SMV-2	UC-20 PuC	VIPAC	83.8	304 SS	0.296	0.021	24.0	675	6.0	16.8
XG05	HMV-5	UC-20 PuC	VIPAC	80.0	Hastelloy-X	0.295	0.015	26.6	685	6.3	16.5
XG05	NMV-11	(U _{0.8} Pu _{0.2})C	VIPAC	82.9	Nb-1 Zr	0.281	0.012	25.5	635	6.3	17.2
XO08	NMP-2	(U _{0.8} Pu _{0.2})C	PELLET	81.6	Nb-1 Zr	0.284	0.013	17.4	545	3.8	10.2
XO08	NMV-4	UC-20 PuC	VIPAC	80.0	Nb-1 Zr	0.283	0.013	27.0	645	5.6	14.8
XO08	NMV-7	UC-20 PuC	VIPAC	80.0	Nb-1 Zr	0.283	0.013	26.0	630	5.5	14.6
XO08	NMV-12	(U _{0.8} Pu _{0.2})C	VIPAC	85.5	Nb-1 Zr	0.284	0.014	26.7	645	5.6	15.7
XO08	HMV-1	UC-20 PuC	VIPAC	80.0	Hastelloy-X	0.295	0.015	25.2	660	5.1	13.5
XO08	HMV-4	UC-20 PuC	VIPAC	80.0	Hastelloy-X	0.295	0.016	27.2	690	5.5	14.5
XO08	HWMP-1	(U _{0.8} Pu _{0.2})C	PELLET	81.4	Hastelloy-X + W	0.296	0.020	16.9	565	3.7	10.1
XO08	HWMV-1	(U _{0.8} Pu _{0.2})C	VIPAC	82.9	Hastelloy-X + W	0.295	0.018	26.4	685	5.5	15.1
XO15	NMP-1	(U _{0.8} Pu _{0.2})C	PELLET	81.6	Nb-1 Zr	0.284	0.013	16.5	535	1.2	3.2
XO15	NMV-3	UC-20 PuC	VIPAC	80.0	Nb-1 Zr	0.284	0.013	26.0	635	1.7	4.6
XO15	HMV-2	UC-20 PuC	VIPAC	80.0	Hastelloy-X	0.295	0.015	27.0	685	1.7	4.5
XO15	TVMV-1	(U _{0.8} Pu _{0.2})C	VIPAC	83.7	V-20 Ti	0.298	0.021	23.4	640	1.6	4.3

^aVIPAC = Vibratorily compacted into cladding; PELLET = Pressed and sintered pellets.

^bBased on effective density.

TABLE II.A.6. Design Parameters and Operating Conditions for
Carbide Elements Irradiated in EBR-II and Now
Undergoing Postirradiation Examination

	Element Number		
	SMP-1	VMV-1	SMV-1
Fuel composition, w/o	(U _{0.8} Pu _{0.2})C	(U _{0.8} Pu _{0.2})C	UC-20 w/o PuC
Fuel synthesis	Arc-melted	Arc-melted	Arc-melted
Fuel form	Pellets	Vib compacted	Vib compacted
Fuel diameter, in.	0.253	0.253	0.257
Fuel length, in.	13.4	14.0	14.0
Effective density, %	81.4	85.9	80.0
Cladding material	Type 316 SS	Vanadium	Type 316 SS
Cladding OD, in.	0.306	0.301	0.306
Cladding wall, in.	0.024	0.024	0.025
Max heat rating, kW/ft	15.9	25.8	21.3
Max cladding temp, °C	565	645	630
Max burnup, a/o	1.91	3.02	2.59

Element No. SMP-1, irradiated to 1.91 a/o burnup, contained both stoichiometric (single-phase monocarbide) pellets and hyperstoichiometric (monocarbide and dicarbide and/or sesquicarbide) pellets before irradiation. The same pellets were single-phase or restructured polyphase after irradiation. Abundant fine porosity within grains was observed in the hyperstoichiometric material, and to a lesser extent in the stoichiometric material. The stoichiometric pellets, however, were not located in the hottest part of the fuel column.

Fine porosity also was observed within the grains of the sintered center arm of Element No. VMV-1, loaded with vibratorily compacted hyperstoichiometric particles. The particles consisted of monocarbide plus M₂C₃ before irradiation. The sintered area of this element, which covered more than one-half of the radius at the fuel midplane, appeared to have very little second phase after irradiation. Large primary particles at the periphery of the sintered area retained their as-cast structures.

b. Compatibility of (U,Pu) Carbides with Potential Jacketing Materials (T. W. Latimer)

(i) Incoloy 800. Metallographic examination of Incoloy 800 (46 w/o Fe, 32 w/o Ni, 21 w/o Cr) after contact with (U_{0.8}Pu_{0.2})C for 1000 hr at 800°C revealed interactions similar to those previously observed with Hastelloy-X (18 w/o Fe, 48 w/o Ni, 22 w/o Cr, 9 w/o Mo) and Inconel 625 (3 w/o Fe, 61 w/o Ni, 22 w/o Cr, 9 w/o Mo). The reaction products consisted of a 5μ band of (U,Pu)Ni₅ on the fuel side of the interface and a large number of small precipitates surrounding and within the grains of the Incoloy 800 to an average depth of 125 μ. The depth of the (U,Pu)Ni₅ band in the Incoloy 800 test was the same as that after identical testing conditions

with Hastelloy-X, although the nickel content of Incoloy 800 is considerably less than that of Hastelloy-X. Chromium- and molybdenum-rich zones adjacent to the interface, previously found in compatibility tests with Hastelloy-X and Inconel 625, were not observed in the tests with Incoloy 800 (which does not contain molybdenum).

(ii) Vanadium-base Alloy. After 1000-hr heating at 800°C, reactions were not observed between V-10 w/o Cr and either single-phase (U,Pu)C or hyperstoichiometric (U,Pu)C containing approximately 20 v/o (U,Pu)₂C₃. However, etching has revealed a widening of grain boundaries of the V-10 w/o Cr within approximately 150 μ of the interface, and a second phase has been observed between these grain boundaries. This effect was not observed after similar tests with V-15 w/o Cr-5 w/o Ti, which has shown no effects in previously reported tests with (U,Pu) carbides.

In order of decreasing compatibility with (U,Pu)C at 800°C, vanadium alloys produced at Argonne ranked as follows: V-15 w/o Cr-5 w/o Ti, V-10 w/o Cr, V-15 w/o Ti-7.5 w/o Cr, and V-20 w/o Ti.

(iii) Compatibility of Uranium Phosphide with Selected Jacketing Materials. To compare the interaction of various jacketing alloys with ceramic fuel materials other than (U,Pu) carbides, certain alloys, previously tested with stoichiometric and hyperstoichiometric (U,Pu)C, were heat-treated at 800°C for 1000 hr with uranium monophosphide. Metallographic examination of Type 304 stainless steel has shown the jacketing to be affected to an average depth of 22 μ , while the average affected fuel zone was 18 μ .

For Hastelloy-X, the average depth of the affected zones was 15 μ into the jacketing and 45 μ into the fuel. Preliminary microprobe results indicate that in both cases the jacketing material has been depleted of nickel. A more complete analysis of these reactions is being performed.

c. Fabrication Development (N. J. Carson, Jr.)

Last Reported: ANL-7329, pp. 45-48 (April 1967).

The purpose of this program is to prepare dense granules of (U,Pu)C, suitable as feed to the vibratory compaction process, from powders synthesized by the Chemical Engineering Division. Suitable granules are made in four main steps: (1) The carbide powders are pressed into tablets; (2) the tablets are granulated; (3) the green granules are sintered; and (4) the sintered granules are sized. The granules will then be loaded into jackets by means of the vibratory compaction equipment in PF-16, a new glovebox that became operational during this reporting period.

The work is being carried out in an argon-atmosphere glovebox containing two main pieces of equipment: a 20-ton-capacity press and a 2700°C sintering furnace. During this reporting period, a set of tableting dies was constructed and installed in the press and several tablets were pressed from uranium carbide. Uranium carbide is being used during this learning period to minimize contamination of the equipment and conserve the more expensive (U,Pu)C. Powders have been pressed with and without a binder, and the results are being evaluated. The sintering furnace has been operated at temperatures above 1900°C. The temperature indications of the graphite-boronated graphite thermocouple have been correlated with those of an optical pyrometer, and temperature uniformity within the heating chamber has been established. Tantalum carbide sintering boats have been used and appear to have excellent thermal shock resistance. The flow of cooling water to the furnace appears adequate, but some redistribution was required when the water-cooled thermocouple was added.

Solid solution (U,Pu)C has been ordered from the Chemical Engineering Division and will be worked on during the next reporting period.

4. Fuel Cladding and Structure--Jacket Alloys

a. Mechanical Properties of Fuel-jacket Alloys (F. L. Yaggee)

(i) Effect of Alloy Composition, Annealing Temperature, and Testing Temperature on Short-term Tensile Properties of Vanadium-base Alloys

Last Reported: ANL-7382, pp. 82-83 (Sept 1967).

The effects of annealing temperature and of test temperature on the short-term engineering properties of V-20 w/o Ti and V-40 w/o Ti are illustrated in Tables II.A.7 and II.A.8. These properties are important in assessing the probability that a fuel cladding will or will not survive an unexpected reactor transient. The engineering values are presented because these are the values most often quoted. The true-stress true-strain values are unique to the material and are therefore more useful in design. These latter values are being determined by computer and will be presented as they become available.

The tests were conducted on 1.5-mm (0.060-in.)-thick sheet specimens in a vacuum environment of 4×10^{-6} mm Hg or better. The strain rate of 0.02 min^{-1} was held constant for all tests.

Although the V-20 w/o Ti alloy annealed at 900°C has short-term tensile properties not too different from the same alloy annealed at 1000°C, the higher annealing temperature doubles the creep resistance of this alloy.

TABLE II.A.7. Short-term Tensile Properties of V-20 w/o Ti

Test Temp (°C)	1000 psi			Percent Strain	
	UTS	0.2% Y.S.	Fracture Stress	Fracture	Uniform Eng
<u>Vacuum-annealed 1 hr at 900°C</u>					
R.T.	-	-	-	-	-
200	82.7	62.0	74.8	22.3	15.6
400	89.2	57.5	72.8	17.2	14.5
600	81.3	51.2	64.4	28.3	17.0
800	45.7	38.4	Zero	59.4	3.3
<u>Vacuum-annealed 1 hr at 1000°C</u>					
R.T.	91.5	79.9	67.2	34.4	18.7
200	83.2	62.5	65.8	24.9	14.0
400	86.3	55.7	69.0	13.0	12.0
600	85.9	54.6	68.8	28.9	21.3
800	51.3	43.4	16.6	43.3	3.5

TABLE II.A.8. Short-term Tensile Properties of V-40 w/o Ti

Test Temp (°C)	1000 psi			Percent Strain	
	UTS	0.2% Y.S.	Fracture Stress	Fracture	Uniform Eng
<u>Vacuum-annealed 1 hr at 900°C</u>					
R.T.	106.9	97.5	99.2	31.2	21.5
200	99.4	79.2	56.7	21.6	14.3
400	100.3	70.7	21.1	24.3	18.3
600	79.5	64.4	54.7	30.5	9.6
800	38.6	37.5	Not Fractured	-	7.5
800	53.8	46.2	3.3	19.6	10.2
<u>Vacuum-annealed 1 hr at 1250°C</u>					
R.T.	104.1	97.0	99.8	17.9	12.2
200	95.3	75.1	91.6	17.5	12.4
400	94.0	69.2	81.7	20.3	12.7
600	76.4	62.5	64.7	19.5	10.0
800	49.3	47.8	20.9	27.7	7.5

(ii) Effect of Surface Defects on Mechanical Properties of Type 304 Seamless Stainless Steel Fuel Cladding. Specifications for Type 304 stainless steel fuel tubing demand a 100% inspection for defects by nondestructive techniques, but designate as acceptable material

containing defects (scratches, laps, seams, etc.) that are 10% or less of the wall thickness. This specification is presumably based upon the belief that such defects will not influence, on a statistical basis, the location of the tube failure or the values of stress and strain obtained at failure. Although reports in the open literature tend to support this view (and probably are the basis for the specifications as originally written), recent tests indicate that artificially defected tubes will fail preferentially at the defect site even when a defect depth of 5% is used.

The tubing used in the present experiments was purchased by ANL for use as cladding in fuel-irradiation experiments. The purchase specifications require that the chemistry of the material comply with ASTM Specification A-213-64. Mechanical properties for this material reported by the vendor include a 90,000-psi UTS and 61% elongation in 2 in. (values at room temperature). The tube dimensions were 0.290-in. OD and 0.250-in. ID.

Before the introduction of the artificial defects, the tubing was inspected by nondestructive techniques and certified to be free of defects that would give a signal greater than or equal to that of a defect made by machining with an Elox electric-discharge machine to a depth equivalent to 5% of the wall thickness. In addition, the ID and OD of the tube specimens were measured at five perimeters along the specimen length. Four such diameter measurements, made at each perimeter, were spaced at 45°. Artificial defects were introduced by electric-discharge machining (Eloxed) on the external surface of some tubes, the inside surface of others, and on the inside and outside surfaces of a third group of tubular specimens. The defects were oriented parallel to the tube axis and had a depth equivalent to 5% of the wall thickness. The exact location of each defect in an individual specimen was indicated by a reference mark on an appendage to the specimen. Tests were conducted with the tube specimen in vacuum (2×10^{-7} mm Hg) at 650°C by applying internal gas pressure to impress a biaxial stress on the specimen. Results are summarized in Table II.A.9.

TABLE II.A.9. Effect of Surface Defects (Depth 5% of Wall Thickness) on Mechanical Properties of Type 304 Stainless Steel

Location of Defect	Calc Hoop Stress (psi)	Time to Rupture (hr)	Location of Failure	Rupture Strain (%)
None	36,260	1.5	Mid Specimen	28.8
OD	32,990	3.6	At defect	22.0
OD and ID	28,460	2.4	At ID defect	7.7
ID	21,360	178.4	At defect	18.0

The rupture strain of 28.8% shown for the specimen that was free of defects, as certified by nondestructive testing techniques, is normal for this material under biaxial stress. Tubular specimens tested under the same conditions of temperature, stress, and environment will always show a lower rupture strain than does the sheet material pulled in uniaxial tension.

b. Irradiation Studies (R. Carlander)

Last Reported: ANL-7382, pp. 80-81 (Sept 1967).

Tensile specimens of unirradiated and irradiated Hastelloy-X that were tested at a crosshead speed of 0.5 mm/min at room temperature and at 400, 550, and 650°C are being examined metallographically. The irradiated specimens are from EBR-II subassembly XO13, which was irradiated to a nominal fluence of 5×10^{21} in the temperature range of 510-704°C.

Transverse sections of the strained and unstrained areas and longitudinal sections of the fracture surface are being analyzed to determine the cause(s) for the unexpectedly high ductility (50 to 55% total elongation) that the specimens exhibited after irradiation (Progress Report for June 1967, ANL-7349). The unirradiated Hastelloy-X microstructure contained a large (2- to 6-micron-diameter) randomly distributed globular precipitate and a fine uniformly distributed precipitate. Neither irradiation nor temperature changed the distribution, volume, or morphology of the precipitate particles. The fracture areas of both the unirradiated and irradiated specimens were ductile transgranular failures. There was no evidence of a transition from transgranular to intergranular fracture. Since no gross changes in microstructure were observed, the reason(s) for the high ductility of the irradiated Hastelloy-X have not yet been determined. Explanation of the ductile behavior may result from projected examinations of irradiated material by electron transmission microscopy.

c. Sodium Corrosion of Fuel-jacket Alloys (W. E. Ruther and C. F. Cheng)

(i) Development of Refractory-metal Alloys for Service in Oxygen-contaminated Sodium

Last Reported: ANL 7382, p. 83 (Sept 1967).

The dynamic (6.1 m/s) corrosion experiment at 450°C has been terminated, and a new experiment has been started at 600°C. The samples from the 450°C experiment are being examined microscopically and will be analyzed chemically. Except for V-20 w/o Ti, the specimens have retained a metallic appearance and have very small (~ 0.1 mg/cm²)

weight changes. The thin corrosion film on the V-20 w/o Ti surface has spalled off in areas. These areas have darkened, giving the specimen a splotchy appearance. The weight losses of these samples are higher than for the other alloys.

Exposure of vanadium and vanadium-alloy samples in static, oxygen-refreshed sodium at 600°C was started during the last month.

An attempt to measure the carburizing ability of the sodium in one static refreshed system with pure iron foils resulted in the complete decarburization of the foils after 10 days at 650°C. Similar iron foils simultaneously exposed picked up about 30 ppm by weight nitrogen.

(ii) Analysis of Oxygen and Carbonate in Sodium-Nickel Mass-transfer Loop

Last Reported: ANL-7382, pp. 83-86 (Sept 1967).

The nickel mass-transfer loop has been modified to evaluate the mass transfer of nickel from 645°C to lower temperatures along the entire test section as well as in the cold trap. Small, auxiliary, detachable cold traps have been added to permit periodic analyses for nickel at the coldest point in the system.

d. Fabrication Development of Fuel-jacket Alloys (R. M. Mayfield)

(i) Fabrication of V-15 w/o Cr-5 w/o Ti

Last Reported: ANL-7357, p. 84 (July 1967).

Two additional production-size (3.8-in.-dia) ingots have been consolidated since the last report. Results of chemical analysis are outlined in Table II.A.10.

TABLE II.A.10. Chemical Analysis of V-15 w/o Cr-5 w/o Ti Ingots

Ingot Number	Alloy Content (w/o) ^a		Interstitial Content (ppm by wt) ^b			
	Cr	Ti	Carbon	Oxygen	Nitrogen	Hydrogen
AM-105	15.1	5.1	255	740	195	35
AM-107	15.0	5.1	315	745	210	30

^a Average of three values from top, center, and bottom, respectively, of ingot.

^b Average of two values from top and bottom, respectively, of ingot.

The interstitial content of both ingots was controlled by blending vanadium crystals (~300 ppm by weight oxygen), electrolytically refined in fused salt, with vanadium granules, calcium-reduced, having a higher oxygen content.

Workable secondary fabrication schedules have now been established for double-extruded rod, sheet bar, and tube-blank products. The as-extruded rod and sheet bar are heat-treated for 2 hr at 900°C and then reduced 40 to 50% by rolling at 300°C. Average reduction per pass for rod and sheet has been about 10%. After an in-process anneal of 1 hr at 900°C, the materials can be cold-reduced an additional 50%, or to finished size, by similar reductions. All ends of rod stock are nosed before rolling to prevent splitting.

The grain-growth behavior of 70% cold-reduced V-15 w/o Cr-5 w/o Ti alloy specimens annealed at temperatures of from 900 to 1500°C is illustrated in Table II.A.11.

TABLE II.A.11. Grain-growth Behavior of
70% Cold-reduced and Annealed
V-15 w/o Cr-5 w/o Ti

Annealing Temp (°C) (Time: 1 hr)	Average Grain Diameter (μ)	Annealing Temp (°C) (Time: 1 hr)	Average Grain Diameter (μ)
900	16	1400	22
1100	22	1450	32
1300	22	1500	152
1350	22		

Rapid grain growth occurs between 1450 and 1500°C. The presence of a fine, uniformly distributed precipitate phase effectively restricts grain growth at annealing temperatures below 1400°C for times of 1 hr. Above this temperature the precipitate is taken into solution, the grain-growth inhibiting effect is lost, and rapid grain growth results. Upon cooling the precipitate reappears, but in a form more reminiscent of the casting than the initial cold worked and annealed structure. These microstructural changes are illustrated in Fig. II.A.1.

Fig. II.A.1. Grain Size and Precipitate Morphology of 70% Cold Worked V-15 w/o Cr-5 w/o Ti Rod Specimens Annealed for One Hour at the Indicated Temperatures

B. Physics Development--LMFBR

1. Theoretical Reactor Physics--General Fast Reactor Physics

a. Multigroup Methods (H. H. Hummel)

(i) Calculations of Detailed Neutron Fluxes across Interfaces between Resonance-scattering Media. Detailed neutron-flux spectra in homogeneous bare-core fast systems containing resonance scattering materials may be calculated with the Argonne program ELMOE.* For a given composition, the ELMOE code determines the bare-core neutron-flux spectrum by using many hundreds of very fine-energy groups.

However, resonance-scattering materials can also be present in regions in which the assumption of a normal mode flux may not be satisfactory, for example, in or near reflectors. In this regard, a modified usage of the ELMOE program was reported.** This method employed iterations between coarse-group, multiregion criticality calculations and ELMOE calculations. The effects of gross space- and energy-dependent bucklings were simulated by substitutions of approximate coarse-energy and space-dependent positive and negative group-absorptions. These leakage-simulating absorptions, however, were based upon regional coarse-group leakages derived from coarse-group criticality calculations. Thus, the possible effects of interactions of differing fine-energy variations of the fluxes across an interface between media of different resonance scattering characteristics are not included in that method.

Spatially dependent fine-energy-detailed fluxes through such interfaces between scattering media are currently being calculated more directly for simple situations.† The fine-group, spatially dependent calculations are carried out by using fine-group cross sections for transport and elastic scattering obtained by using the ELMOE library of angular scattering data as fine-group cross sections. These fine-group cross sections are then used in a modification of the Argonne version†† (2D-SNARG) of a two-dimensional discrete S_n transport program.§

*Rago, A. L., and Hummel, H. H., An IBM-704 Program Treating Elastic Scattering Resonances in Fast Reactors, ANL-6805 (1964).

**Meneghetti, D., Evaluation of Group Cross Sections for Light Reflectors, Trans. Am. Nucl. Soc. 8, 247 (1965); also, Meneghetti, D., Effects of Leakage Upon Evaluation of Fast-reactor Multigroup Cross Sections for Reflectors, ANL-7170 (1966).

† Meneghetti, D., Rago, A. L., and Phillips, K. E., Use of the ELMOE-SNARG Programs to Study Details of Neutron Spectra Through Regions of Resonance-scattering Media, Resúmenes de los Artículos Presentados en la Conferencia Internacional sobre Utilización de Reactores de Investigación y Computo y Matemáticas de Reactores, Mexico City (1967).

†† Duffy, G. J., Greenspan, H., Sparck, S. D., Zaparka, J. V., and Butler, M. K., SNARG-1D, A One-Dimensional Discrete-Ordinate Transport-Theory Program for the CDC-3600, ANL-7221 (1966).

§ Carlson, B., Lee, C., and Worlton, J., The DSN and TDC Neutron Transport Codes, LAMS-2346 (1960).

TABLE II.B.1. Regional Atomic Densities,
 10^{24} atoms/cm³

Case	Core		Reflector	
1	Al ²³⁵ U	0.0603 0.004 ^a	Fe	0.0847
2	C ²³⁵ U	0.054 0.004 ^a	Ni	0.0913

^aOnly the absorption property of fission and not source property is used. These elements are not treated by the detailed Legendre-polynomial expansion coefficients.

diluent and another resonance scatterer (iron) as the reflector material. The second case has a nonresonance scatter (carbon) as the core diluent and a resonance scatterer (nickel) as the reflector material.

For example, consider the two interface examples described in Table II.B.1 which have identical slab-geometries. They simulate 60-cm-thick core regions, with 30-cm-thick abutting reflector regions. For simplicity and for reasonable machine-running time, only the case of an applied flat distributed fission source in the core region was evaluated. The first case has a resonance scatterer (aluminum) as the core

For these cases, S_2 flux solutions using linearly anisotropic and isotropic scattering assumptions were obtained. Transport approximations were employed for the isotropic cases. These solutions thus correspond to consistent-P1 and to diffusion-theory solutions since the applicable S_2 quadrature corresponded to that of a P1 or Gaussian quadrature. Indeed, if the only projected usage of an ELMOE and SNARG linkage were for S_2 solutions, a large saving in computation time could be obtained by employing a more direct (nondiscrete-ordinate) mode of solution.

Since, for these studies, detailed interest is primarily in the energy region of the scattering resonances, we will present only the fine-group solution in an energy range sufficiently wide to exhibit the characteristics of effects due to resonance-interface phenomena. For example, in these cases only the energy range from 0.5 MeV to 15 keV have been subdivided into 172 fine groups. The coarse-group cross sections and coarse energy intervals are those of the fast-group cross-section set of Yiftah *et al.**

The fine-group flux spectra at various positions on both sides of the interface between the aluminum diluent core and the iron reflector are shown in Fig. II.B.1 as functions of lethargy. For the cases to be described, the flux spectra from the isotropic solutions did not differ sufficiently from those of the anisotropic solutions to warrant reproducing a comparison of the flux spectra.

Furthermore, a higher-order, S_4 , flux solution for this case results in larger variations than the differences between anisotropic and isotropic S_2 . Because of troughs between resonances of one regional composition, the flux peak penetrates quite deeply into the adjacent region and vice versa.

Analogous spectra for the case with the carbon-diluent core having the nickel reflector are shown in Fig. II.B.2. Because carbon is not

* Yiftah, S., Okrent, D., and Moldauer, P. A., Fast Reactor Cross Sections (Pergamon Press, New York, 1960).

a resonance scatterer, its characteristic fine flux affects only the spectral envelope on the nickel side of the interface. The flux peaks in the carbon are due to the characteristic fine-flux peaks of the nickel reflector. The penetration of these flux peaks into the carbon core are less than those shown in Fig. II.B.1.

Fig. II.B.1. Neutron-flux Spectrum at Various Spatial Positions in the Aluminum-diluent Core and Iron Reflector

Fig. II.B.2. Neutron-flux Spectrum at Various Spatial Positions in the Carbon-diluent Core and Nickel Reflector

Work is currently continuing on the study of differing interfaces, some of which simulate more usual core-reflector compositions of fast-reactor systems.

(ii) Application of Second-order Perturbation Theory to the Collapsing of Multigroup Isotopic Cross Sections. Most of the computer codes currently in use for the production of multigroup cross sections collapse the macroscopic cross sections for the homogenized mixture under consideration on a fundamental-mode basis and, in addition, collapse the microscopic cross sections for each of the isotopes in the mixture. Although the procedure used in collapsing the macroscopic cross sections is fairly well established, the procedure used for the collapsing of the microscopic cross sections could probably be improved.

The main operations that one would desire to perform for a fundamental mode by using the cross sections produced by the collapsing operation are as follows:

- (1) To derive macroscopic cross sections for the standard reactor composition, yielding exact values for the material buckling and for the fundamental-mode flux.
- (2) To calculate exactly the reaction rates of the various isotopes under consideration for the standard core composition.
- (3) To calculate exactly the total material worths of the various isotopes for the standard core composition.
- (4) To calculate exactly the spectral material worths of the various isotopes for the standard core composition.
- (5) To derive macroscopic cross sections for core compositions close to the standard composition, yielding accurate* values of the corresponding material bucklings and fundamental-mode fluxes.
- (6) To calculate accurately* the reaction rates of the various isotopes for core compositions close to the standard composition.
- (7) To calculate accurately* the total material worths of the various isotopes for core compositions close to the standard composition.
- (8) To calculate accurately* the spectral material worths of the various isotopes for core compositions close to the standard composition.

Few of these requirements are met by the cross-section sets produced by the present multigroup codes. For example, the formalism used in collapsing the microscopic cross sections by the MC² code satisfies adequately only the first two requirements of the list. Of course, the other calculations may be performed by using the MC²-collapsed microscopic cross sections, but the error involved is often so large that a new MC² calculation is needed to satisfy requirements 5 and 6, even for compositions very close to the standard composition or to satisfy approximately requirement 3. Two additional MC² calculations would be needed to satisfy requirement 7, and there is no way, at present, to satisfy, adequately, requirements 4 and 8 with the MC² collapsing procedure.

The use of a collapsing formalism based on second- (or higher)-order perturbation theory promises to be very useful in producing

*By "accurate" we mean that the error should be at least of the second order with respect to the difference between the composition under consideration and the standard composition.

multigroup cross-section sets that would answer satisfactorily most or all of the listed requirements with only a modest expenditure of computer time.

The formalism is based on the use of the first and second derivatives of the real fluxes with respect to the atomic concentrations of each isotope, or of each possible combination of two isotopes. The flux derivatives are used to produce a cross-section set that contains considerably more cross sections than the standard sets, but can be used for any of the listed applications at the cost of only a minor additional effort on the part of the user. For example, in second-order perturbation theory, the cross sections answering requirement 5 will be obtained by

$$\Sigma = \Sigma^0 + \sum_{i=1}^M \sigma_i N_i + \sum_{i=1}^M \sum_{j=i}^M \sigma_{ij} N_i N_j,$$

where Σ is the desired macroscopic cross section, Σ^0 is a constant, N_i is the atomic concentration of the i th isotope, σ_i is the first-order perturbation cross section of the i th isotope, and σ_{ij} is the second-order perturbation cross section which represents the interference of the i th and j th isotopes. The errors of the material buckling and of the flux obtained with these cross sections would be of the third order with respect to differences between the atomic compositions in the actual and standard compositions.

A computer code is being prepared for the CDC-3600 to test this procedure in conjunction with the ultrafine cross sections used by the ELMOE code.

b. Reactivity Coefficients (H. H. Hummel)

Because of great concern about the uncertainties in the usual statistical treatment of the unresolved resonances used in fast-reactor Doppler-effect studies, detailed investigations, as an extension of the work by Kelber and Kier,* have been carried out. These investigations included both the resonance-integral method using the RANSRES code* and the direct Monte Carlo method using the AMC code.** The results of these investigations lead to the following conclusions:

(1) The large dispersion from the mean value in the Doppler effect of the fissile isotope (in terms of the fractional change in the fission rate, $\Delta R/R$) is almost entirely the result of the accidental overlap effect due to the presence of the resonances of the fertile isotope. The dispersion in $\Delta R/R$ for the fissile isotope was reduced by approximately one order of magnitude if the resonances of the fertile isotope were deliberately removed in the calculation.

* Kelber, C. N., and Kier, P. H., Nucl. Sci. Eng. 24, 389, 26, 67 (1966).

** Miller, L. B., Monte Carlo Analysis of Reactivity Coefficients in Fast Reactors; General Theory and Applications, ANL-7307 (March 1967).

(2) The dispersion in $\Delta R/R$ is sensitive to the number of levels under consideration. The dispersion in $\Delta R/R$ generally reduces when the number of resonances under consideration increases.

(3) For a given 200-eV interval, considered by Kelber and Kier (see p. 20), the dispersion of the Monte Carlo results is generally in agreement with that reported.

(4) The preliminary Monte Carlo results seem to indicate that the probable error in the fractional change in fission rate within a $7^{238}\text{U}:1^{239}\text{Pu}$ system obtained by direct calculation (bypassing the multi-group method) is only of the order of 40%, which is much smaller than that reported by Kelber and Kier (see p. 20). Further studies are still under way.

2. Fast Critical Experiments--Theoretical Support (F. W. Thalgott/ R. Nicholson and R. Palmer)

The work conducted under this activity consists of theoretical and analytical support in fast-reactor physics for critical experiments in the Idaho Division. The emphasis is on the planning, preanalysis, and post-analysis of experiments. The interaction between experimental work and theoretical support necessitates a close relationship between these efforts. Coordination is also maintained with efforts of the same general nature performed at the Argonne, Illinois Site.

Postanalysis of ZPR-3 Assemblies 48 and 49 continues. The effective cross sections in the three types of fuel and fertile plates for the complete cell were derived from three separate MC² runs, each being a simulated two-region cell for the plate in question. The RABZPR code is being used to determine the ^{238}U foil activity through the cell.

Planning and preanalysis of the first two cores of ZPPR have continued. Core 1 will be the simplest two-drawer cell that will have a critical volume of 1500-2000 liters and a composition similar to that of a typical power reactor. Core 2 will be a simple one-drawer cell with an initial volume of 3500-4000 liters. The heterogeneity effects of the normal drawers and the special oxide-plate drawers in Core 2 are being studied using the TESS double- S_n transport code. This code is now being modified to treat $\ell = 1$ anisotropic scattering.

3. Experimental Reactor Physics--Fast Critical Experiments-- Experimental Support (W. G. Davey)

a. Neutron Spectrometry. This task encompasses the development and utilization of techniques for measuring neutron spectra in the plutonium-fueled fast critical assemblies (ZPR-3 and ZPPR). Developmental problems

involving high gamma activity are particularly severe in plutonium systems containing high (10-20%) concentrations of ^{240}Pu .

Measurements have been made in ZPR-3 Assembly 50 (see ZPR-3 Operations and Analysis, Section II.B.4).

A neutron spectrometer system based on utilization of a small digital data-acquisition system as a multiparameter analyzer is being developed. The system will be tested after a modular real-time clock control system has been constructed. Preliminary checkout of subsystems has indicated excellent gamma discrimination capability and a high limit on data-acquisition rate.

b. Heterogeneity Studies. Large, dilute, fast-reactor systems now under consideration present increasingly more serious and complex problems associated with local flux and reactivity perturbations. Special precautions will be necessary to ensure that data measured in a critical assembly will be accurately interpretable in relation to microscopic theory and capable of extrapolation to power reactor configurations.

Potential techniques applicable to soft-spectrum systems in ZPR-3 and ZPPR have been evaluated by two recently recruited scientists. Initially, technique development utilizing activation analysis has been considered. In-cell energy-flux measurements should evolve using a variety of spectral indicator techniques. Preliminary emphasis is being given to establishing equipment specifications that are consistent with the demands of these potential techniques.

c. Computer Applications. Optimized experimental performance and high reactor utilization on ZPPR have been needed for some time. The physical size of systems to be investigated, coupled with the consequent dilution of fissile material, requires that all present reactivity measuring techniques, loading schemes, fissile material accounting, and traverse measurement devices be optimized from an operational and a data-acquisition-and-evaluation point of view. This task encompasses the solution to these problems.

Preliminary evaluation of the data collecting and handling requirements for ZPPR indicated that electronic digital data acquisition and reduction would be necessary to satisfy the accuracy and reactor utilization requirements of ZPPR.

The adaptability of proposed experimental techniques to on-line digital data collection and evaluation has been studied. Investigations into potential cost and manpower savings have resulted in discussions of compatibility between proposed ZPPR equipment and similar devices

projected for use with ZPR-6 and -9 in the Reactor Physics Division. These discussions were held as interdivision meetings on compatibility of equipment and experimental techniques.

A composite request for proposal is being formulated to determine if the appropriate industrial suppliers of such equipment can comply with the necessary compatibility requirements to achieve a substantial saving in cost and projected manpower utilization.

d. Integral Studies of Cross Sections. The objective of this activity is the study in integral experiments of basic cross sections used in fast power breeder reactor design.

A revised study of the best fission cross sections to be used in the analysis of fast integral experiments has been completed and submitted to Nuclear Science and Engineering for publication. This study indicates the areas of ambiguity in fission data and is used to direct the integral experimental effort.

4. ZPR-3 Operations and Analysis (W. G. Davey and R. L. McVean)

Last Reported: ANL-7382, pp. 55-58 (Sept 1967).

a. Basic Studies of Plutonium Systems. A neutron-spectrum measurement completed the experimental program on Assembly 50. The spectrometer used was a proton-recoil device that uses pulse-shape discrimination to differentiate between background gamma rays and neutrons.

Data were taken over the energy range of 1 keV to 2.5 MeV. Two different counters were used. One counter, filled with hydrogen, was used over the range of 1 to 150 keV. The other counter, filled with methane, was used from 100 keV to 2.5 MeV.

To reduce the gamma background for the spectrum measurement, fuel which had become gamma active from recent runs was replaced with relatively cold fuel from the vault. This replacement was made in the region surrounding the spectrometer. The reactor was then run at $4\frac{1}{2}\%$ subcritical to achieve reasonable counting rates for the spectrometer as well as spectral reproducibility.

After this measurement, the reactor was reloaded to Assembly 49 and the neutron-spectrum measurements repeated. It was not practicable to make this measurement when the other experiments were run on Assembly 49 because there was insufficient cold fuel in the vault to reduce the gamma background to an acceptable level. For this measurement, the reactor was run at 8% subcritical.

The data are now being analyzed, and the results will be reported at a later date.

b. Doppler Coefficient. Recent work on this task has involved soft-spectrum, plutonium-fueled assemblies. The obtaining of more data on the Doppler coefficient in these assemblies should greatly aid in the confirmation and testing of analytical methods for computing the magnitude of this effect in large fast breeder reactors. Confidence in establishing the safety criteria for fast reactors should be considerably enhanced by measurements of the Doppler coefficient in the ZPR-3 series of simple plutonium-fueled assemblies.

Current work involves the FFTF critical assemblies. Plans for these assemblies call for measurement of the Doppler coefficient of ^{238}U at locations in the reactor core other than in the central position used in previous ZPR-3 assemblies. To accommodate these requirements, the evacuated portion of the Doppler equipment has been greatly modified to fit within the confined space among the ZPR-3 control-rod drives. The Doppler manifold has been reduced in volume by an order of magnitude while retaining the electrical-connection and access ports and the alpha-particle detector. The connection from the manifold to the vacuum pump and valve assembly has been lengthened to place the latter items away from the control rods even at full insertion of the Doppler equipment. A new, higher-capacity ion pump is being fitted to the system and can be used interchangeably with the present ion pump.

c. Reactor Technique Development. ZPR-3 operations and utilization will be improved by the gradual upgrading of equipment and techniques. This improvement will involve reactor operation and techniques specifically, and not development of, for example, general counter and foil measuring techniques.

Broadly speaking, the objectives are improved "plant factor" and improved accuracy. The general trend of the development is the digitizing of output data so that on- or off-line computer data acquisition and reduction can be accomplished.

Current work involves improving the capability for making reactivity traverses. Reactivity traverses in previous ZPR-3 assemblies have been limited to one radial traverse position where samples could be changed manually without shutting the reactor down. This was achieved by providing a penetration through the shielding wall between the control room and the reactor. Any other radial and axial positions required a reactor shutdown to change samples. The length of time necessary to re-establish a reasonable temperature equilibrium after such a shutdown resulted in poor reactor and manpower economy compared to traverses where samples could be changed without a shutdown.

The future program for ZPR-3 calls for the eventual construction of a conical, split core. Nine radial traverse penetrations and several axial traverse penetrations will be needed.

To eliminate drilling eight more holes through the cell wall for radial traverses, and to eliminate the necessity for many shutdowns for axial traverses, a sample changer has been designed. The changer can be adapted to mechanically traversing a sample through the reactor, but this technique would require several undesirable corrections that would reduce the accuracy of the measurement. To overcome these difficulties, the sample will be transferred pneumatically in and out of the reactor within approved limits. Since only the sample would be moved or oscillated, the reactivity effect would be measured directly, drifts due to temperature effects being cancelled by multiple measurements at each in-reactor location of interest.

In its present design, the changer can handle up to 12 samples and/or reference capsules. Increasing this number would merely require enlarging the changer drum and housing.

d. Mockup Studies. Detailed mockup studies of specific reactor designs have constituted an important part of the ZPR-3 program. ZPR-3 critical studies in support of the Fast Flux Test Facility (FFTF), including both basic studies and more exact mockup designs, constitute a major part of the current and projected ZPR-3 effort.

The FFTF Phase B critical program on ZPR-3 was started on October 18 with the drilling of additional access holes in the ZPR-3 matrix. The entire matrix had been unloaded before this operation. These access holes are located in positions mutually agreed upon in detailed planning discussions between representatives of Idaho and Reactor Physics Divisions of ANL, and of Battelle Northwest Laboratory. In these discussions, detailed plans were also formulated for all experiments with Assembly 51, which is the first assembly in the Phase B program.

The additional access holes are essential because of the complexity of the FFTF core, a split conical system. They are not essential in Assembly 51, a simple, nonsplit cylinder, but the future FFTF criticals in the split-core and conical-core geometries will require worth and reaction rate traverses at off-core-center and off-core-axis positions. The dimensions of these cores are not well established at this time, and the traverse positions have been chosen to provide maximum flexibility to the experimental program over a wide range of geometries.

In the fixed half of ZPR-3, the holes are located $13\frac{1}{4}$ in. from the midplane in the P, K, and G rows, and at the midplane in the P, L, and H rows. In the movable half, the holes are located $12\frac{1}{2}$ in. from the midplane in the P, M, and J rows.

The detailed loading plans for Assembly 51 have been completed, and the necessary fuel and other materials are expected to be delivered in early November.

5. ZPR-6 and -9 Operations and Analysis

a. Integral Studies of Large Systems (W. Y. Kato)

Last Reported: ANL-7382, pp. 58-59 (Sept 1967).

This task covers the investigation and measurement of the physics parameters of large dilute uranium- and plutonium-fueled fast reactors.

Following the completion of the investigations of a large 2600-liter uranium carbide core, Assembly 5 on ZPR-6, the facility was completely unloaded of ^{235}U fuel. The annual fuel-inventory audit, maintenance checks, and cell-leakage rate measurements have been carried out. In addition, the argon gas storage tanks have been relocated from the west side of the building to the far southeast area away from the building. This was done to allow the contractor for the confinement shell construction free access to the cell exterior.

The ZPR-6 is currently being prepared for Assembly 6, a 3700-liter uranium oxide system.

The ZPR-9 was also completely unloaded to make the annual fuel audit in the early part of this reporting period. While the facility was unloaded, annual maintenance checks and cell-leakage checks were made.

The ZPR-9 is being loaded for a series of small, uranium oxide central zone core studies.

b. Doppler Effect (C. E. Till)

Last Reported: ANL-7382, pp. 58-59 (Sept 1967)

Exploratory measurements were done in ZPR-9 Assembly 18 using an all-graphite furnace designed for very high temperatures. The purpose of the measurements was to test the element behavior in-reactor and to test the adequacy of the existing vacuum and cooling systems for this element. The furnace contained a UO_2 sample and was run at a peak temperature of about 2000°K . The measurements indicate that some redesign of the element and cooling system is desirable, and these modifications are now being made.

c. Heterogeneity Effects (C. E. Till)

Last Reported: ANL-7382, p. 58 (Sept 1967).

One facet of the general heterogeneity effect problem presented to the fast-reactor critical experiments section is the effect on overall reactivity of the plate structures used in the ZPR's. Tests of the magnitude of these effects were done on Assembly 5 of ZPR-6. The reactivity worths of nominal 2-in.-cubic samples, of similar compositions but with differing internal geometry, were compared. The sample types were: first, the normal plate geometry; second, rodded geometry; and third, homogeneous slurries of the same materials. The sample compositions were similar, but not identical to the composition of Assembly 5 itself. The enrichment in each case was 12.7% of ^{235}U . The homogeneous samples were mixtures of UC powder and sodium or UC powder, sodium, and SS powder, the latter to mockup the tubes present in the rodded samples. Figures II.B.3 and II.B.4 show the geometry of the plate and rodded samples, respectively, and Table II.B.2 gives the compositions of each sample used.

Fig. II.B.3. Heterogeneous-sample Loading Pattern

Fig. II.B.4. Pellet-sample Loading Pattern

TABLE II.B.2. Samples Used in Heterogeneity-effects Study

Sample No.	Physical Description	Composition (g)				Stainless Steel	
		^{235}U	^{238}U	C	Na	In-sample	Can
1	Heterogeneous	82.95	635.78	25.00	0.00	49.51	23.74
2	Heterogeneous	82.95	635.78	25.00	48.00	49.65	23.74
3	Homogeneous	107.31	739.68	45.05	0.00	0.00	24.45
4	Homogeneous	108.96	751.01	45.74	34.36	0.00	43.37
5	Homogeneous	86.63	597.08	36.36	0.00	60.14	30.08
6	Homogeneous	87.86	605.59	36.88	44.44	61.01	29.08
7	Pellets	86.73	597.83	33.47	0.00	59.96	54.52
8	Pellets	87.38	602.30	33.71	42.29	59.90	54.56

The reactivity was measured in the following manner: The reactivity of one sample of each general composition was measured relative to void, using repetitive period measurements; the reactivity worths of the other samples with similar compositions were then measured relative to the first sample, using an oscillator in conjunction with a calibrated auto rod.

The small size of the samples used could cause the results to depend on the geometry of the environment in which the samples were placed. As a test of the sensitivity of the results to this, two environments were used: First, the normal reactor loading pattern, and second, a loading pattern in which the ^{238}U was bunched to increase its self-shielding (Fig. II.B.5); for both environments, the oscillator was the second drawer position. In the bunched-environment experiments, the bunched loading pattern was present in the eight drawers surrounding the sample, as well as 8 in. each way from the sample in the oscillator drawer itself.

Fig. II.B.5. Loading Pattern in Assembly 5 of ZPR-6 (End View)

The results are given in Table II.B.3 and are to be understood in the following way: Two general groups of samples are shown in the table, the first group without sodium, the second with sodium; to compare the results of samples within each group, differences in the compositions of the sample require that corrections, in most cases minor, be made to the measured results. The results shown in the table have been corrected using linear interpolations of the measured individual material worths. For the group without sodium, these corrections were made to bring each of the other samples to the same ratio of ^{235}U , ^{238}U , and carbon as

sample No. 5, and all the results were then normalized per kilogram of UC. For the group with sodium, an analogous procedure was used, except that the composition of sample No. 6 was used as the reference composition for the other two samples in the group, and the normalization is per kilogram of the uranium, carbon, stainless steel and sodium mixture. Because of the difference in normalization, meaningful comparisons should only be made within each group. The standard deviations listed in the table reflect only the reactor statistics, and do not, for example, reflect uncertainties in the corrections described above.

TABLE II.B.3. Measured Reactivity Worths at Center of Assembly 5, ZPR-6

Sample No.	Sample Type	Sample Worth Excluding Can	
		Normal Environment	Bunched Environment
		Worth of UC Only, $I_h/\text{kg of UC}$	
3	Homogeneous UC, no Na	3.027 ± 0.007	2.982 ± 0.007
5	Homogeneous UC plus SS, no Na	2.991 ± 0.011	2.932 ± 0.012
7	Rodded UC, SS clad, no Na	2.970 ± 0.009	-
1	Plate U, C and SS, no Na	3.436 ± 0.007	3.391 ± 0.009
		Worth of UC, SS, Na Mix, $I_h/\text{kg of Mix}$	
6	Homogeneous UC plus SS, with Na	2.234 ± 0.006	2.221 ± 0.008
8	Rodded UC, SS clad, with Na	2.227 ± 0.007	-
2	Plate U, C and SS, with Na	2.698 ± 0.010	2.644 ± 0.008

The results indicate, first, that the measurements are not sensitive to the change made in the sample environment in these measurements. Second, the homogeneous and rodDED samples had almost equal reactivity worths. Third, the reactivity worth of the plate-type heterogeneous sample without sodium, is about 13% greater than that of the equivalent homogeneous sample. Fourth, the reactivity worth of a plate-type heterogeneous sample, with sodium present, is about 20% greater than the equivalent homogeneous sample.

As an indication of the magnitude of these effects, a direct extrapolation of the reactivity difference between the plate and homogeneous samples, using a J_0^2 radial worth dependence and a $\cos^2$ axial dependence, would lead to an increase in the critical mass of Assembly 5 of 75 kg of ^{235}U for fuel distributed uniformly throughout the system.

6. ZPPR Operations and Analysis (H. Lawroski)

Last Reported: ANL-7382, pp. 59-62 (Sept 1967)

The contractor continued to have favorable weather for placing sand-gravel fill over the reactor cell and for completing the earth mound. Placement of the 26-by-26-mesh screen was completed, and 70% of the 18-in.-thick layer of 20-to-50-mesh sand has been installed; completion of this sand layer is awaiting the processing of 44 more yards of sand in Idaho Falls. The earth mound was completed to elevation 5164.0 on October 16, and hand finishing of the slopes has been completed preparatory to resuming installation of the asphaltic membrane. The ramp for the earth mound has been removed.

Six additional panels of flexboard siding were installed on the outside of the backup containment structure. Twenty-three of a total of 24 have now been installed. Seal welding was completed on 22 of a total of 24 of high-efficiency filter frames in the backup containment structure. The frame for the access door to the backup containment area remains at the fabricator for installation of the door.

Installation of ductwork, refrigeration piping, and electrical systems continued in the reactor cell. Erection of the frangible ceiling grid and structural steel for the personnel shield trolleys was started. The reactor cooling-system blowers were received and moved into the cell. Installation of the cell temperature control was started. The 1-ton monorail was erected in the blanket storage room. Painting continued in the mound area.

Installation of hardware and insulation was essentially completed in the support wing. Erection of the AFSR crane was completed. Modifications to reduce excessive leakage in the dampers above and below the filters in the suspect exhaust system in the fan loft were completed. Floor hardener was applied to the basement floors. Installation of asphalt tile was essentially completed. The fuel-coating machine was received and erected in the fuel-coating and -cleaning room. Installation of acoustical ceiling panels was completed in the control and conference rooms. Acceptance tests of the electrical systems in the support wing were about 80% complete at the end of the month. Overall construction of the facility is about 93% complete.

The due date for proposals for installation of the reactor and associated equipment was extended to October 11 from October 2. Seven proposals were received and are being evaluated.

The month's-end status of outstanding reactor components is as follows:

a. Rod-drive Mounting Plate: One of the two plates has been received. The rejected plate (No. 2) has been repoured and is now being machined by the fabricator.

b. Drawer Conveyor: The bid package has been assembled, but bids will not be requested until November 9, because of the current hold in funds.

c. Fission Counter and Sample Changer: Design work for the fission-chamber drive mechanism is continuing. Preliminary drawings of the sample changer to fit this drive have been completed.

7. ZPR Fuel and Nonfissile Materials--Technical Assistance

a. Fuel-element Fabrication Development (J. E. Ayer)

(i) Oxide Rod Elements. During fiscal year 1969, Reactor Physics studies will commence on simulations of oxide-fueled reactors of 2000-, 3500-, and 5000-liter sizes in ZPPR. The fuel elements required for these studies will be commercially procured to specifications generated by the Laboratory. The responsibility of the Plutonium-Materials Fabrication Group in this activity is to develop techniques for loading the fuel, and to write a feasibility report that will serve as a basis for product specification and will permit the procurement of acceptable fuel elements from a commercial source. The scope of the work includes design and specification of the fuel jacket, specification of the form and quality of the fuel, development of loading techniques, fuel-element testing, and generation of information leading to an overall product and process specification.

In the work so far, the following facts have been established and decisions made that merit consideration in further development. Minimum clearance between fuel and jacket, and between jacket and reactor collandria tubes, is of utmost importance. The clearance between fuel and jacket and between jacket and collandria was therefore set at no more than 0.003 in. and the jacket thickness at not more than 0.012 in. When the clearance between fuel and jacket is a minimum, loading techniques are needed that will permit contamination control with minimum effort. The desired clearance between jacket and collandria necessitates minimum protrusion of the end-plug girth-weld beyond the jacket wall. The oxide rods will be subjected to "hot zone" experiments that will require jackets resistant to thermal cycling between room temperature and 600°C in an air or nitrogen atmosphere. The choice of jacket material depends upon its compatibility with the fuel and its surrounding environment. A common reactor-grade tubing for containment of (U-Pu)O₂ fuel is Type 304 or Type 304L stainless steel. Work by W. E. Ruther and S. Greenberg, ANL, and E. A. Gulbransen and K. F. Andrew, Westinghouse, shows that at 600°C

the rate of weight change of Type 304 stainless steel in purified oxygen at 76 mm Hg pressure is linear with time between 2 and 6 hr. A weight-gain rate of about $0.2 \mu\text{g}/\text{cm}^2/\text{hr}$ was found for that condition. Such a weight gain calculated for a 0.375-in.-dia tube 6 in. long would be 3.6×10^{-6} g/hr. None of the oxide films or scales spalled away from the Type 304 stainless steel on cooling to room temperature. These findings support the selection of Type 304 or of Type 304L stainless steel for this application.

During this reporting period, sufficient Type 304L reactor-grade stainless steel tubing 0.385-in. OD by 0.012-in. wall has been received to satisfy the requirements of the oxide rod elements. The tubing was obtained commercially, fully annealed, and accompanied by a statement of the chemical composition. The final size was achieved by drawing the fully annealed tubing through a 0.375-in. die, resulting in a 0.367-in.-OD finished size. A 0.351-in.-dia pin was then pulled through the tubing to give a uniform size enabling 0.345-in.-OD pellets to be introduced. After the final sizing operation, the tubing was nondestructively tested for defects. One defect, consisting of a blemish on the outside diameter, was cut from the tubing. Metallography to determine the extent of cold work contributed by the sinking and redraw operations is now in progress.

Thirty tubes were cut to length. End plugs are now being made of Type 304 stainless steel to be TIG-welded into the jackets. The end-cap design has been proven with respect to our ability to make a girth weld with minimum protrusion beyond the outside diameter of the jacket. The shape of the end cap conforms to the requirements of the Reactor Physics and Metallurgy Divisions with respect to fixed length, minimum clearance between rod and collandria tube, a bearing surface apart from the weld, and a compromise between end-plug design and relatively insensitive welding parameters.

The development of techniques for the loading of plutonium-containing pellets into close-fitting jackets will be heavily influenced by work now in progress. Nineteen elements, consisting of 0.245- to 0.247-in.-dia (U-20% Pu)O₂ pellets, were loaded into jackets of 0.250-in. ID. The fabrication method and fixturing required for this work is described in the Progress Report for June 1967, ANL-7349 (pp. 124-125). A similar loading is now being made, but techniques are simplified to refine the process. Although the oxide rods for ZPPR have a smaller distance between the fuel and the rod top, other clearances are similar. Fixtures for preparing similar jackets for loading, and for reforming the jackets, have been built and are being used. Welding setups including chills to obtain welds of the type shown have been built for test welds on smaller components. Gauging to measure pellets and to ensure against oversize fuel pellets being introduced into assembly is now in operation on fuel loadings for Activity No. 46091-15, and will be done similarly for the ZPPR oxide rod

elements. The final design and fabrication of loading fixtures and jacket preparation and reforming tooling for this subtask is now under way.

Fuel for this subtask is being ordered through a development contract that is in preparation. The fuel-assembly development will proceed immediately upon review and approval of the preliminary fuel specifications by Reactor Physics Division and Metallurgy Division representatives.

(ii) Instrument Traverse Elements. The ZPPR series of Reactor Physics experiments will require about 250 instrument-traverse sensing elements to be made in FY 1969. These elements provide a means to achieve fine-flux mapping of the reactor core. The objectives of this subtask are to design the element, to develop the fabrication process, to estimate fuel losses, and to write a process specification based on experience gained by the fabrication of 50 elements. The cores for these elements will be selected from core plates submitted for inspection and sampling.

During this reporting period, an approved design of the traverse element has been achieved. The requirement of a somewhat precisely located hole through the traverse element core may be met in any of a number of ways. One of the most obvious is to saw a ZPPR element into pieces, clamp the raw edges together, and drill or bore a hole so that the saw cut is on the hole diameter. This method, however, is a waste of valuable material, places a premium upon cleanup and accountability, and requires multiple machine setup operations. The above objections should be avoidable by a simultaneous punching and shearing operation. A die to accomplish this operation has been designed and is being built. A loading fixture has been designed and built to permit the loading of halves of core plates into each end of a traverse-element jacket and to maintain an alpha-free exterior. Welding fixtures such as those used in ZPPR fuel element development will be used for end closure.

The traverse-element jackets are standard ZPPR hardware with a ferrule welded into the sides. The welding parameters have been established, and the welding operation has been fixtured and automated. The 50-element cans required for this subtask are now being welded by the techniques that have resulted in the work described.

(iii) Special Measurement Elements

Last Reported: ANL-7371, p. 67 (Aug 1967).

(a) Objective and Scope. The objective of this task is to fabricate special fuel bodies for experimental purposes. The scope of

work includes the fabrication of limited numbers of elements for Doppler- and danger-coefficient measurements, stacked-foil elements, and homogeneous drawer elements for reactor-physics studies.

(b) Danger-coefficient Elements. During this reporting period, 12 danger-coefficient elements (including eight dummy elements) were completed and delivered to Reactor Physics. The fueled elements contained 0.015- and 0.030-in.-thick foils of Pu-1.25 w/o Al alloy. The alloy was made in a vacuum induction furnace. The procedure involved placing electrical-grade aluminum wire on the bottom of an yttria-coated magnesia crucible and placing a broken plutonium button on top of the wire. The alloy is melted, heated to 950°C, and bottom-poured into an yttria-coated carbon book mold with a stepped cavity. This casting technique was described in the Progress Report for August 1967, ANL-7371, p. 67. After the casting cools, the furnace is opened, and the billet is removed and carefully cleaned. The foils required were formed by hot-rolling the 0.050-in. step casting to required thickness. Reductions were 15% per rolling pass between 5- by 8-in.-dia, two-high rolls that were electrically heated to 235°C until final dimensions were obtained. The foils were then wire brushed, weighed, rinsed in ethanol, dried, and assembled into jackets.

A new method of loading the plutonium and uranium isotopes into the stainless steel cans was developed. The aluminum funnels previously used for this purpose were costly to fabricate and were discarded because of contamination after loading each can. The new loading fixture shown in Fig. II.B.6 required no loading funnel and can accommodate 0.125- and 0.250-in.-thick element sizes. The element can is inserted from one end to a stop, and the core plate from the other. A 0.003-in. stainless steel shim enters the can on all four faces as shown on the insert in Fig. II.B.6. These shims prevent the core plate from contacting the lip of the element can. The core plate is forced in past the shims, which act as a retainer spring. After the core plate bottoms in the element can, the assembly is removed from the loading fixture and placed in a welding fixture where the end plugs were inserted.

(iv) Oxide Plate Elements. Cold-pressing and sintering, and hot-pressing procedures for fabricating the modular oxide core plates for ZPPR are being investigated. Pellets of (U,Pu)O₂ have been pressed to determine their sintering and shrinkage characteristics. These specimens were pressed from mixtures of UO₂ and PuO₂, and were sintered in vacuo, helium-6 v/o hydrogen, and helium at 1500 and 1650°C. The pellets were of 91 to 96% dense. The shrinkage range of the pellets was 16.3 to 19.5%. X-ray diffraction analysis and metallographic examination indicates that solid solution of the UO₂ and PuO₂ was essentially complete. The oxide/metal ratio of the sintered pellets was 1.98 to 1.99. The densities of the pellets from these initial studies are higher than the specified

density for the oxide fuel tile. Additional experiments are being performed on the effect of processing parameters on the final density of the mixed oxide. A 2.54-by-2.54-cm modular die has been ordered, and on receipt of this die, plates of the mixed oxide will be made.

Fig. II.B.6. Fuel-loading Fixture for Alpha-contamination Control

A series of experiments was performed in which multi-specimens of UO_2 were hot-pressed. The purpose of these experiments was to determine some parameters involved in this fabrication technique, and to develop a hot-press design that would be suitable for hot pressing $(\text{U,Pu})\text{O}_2$. Five and eleven plates were pressed at one time at 1600 and 1625°C. The thickness of the pressed plates varied between ± 0.01 cm from the mean dimension. Densities of 82.4 to 88.9% of theoretical were obtained on various pressings. Some carbon contamination occurred, and the pressed plates were heat-treated in a hydrogen atmosphere in an effort to remove or reduce the amount of carbon.

8. ZPR Fuel and Nonfissile Materials--Procurement

Last Reported: ANL-7382, pp. 61-62 (Sept 1967)

a. Technical Administration and Quality Control

(i) Plutonium Ingotting. The Atlantic Richfield Hanford Company blended and ingoted lot 12, from which they obtained 95 kg of plutonium to 11.5% ^{240}Pu . The blending lot 13, containing about 135 kg of plutonium, was

approved for ingoting. This lot contains near the specified maximum limit of 2.5% ^{241}Pu . It has become increasingly difficult to obtain plutonium of the isotopic composition necessary to produce ingots that meet the ZPR program specification ($11.5 \pm 0.5\% \text{ }^{240}\text{Pu} < 2.5\% \text{ }^{241}\text{Pu}$ and $< 0.1\% \text{ }^{238}\text{Pu}$). The average composition of 1330 kg of plutonium ingoted in FY 1967 was 0.035% ^{238}Pu , 86.75% ^{239}Pu , 11.53% ^{240}Pu , 1.54% ^{241}Pu and 0.14% ^{242}Pu . An increase in the ^{241}Pu results in increased gamma radioactivity from the daughter product ^{241}Am . Increased amounts of ^{238}Pu result in higher alpha and neutron levels and more self-heating of the fuel. Additional supplies of plutonium of suitable composition for ingoting are expected to become available in February 1968. Careful management will be required to meet the plutonium needs of the fabricators until the additional plutonium has been ingoted.

(ii) Fuel-element Fabrication at Dow. On October 31, 1967, the Dow Chemical Company reported the following progress:

Number of casting melts	239
Core plates machined	1835
Fuel elements assembled	1155
Fuel elements inspected and accepted by Dow	1016
Fuel elements shipped to ANL	759

The Dow-accepted and -shipped fuel elements were of the following sizes:

<u>Element Length (in.)</u>	<u>Inspected and Dow-accepted</u>	<u>Shipped</u>
4	146	15
5	583	497
6	18	14
7	253	218
8	16	15

The first shipment of fuel elements arrived in Idaho on October 30, 1967. Additional elements are scheduled to be shipped to Idaho during the second week of November 1967 to meet the requirement for 810 five-inch elements and 405 seven-inch elements.

Gamma radiation continues to be a problem in all areas where the ZPR plutonium is handled by Dow. Americium has been the principal source of this radiation. The plutonium ingots usually contain less than 1000 ppm of americium. At 1300°C, part of the americium evaporates from the melt and deposits on the molds, crucibles, and exposed furnace parts. Scrapings from the mold surfaces were analyzed to contain approximately 14% of americium. The casting skulls and furnace residues

were also strong radiation sources. Control of radiation exposure to personnel has required frequent cleanup of gloveboxes, minimized residue storage, local shielding within the gloveboxes, shielding glass on windows, leather gloves, and personnel rotation.

Argonne National Laboratory received 25 core plates and 25 fuel elements on October 3 for preproduction evaluation of the NUMEC processes. NUMEC inspection reports were received on the core plates and elements on October 10, analytical samples on October 18, and the NUMEC analytical results on October 20. The ZPR fuel-element specification requires that inspection reports and analyses be shipped with the fuel elements, and that analytical samples from castings be shipped within 10 days of the time the castings are made. The physical evaluation of the fuel elements has been completed, and chemical analyses are still in progress. A revised quality-control plan and inspection outline was received on October 26. Manufacturing instructions were received for binary-alloy preparation on September 29, and welding manufacturing instructions were received on November 2. NUMEC was authorized by ANL to prepare and cast the binary alloy on October 20. Preliminary approval to weld bottom plugs in 300 jackets was given on October 20. As of October 31, detailed NUMEC manufacturing instructions had not been received on ternary alloying and casting, machining of core plates, and fuel-element assembly.

9. FFTF Critical-experiment Program (P. J. Persiani)

The purpose of this study is to plan and execute a series of critical experiments in support of the FFTF project. An objective is to perform critical experiments from which could be developed the computational technique and models to be used in the design study of the FFTF reactor.

The planning of experiments designed to investigate problem areas that may arise in the FFTF design studies is necessarily a continuous effort since it depends on the evolution of the design. The second series of critical experiments (Phase B) are planned to investigate physics design problems associated with the current FFTF split-core inverted conical design. The engineering mockup experiments that will probably be planned for the later phase of the critical program would be in part directed to establish the fuel-composition specifications, the major effort being directed toward finalizing important design parameters. This series of experiments would include studies related to the experimental program planned for the FFTF.

An integral phase of this activity is to develop a theoretical effort to be used not only in defining a proper sequence of design-related critical experiments but also in analyzing the final experimental results. These analytical models would thus serve as a base for extrapolating to the actual FFTF design.

a. Analysis

(i) Cross-section Set Development. The composition of Phase B, Assembly I, of the FFTF Critical Experiments has been determined from the best available component composition and inventory information. However, not all nuclides in Assembly I are available in library tapes which can be used with the MC² code to generate a cross-section set. The composition was, therefore, modified by representing aluminum as sodium and by replacing Mn, Si, and Mo by Fe, all on an atom-for-atom basis. The resulting composition, shown in Table II.B.4 was used in MC² to generate a new cross-section set, designated as 29001. Nuclides of interest on the tape, but not present in the composition, were added with a concentration of 10^{-9} atom/b-cm, core-averaged cross sections were generated using a heterogeneous fundamental-mode approach and reflector-averaged cross sections with zero buckling in the homogeneous composition of Radial Reflector I. No separate cross sections were produced for Radial Reflector II and the axial reflector. The cross sections for the 29th group, averaged over all energies below 0.532 eV were produced by the THERMOS code. For Doppler calculations, core-averaged cross sections at 1000°K were also produced. The 29001 set is not yet in the proper form to be used in diffusion and transport calculations.

TABLE II.B.4. Composition of FFTF Critical Assembly I (ZPR-3 Assembly 51)
atom/b-cm

Material	Core	Axial Reflector	Radial Reflector I	Radial Reflector II
²³⁹ Pu	0.001731		10 ⁻⁹	
²⁴⁰ Pu	0.0001706		10 ⁻⁹	
²³⁵ U	0.0000150		10 ⁻⁹	
²³⁸ U	0.007018		10 ⁻⁹	
C	0.003113		10 ⁻⁹	
O	0.01279		10 ⁻⁹	
Fe	0.0162594	0.01076	0.007925	0.07547
Cr	0.003657	0.00259	0.001870	0.00115
Ni	0.001600	0.02893	0.05640	0.00049
Na	0.0043113	0.01037	0.00416	
²⁴¹ Pu	10 ⁻⁹		10 ⁻⁹	
¹⁰ B	10 ⁻⁹		10 ⁻⁹	

(ii) Drawer-loading Diagrams. Figure II.B.7 shows the core drawer loading on which the heterogeneous MC² and THERMOS computations were based. This loading was chosen to space the pairs of plutonium- and uranium-bearing plates as uniformly as possible. Also, the carbon, oxygen, and sodium are spread evenly, within limits. The stainless steel in the cans is not negotiable. The remaining stainless steel in the 1/8-in. pieces and in the side walls of the matrix tubes and drawers is positioned next to the fuel as the cladding would be and is rather evenly spaced. The position of the iron in the Fe₂O₃ was sacrificed somewhat for the sake of fuel spacing.

Figure II.B.8 gives the drawer loadings for the reflectors.

Na
Na ₂ CO ₃
U ₃ O ₈
ZPPR FUEL
SS
Na
Na ₂ CO ₃
Na ₂ CO ₃
DEP-U
Pu

Na
Na ₂ CO ₃
DEP-U
SEFOR FUEL
SS
Na ₂ CO ₃
Na ₂ CO ₃
Fe ₂ O ₃
U ₃ O ₈
ZPPR FUEL

Fig. II.B.7. Core Two-drawer Cell Loadings,
ZPR-3 Assembly 51

Na
Na
Na
Na
Na
Na
Na
Na

AXIAL
REFLECTOR

Na
Na
Na
Na
Na
Na
Na
Na

RADIAL
REFLECTOR I

Fe

RADIAL
REFLECTOR II

Fig. II.B.8. Reflector Drawer Loadings,
ZPR-3 Assembly 51

(iii) Preloading Computations for Assembly I. Using the previous cross-section Set 28001, which is operational, with MACH-1 one-dimensional diffusion code, the compositions given in Table II.B.4, and the dimensions shown in Fig. II.B.9, the critical-core volume without heterogeneity correction was 310 liters and the critical-core radius was 33.8 cm. The worth distribution along the core axis computed by the Mach-1 perturbation option of ²³⁹Pu and ²³⁸U is given in Table II.B.5. The Doppler coefficient will be calculated using cross-section Set 29001.

b. Experimental Work

(i) Preparation for FFTF Phase B Critical Experiments. The FFTF Phase B critical program on ZPR-3 was started in October with the drilling of additional access holes in the ZPR-3 matrix. The entire matrix was unloaded before this operation.

Fig. II.B.9. FFTF Critical Assembly I,
ZPR-3 Assembly 51

TABLE II.B.5. FFTF Assembly I Axial Worth

Distance from Midplane (in.)	Reactivity Coefficient ($\Delta k/k$)	
	^{238}U	^{239}Pu
0.5	-39.0	796
1.5	-38.7	792
2.5	-38.1	781
3.5	-37.0	763
4.5	-35.5	739
5.5	-33.6	708
6.5	-31.5	672
7.5	-29.0	630
8.5	-26.2	585
9.5	-23.3	536
10.5	-20.0	484
11.5	-17.2	431
12.5	-14.2	378
13.5	-11.2	325
14.5	-8.86	279
15.5	-6.54	231
16.5	-4.55	183

This first Phase B experiment is designated as ZPR-3 Assembly 51.

These access holes are located in positions mutually agreed upon in detailed planning discussions with representatives of Idaho and Reactor Physics Divisions of ANL, and of Battelle Northwest Laboratory. In these discussions, detailed plans for all Assembly I experiments were formulated. The additional access holes are essential because of the complexity of the FFTF core, a split conical system. They are not essential in Assembly I, a simple nonsplit cylinder, but future FFTF criticals in the split-core and conical-core geometries will require worth and reaction rate traverses at off-core-center and off-core-axis positions. The dimensions of these cores are not well established, and the traverse positions have been chosen with the view of providing the maximum flexibility to the experimental program over a wide range of geometries.

These traverse holes are located in the fixed half $13\frac{1}{4}$ in. from the midplane in the P, K, and G rows, and at the midplane in the P, L, and H rows, and in the movable half $12\frac{1}{2}$ in. from the midplane in the P, M, and J rows.

The detailed loading plans have been completed, and the necessary fuel and other materials are to be delivered in early November.

(ii) Modifications to Improve Flexibility of Equipment for Measurement of Doppler Coefficient. Plans for the FFTF critical assemblies call for the measurement of the Doppler coefficient of ^{238}U at locations in the reactor core other than in the central position used in previous ZPR-3 assemblies. To accommodate these requirements, the evacuated portion of the Doppler equipment has been grossly modified to fit within the confined space among the ZPR-3 control-rod drive units. As a result, the Doppler manifold has been reduced in volume by an order of magnitude, still retaining the electrical connection and access ports and the alpha-particle detector. In addition, the connection from the manifold to the vacuum pump-vacuum valve assembly has been lengthened to place these items away from the control rods even at full insertion of the Doppler equipment. A new, higher-capacity ion pump is being fitted to the system and can be used interchangeably with the present ion pump.

(iii) Improvement of Reactivity-traverse Measurements. Reactivity traverses in previous ZPR-3 assemblies have been limited to one radial traverse position, where traverse samples could be changed manually without shutting the reactor down. This was achieved by providing a penetration through the shielding wall between the control room and the reactor. The other available radial and axial positions required a reactor shutdown to change samples. The length of time necessary to re-establish a reasonable temperature equilibrium after such a shutdown resulted in poor reactor and manpower economy relative to traverses where samples could be changed without a shutdown.

The future program for ZPR-3 calls for the eventual construction of the FFTF split conical core. Nine radial-traverse penetrations and several axial-traverse penetrations will be needed.

To eliminate drilling eight more holes through the cell wall for radial traverses, and to eliminate the necessity for many shutdowns for axial traverses, a sample changer has been designed. The changer can be adapted to moving a sample through the reactor, on the end of a rod or other mechanical device, but this technique would require several undesirable corrections that would reduce the accuracy of the measurement. To overcome these difficulties, it is planned to pneumatically transfer the sample into and out of the reactor within limits established by the operating manual. Since only the sample would be moved or oscillated, the reactivity effect would be measured directly with drifts due to temperature effects cancelled by multiple measurements at each in-reactor location of interest.

Up to twelve samples and/or reference capsules can be handled by the changer in its present design. Increasing this number would merely require enlarging the changer drum and housing.

C. Component Development--LMFBR

1. Sodium Technology Development--Engineering Development

a. Sodium Quality Measurement (S. B. Skladzien)

Last Reported: ANL-7382, pp. 63, 64 (Sept 1967).

(i) Laboratory Techniques. An analytical technique for concentrating and separating certain specific minor impurities from sodium samples has been investigated. The technique involves using lanthanum as a carrier added to the sample solution. By the addition of ammonia gas, the pH is adjusted to 9, producing a hydroxide precipitate that carries the impurities. The separation produces two improvements in the detection and measurement of the impurity concentrations by atomic absorption: (a) it eliminates the high sodium-salt content of the sample that frequently leads to clogging of the fuel passages in the burner, and (b) it permits concentrating the impurities of interest at a level well above their minimum detection limit.

This technique was used to analyze a sample, identified as Lot V, of the sodium to be used in the Core Component Test Loop; the analyses of the sodium from the storage tank yielded values of 4.2 ppm Cr, 1.3 ppm Cu, 27 ppm Fe, 0.8 ppm Mn, and <0.2 ppm In. Analyses of the Lot V sample for oxygen by the mercury-amalgamation method gave 29.6 and 33.6 ppm; the vacuum-distillation method indicated oxygen contents of 34.4 and 39.6 ppm. Bulk-sodium temperature at the time of sampling was 400°F. Earlier, samples of the storage-tank sodium sampled from three different elevations were analyzed for total carbon with the following results:

<u>Sample</u>	<u>Carbon (ppm)</u>
II-38-C	17, 13
II-66-B	9, 11
II-66-T	13, 10
I-102-B	9, 10, 26
I-102-T	11, 7, 9, 20

(ii) In-line Techniques. In the Sodium Analytical Loop, the electrodes have been installed in the United Nuclear oxygen-meter sensor housings, and the resistivity meter was adjusted.

The oxygen-meter electrodes are closely matched; their outputs varied between 0 and 4 mV of each other during a 15-day period with the system at $600 \pm 1.5^\circ\text{F}$ and no known change in oxide concentration. During this period, the resistivity-meter indications varied. Monitoring will continue for indications of improvement or possible clues as to the cause of the variations.

Work is continuing on the correlation of the in-line instruments and vacuum-distillation sampling. So far, control and instrument variations can only be traced to stray electrical currents or malfunctions.

b. Sodium Quality Control (S. B. Skladzien)

Last Reported: ANL-7382, p. 63 (Sept 1967).

Construction of the prototype submerged oxide collector is ~50% complete. A small test arrangement to determine large Na_2O concentrations in cold-trap mesh material has been completed. The method will react steam with the sodium and Na_2O mixtures and monitor the evolution of hydrogen. Initial purging with hydrogen and system checkout will begin soon.

2. Reactor Mechanisms Development--Materials Evaluation

a. Physicochemical Mechanics of Metals (F. A. Smith)

Last Reported: ANL-7382, pp. 64, 65 (Sept 1967).

As reported previously, after Type-304L stainless-steel EBR-II fuel tubes were exposed to sodium for 1000 hr, tests with the laser microprobe showed no significant penetration of sodium into the grain boundaries. Since then, to determine whether residual sodium in microcracks affects the response of stainless steel to Strauss solution, sodium-exposed tubes were cleaned by two alternate methods and then were subjected to standard Strauss reagent for 2 hr. Four tubes were tested. The first two were filled with sodium and held for 1229 hr at 1200°F. Then the first tube was cleaned with ethanol; the second tube was cleaned by induction-heating the tube in vacuo to distill the sodium from the interior. Subsequent tests with the laser microphotospectrometer showed no detectable sodium on the interior surface. The third tube was exposed to sodium for 813 hr at 1200°F, then cleaned with ethanol; the laser test showed no sodium on the inside surface. The fourth tube, which served as a control sample, was held at 1200°F for 24 hr but was not exposed to sodium.

The results of the exposure of these tubes to the Strauss solution are still considered to be very preliminary. However, both tubes that had contained 1200°F sodium for 1229 hr showed resistivity increases: 23% for the ethanol-cleaned tube, and 19% for the vacuum-distilled tube. The 813-hr ethanol-cleaned sample showed an increase of only 3% (the reason is not clear). By contrast, the control sample disintegrated, indicating that great sensitization must have occurred during the 24 hr at 1200°F. Other control samples will be exposed for periods to 1000 hr; an additional group of samples is being prepared for comparative measurements.

One micrograph made of the 1229-hr vacuum-cleaned sample showed that the Strauss solution attacked the outer surface of the tube (which was not exposed to sodium) more than the inner surface.

b. Fission-gas Pressure Transducer (J. R. Folkrod)

Last Reported: ANL-7382, pp. 65, 66 (Sept 1967):

(i) NaK-filled Capillary System. An entirely new 1/8-in.-dia. NaK-filled system now being assembled was cleaned with a weak acid, acetone, and alcohol. The system will be evacuated while being heated above 600°F. Pure NaK for fill has been purchased and high-grade bellows valves will be used in the vacuum system and fill lines.

The design and assembly procedures will be reported in detail. Equations have been derived giving the relationship of pressure, void, and fluid-expansion effects on the system.

(ii) Strain-guage System. No progress has been made on this system. The vendor is still making corrections to the equipment.

(iii) Null-point System. A proposal from Atomics International to furnish two null-point pressure transducers has been received and is being evaluated.

c. In-core Flowmeter (G. F. Popper)

Last Reported: ANL-7382, pp. 66-70 (Sept 1967).

(i) Thermal and Mechanical Tests of Magnetic Materials. Determination of the reversible temperature effects of stabilized Alnico magnet samples has begun. Preliminary data have been taken.

Fabrication of the circuitry for the B-H tester has been completed; the circuitry is ready for checkout.

(ii) Prototype Magnetic Flowmeter. Data pertaining to the stability of the preliminary prototype flowmeter have been assessed. The first calibration was made at 400°F after the flowmeter had operated 90 hr between 400 and 500°F and 34 hr between 500 and 600°F. Sensitivity at 400°F was 0.01708 mV/gpm.

After an unsuccessful attempt to operate the recirculation portion of the calibration facility at 700°F continuously so as to wet the flowmeter, a calibration was run at this temperature using the storage-tank sodium, which was heated to 700°F. Sensitivity was 0.01561 mV/gpm. Before this calibration, the flowmeter had been run for only 13 hr between 700 and 800°F.

Additional calibrations were run at 600, 500, and 400°F. The sensitivity at 400°F was 0.01788 mV/gpm, or 5.1% more than at the first calibration. This indicates complete wetting had not been achieved for the first calibration. Between these 400°F calibrations, the flowmeter had operated for 192 hr between 400 and 800°F: 26 hr at 400-500°F, 26 hr at 500-600°F, 123 hr at 600-700°F, and 17 hr at 700-800°F. The calibration facility was then shut down for modifications to increase the heating capability of the recirculation loop.

After additional heaters were installed, the loop was heated and a check calibration made at 400°F. Surprisingly, the sensitivity measured about 30% less than previously. Investigation revealed that large welding currents could have passed directly through the flowmeter assembly when the new heaters were being attached to the loop piping. Therefore, we assume that the welding current demagnetized the flowmeter magnets and that this reduced the sensitivity. When this series of tests is concluded, the flowmeter will be removed from the loop and the gap flux will be measured to verify this assumption.

The sensitivity at 400°F was normalized to that determined previously (0.01788 mV/gpm). Then a calibration was performed at 700°F. Based on the normalization factor, the 700°F sensitivity increased by 1.3%, or to 0.01582 mV/gpm. This increase was probably due to the incomplete wetting during the first calibration. Only an additional 22 hr of operation between 700 and 800°F were accumulated between the 700°F calibrations, however.

TABLE II.C.1. Temperature History of PM Flowmeter between Calibrations of June and August 1967

Temperature (°F)	Time (hr)
300-400	146.0
400-500	137.0
500-600	11.0
600-700	226.0
700-800	197.0
800-900	18.0
Total	735.0

Then the flowmeter was operated for an additional 735 hr at temperatures as indicated in Table II.C.1. After this, check calibrations were made at 400 and 700°F. No further changes in sensitivity were detected. During this period of operation, the flowmeter was subjected to thermal shocks as large as 38°F/sec, with no deleterious effects, either mechanical or electrical. The loop was shut down once for repairs during this period. Particular attention was given to the way welding operations were performed to ensure that little or no welding current could pass through the flowmeter assembly.

Since the last calibrations were performed, an additional 720 hr of operation have been accumulated on the flowmeter. The total temperature history of the flowmeter is shown in Table II.C.2. The flowmeter

TABLE II.C.2. Temperature History of PM Flowmeter as of the End of September 1967

Temperature (°F)	Time (hr)
300-400	231.5
400-500	410.5
500-600	220.0
600-700	444.5
700-800	362.5
800-900	60.5
900-1000	57.0
1000-1100	29.0
Total 1817.5	

has operated at temperatures up to 1100°F during this 1817-hr period. Check calibrations will be made as soon as the loop is operational again (see below).

The expansion bellows in the flowmeter leg of the calibration loop failed early in October; check calibrations will be run on the flowmeter as soon as the loop is repaired temporarily. Then the loop will be shut down for permanent repairs; the expansion bellows will be replaced by hairpin expansion joints, and the dc EM pump will be replaced with an ac model.

(iii) Magnet Irradiation Tests.

Work on the experimental description and hazard analysis is continuing, but progress is being delayed by the lack of accurate data on gamma heating rates in the EBR-II reactor.

Figure II.C.1 shows the mockup of the hot cell to be used for continuous measurements of the magnetic flux of irradiated magnets at any temperature up to 1200°F. In the cell, the magnetic-rod samples are

Fig. II.C.1. Mockup of Hot Cell for Measuring Magnet Properties after Irradiation

stored in individual sample holders in the storage racks. Only one sample is transferred to the furnace assembly at a time. This is done by using a manipulator to remove the holder and sample from the storage rack, dump the sample into the sample transfer tray, place the holder on the catch table, slide the transfer tray into a mating position with the furnace assembly (a ceramic push rod inserts the sample into the furnace), move the sample transfer tray back to the position shown in Fig. II.C.1, rotate the sample thermocouple, and drop it into intimate contact with the magnet.

To make a flux measurement, the furnace assembly is rotated 180° by means of a turntable. An indexing pin provides accurate positioning.

After the magnetic flux has been measured, the manipulator lifts the thermocouple assembly off the sample, mates the transfer tray again with the furnace, pushes the sample back into the transfer tray, picks the sample holder off the table, slides the transfer tray away from the furnace assembly, drops the sample back into the sample holder, and places the sample holder into the only storage position open (to maintain sample identification).

Final design of the experimental Alnico magnet irradiation capsule is being delayed by the lack of accurate data on gamma heating rates in the EBR-II reactor. Such data are needed to ensure that, during irradiation, the magnet temperatures are as close as possible to 1200°F yet never exceed 1300°F . Until recently, the only values available for gamma heating rates were calculated on the basis of neutron flux. But some preliminary experimental values differ considerably. For example, at 45-MW reactor power, the gamma heating rate in a steel sample in Row 7 (12 in. from the reactor radial centerline) was calculated to be 2.37 W/g , but the experimental value was 0.83 W/g . Such differences in heating rates would give considerably different maximum magnet temperatures. To resolve the uncertainty, a mockup irradiation capsule with steel samples and melting-point indicators may need to be inserted to determine actual temperatures in the desired reactor position. This possibility is being investigated.

(iv) Eddy-current Flowmeter. The evaluation analysis predicts that the sensitivity of an eddy-current flowmeter (which has coils external to the flow conduit) first increases as the applied carrier frequency is increased from a low value, reaches a maximum value, and then finally decreases again as the applied frequency is further increased. The analysis also shows that errors due to changing fluid temperature can be minimized if the instrument is operated at a frequency in the region of peak sensitivity.

A changing velocity profile in the fluid, due to varying Reynold's number, introduces a degree of nonlinearity to the signal. If the

laminar-flow condition can be avoided by the proper selection of flow conduit size, the instrument response is given by $E = KQ^{1.028}$, where E is the signal voltage at the secondary-coil terminals, K is the flowmeter sensitivity, and Q is the flow rate in gallons per minute.

3. Fuel Handling, Vessels, and Internals

a. Core Component Test Loop (CCTL) (R. A. Jaross)

Last Reported: ANL-7382, pp. 71, 72. (Sept 1967).

The tentative arrangements for testing the prototype FFTF flow tube (fuel subassembly) were discussed with representatives of the FFTF Project. Plans are to install a full-size fuel and bottom reflector section of the FFTF flow tube in the test vessel of the CCTL for hydraulic testing. This assembly has a total length of 104 in. and will be tilted about 5° to simulate FFTF conditions. Only one assembly will be tested at its design flow rate of 400 gpm. Test temperature will be 1060°F ; depleted UO_2 will be used in the fuel pins.

Several modifications are still in progress on the CCTL. These include installation of throttle valve and a materials-compatibility test facility. Completion of the modifications and delivery of the prototype flow tube are scheduled for January 1968.

D. Systems and Plant--LMFBR

1. 1000-MWe Plant

a. Contract Management and Liaison (L. W. Fromm)

Last Reported: ANL-7382, pp. 87, 88 (Sept 1967).

An information meeting on the design of large sodium-heated steam generators was held at The Babcock & Wilcox (B&W) Co. Boiler Division in Barberton, Ohio, on October 3. At this meeting, B&W and Atomics International personnel presented results of their AEC-sponsored steam generator development work for the benefit of all 1000-MWe follow-on contractors.

Separate meetings were held between representatives of the Liquid Metal Engineering Center (LMEC) and General Electric personnel on October 19, 1967, and Babcock & Wilcox personnel on October 27, 1967, to discuss the relationships between the LMEC Combined Unit Reactor Vessel (CURV) study and the 1000-MWe follow-on studies.

b. The Babcock & Wilcox Company Subcontract. Company effort during the month was primarily on physics methods, refinements of the reference design, and trade-off studies.

The physics-methods work on the fast-reactor design codes is nearly complete. New cross-section libraries are being constructed, chiefly from ENDF/B data.

The architect-engineer subcontract with Sargent and Lundy has been approved by ANL; work is underway.

The reference core has been selected. Task II engineering of the heat-generation and heat-transfer systems is continuing.

The study of 1980 reactor safety criteria has been completed; the results are being used in developing the reference design.

The pot versus loop and the refueling trade-off studies have been combined into one study. Work has begun on this and other trade studies.

c. Westinghouse Corporation Subcontract. The Laboratory reviewed Technical Note 132-2, Evaluation and Trade-off Study of the Number and Arrangement of Loops for a 1000-MWe LMFBR Plant, and sent the results to the contractor.

A final iteration of the reference core design has been carried out to account for changes in fuel-temperature limitations and fuel swelling rate. Fuel venting, fuel, and materials have been evaluated.

Heat-transfer equipment, pumps, and energy-conversion systems information has been developed; plant-systems concept selection studies are underway.

Task II work is being planned. Plans for nuclear calculations including those for control requirements and fuel cycle have been completed.

d. General Electric Company Subcontract. The Laboratory reviewed General Electric Technical Note No. 1, A Summary Status Report of the APO Reactor Physics Developments Since 1964, and sent the results to the contractor.

Three analytical-type safety studies were initiated, concerning fuel meltdown, specific accidents, and effects of blast upon systems. Attractive core designs have been screened by using previously completed thermal-hydraulic and fuel-cycle analyses. Based on this screening, a preliminary selection has been made for advanced core designs.

Mechanical and plant arrangement studies, including alternates in fuel handling, maintenance methods, and numbers of loops, were begun. Data allowing preparation of basic flow sheets and heat balances were developed; rudimentary plant instrumentation and control diagrams were prepared.

e. Combustion Engineering, Inc., Subcontract. The first draft of the Conceptual System Design Descriptions for the five major areas being studied under the contract was completed. The draft of the Task I report is almost complete.

f. Atomics International Subcontract. Preliminary work plans for Tasks II, III, and IV have been completed and are being reviewed. Work plans for Task I have been revised to include parametric investigation of several major core variables to avoid major changes in the reference reactor design in future work. Work on the base conceptual system description is in progress. System design descriptions are being prepared, steam-cycle conditions have been established, and a cost-effectiveness model for the 1000-MWe plant has been developed.

E. EBR-II

1. Research and Development

a. EBR-II System Design Descriptions (E. Hutter)

Last Reported: ANL-7382, p. 12 (Sept 1967).

Collection and organization of source data and tests of retrieval are continuing. The data are being evaluated for depth and scope, using as guidelines parameters established in the working outline. Writing will start after comments on this outline are received.

b. Reactor Experimental Support--Reactor Analysis and Testing (R. R. Smith)

Last Reported: ANL-7382, pp. 12-19 (Sept 1967).

(i) Operational Data Analysis. The results of work performed under this subtask are published quarterly in tabular and graphical form.

(ii) Nuclear Analysis

(a) Neutron Streaming in the Trifluted Reflector Section. SNARG calculations in the R-Z geometry and the S_4 scattering mode were reviewed to estimate the relative importance of neutron streaming with the stainless-steel trifluted reflector as compared with the straight 19-pin stainless steel reflector:

The geometry of the trifluted reflector is such that there are three lines of right semicylindrical channels through each fluted section with diameters of approximately $1/2$ in. Previous calculations have been made with the reflector considered as a homogeneous region of stainless steel and sodium to determine its effectiveness as a reflector and shield. The most recent calculations were intended to estimate, in a non-homogeneous case, the possible effect of excessive neutron streaming through the $1/2$ -in. channel. Three problems were run: (a) the homogeneous case for reference, (b) a $1/2$ -in. streaming channel in the upper reflector, and (c) a $3/8$ -in. streaming channel. The results of the calculations show no significant streaming. These results are not unreasonable in view of the small solid angle involved where even small-angle scattering in the sodium will cause the neutrons to be scattered out of the channel.

(b) Comparative Reactivity Worths in Run 26B. The fuel loading, after the installation of the oscillator rod, provided sufficient reactivity to continue Run 26B until the scheduled 1100 MWd shutdown.

Fuel-transfer operations at the start of Run 26B provided an opportunity to measure the following comparative reactivity worths:

Oscillator Assembly vs Control Rod No. 8	-300 lh
Partial Subassembly (1/2 worth) vs Standard Driver Subassembly in Position 6A1	-80 lh
Partial Subassembly (1/2 worth) vs Standard Driver Subassembly in Position 4B3	-200 lh

As a result of these loading changes, the critical rod-bank position was 11 in. for nine rods, with one control rod at 8 in., and with the oscillator rod and stainless steel rods inserted in the core and occupying position Nos. 8 and 1, respectively. The power-reactivity decrements at 45 MWt, as measured on two successive startups, were 41 and 43 lh, respectively.

(c) Experimental Determination of Effect of Core Size on Power Coefficient. Vital to a definitive understanding of the Run 25 reduced power coefficient is a knowledge of the effects of core size and composition. As a logical supplement to computational efforts, a comparison of experimentally established values of the power-reactivity decrement (PRD) for a variety of core sizes and compositions has been made. Table II.E.1 presents such data from previous power-coefficient measurements.

TABLE II.E.1. Initial Power-reactivity Decrement for Previous Runs

Run No.	Rod Bank Position (in.)	Measured Reactivity Decrement (lh)	Decrement Minus Total Rods Expansion Effect (lh)	Kg ²³⁵ U	Number of Core Subassemblies	Control Rods Calibrated?
2				190.9	73	No
3	11.25	76.1	62.6	190.6	73	No
4	11.8	76.2		189.2	72	Yes
5	12.2	75.6	65.5	188.7	72	No
6				192.2	76	Yes
7	12.5	77		186.2	75	No
8	12.5	75	65.9	186.2	74	Yes
9	12.5	69		190.4	74	Yes
10	11.8	67		190.5	78	No
11	11.5	78		193.9	79	No
12	12.5	72.2		196.8	79	No
13	11.25	70.9	57.4	194.3	78	No
14	11.3	70.4	57.1	198.2	80	Yes
15	13.0	68.9	61.5	198.2	80	Yes
16	11.2	77.6	63.9	192.4	77	Yes
17	11.0	76.5	62.5	188.5	75	Yes
18	11.2	76.1	62.4	191.6	77	Yes
19				189.6	76	Yes
20	11.2	71.0	57.3	184.5	74	No
21	11.2	71.0	57.3	196.7	80	Yes
22	12.0	72.4	61.5	195.5	80	Yes
23	12.0	66.4	55.5	193.3	78	Yes
24	12.0	65.5	54.6	195.5	79	Yes
25	11.0	39.2	25.2	200.6	81	Yes
				206.0	88	Yes

The first column in Table II.E.1 identifies the run number. The second column gives the nominal position at which the control rods were banked. (Usually 9 or 10 rods were banked at the position indicated; other rods, depending on their functions, were located at other positions.) The third column lists values for the measured PRD corrected for burnup during the measurements and for changes in bulk coolant temperature. The fourth column gives PRD values, with control-rod expansion effects subtracted. In a sense, these values are those for hypothetical EBR-II cores that have no control rods. Since corrections for the effects of control-rod growth rely on accurate control-rod calibration data, only those values are given for which reliable calibration data exist. The fifth column gives the mass of ^{235}U for each loading. The sixth column lists the number of subassemblies considered as the core loading, including all partially and fully loaded fuel subassemblies, all 14 control and safety rods, and all irradiation experiments within the inner six rows. The last column indicates the availability of sufficiently reliable control-rod calibration data.

Table II.E.1 permits several interesting observations to be made. First of all, PRD values in the fourth column indicate that no significant trend as a function of time is evident, but as indicated by the number of subassemblies, an inverse correlation may be seen between the PRD and the size of the core. Such behavior is illustrated graphically in

Fig. II.E.1. Reactivity Decrement at Start of Various Runs (Total rod-bank expansion has been subtracted)

Fig. II.E.1. With the exception of Run 3, the data from runs before the radial stainless steel reflector (Run 25) was added indicate that smaller PRD values are measured for the larger cores.

Complicating the interpretation of data given in Table II.E.1 and illustrated in Fig. II.E.1 are the following facts:

(1) Experimental subassemblies containing less fuel than normal driver subassemblies were placed in the reactor at various times throughout Runs 1-25.

(2) The substitution of stainless steel for uranium in the upper and

lower reflectors was begun in Run 13, most of the substitutions occurring in Runs 18-20. Before Run 18, a maximum of seven subassemblies contained stainless steel, but after Run 20 a minimum of 46 subassemblies were with stainless steel axial reflectors. All the remaining uranium axial reflectors were replaced with stainless steel for Run 25.

(3) The major changeover from Mark-I to Mark-IA fuel took place during Runs 15-19. Before Run 15, the maximum number of Mark-IA subassemblies was five, but after Run 19, a minimum of 53 subassemblies contained Mark-IA fuel. The change from Mark I to Mark IA decreased core height from 14.22 to 13.50 in. and an increased enrichment from 48.5 to 52.4%.

After considering the data in Fig. II.E.1, we have assumed that the trend toward lower PRD values with increasing core size is real and that the influence of changes in core height, fuel enrichment, and axial-reflector material is not noticeably significant. Attempts to correlate decreases in PRD values with time, however, have been unsuccessful.

If the earlier data are linearly extrapolated for the Run 25 configuration, only a portion of the decrease in PRD noted in Run 25 can be attributed to the effects of increases in core size. The remainder, approximately 10-12 lh, is believed to be associated with the substitution of uranium radial blanket in Rows 7 and 8 for stainless steel and largely due to the inverse bowing of reflector subassemblies in Row 8.

(d) Transfer-function Results. To establish the magnitudes and time dependencies of Run 25 power-coefficient components, a series of transfer-function measurements was conducted on the Run 26B configuration. Data coverage was limited by a tendency for the oscillator rod to "rub" at lower frequencies under high power conditions, but, valuable information was obtained. The results, thus far, are summarized in feedback form in Figs. II.E.2-4, where the feedback function is defined as

$$H(i\omega) = \frac{1}{G(i\omega)} - \frac{1}{G_0(i\omega)} \quad (1)$$

In this expression, $1/G(i\omega)$ and $1/G_0(i\omega)$ are the reciprocals of the measured and zero-power transfer functions, respectively.

In all cases, the amplitude and phase of the measured transfer function were normalized to corresponding zero-power data at a frequency of about 8.8 Hz.* This procedure is premised on the assumption that the feedback at 8.8 Hz is zero. Since some feedback from fuel-expansion effects at 8.8 Hz may exist, corrections for the loss will be made when the time constant and amplitude for fuel feedback are estimated.

Although the results illustrated in Figs. II.E.2-4 are still preliminary and subject to additional corrections, the following tentative conclusions may be drawn:

* Hz $\equiv$ Hertz unit $\equiv$ cycles/sec.

(1) The feedback guarantees operational stability for all credible reactivity insertions over the frequency range studied and for the power range 0-41.5 MWt, with no indication of instability for credible power increases above 45 MW.

(2) The "shape" of the low-frequency feedback, particularly that at 30 MW for which slightly lower frequency information was obtained, suggests the existence of a "loop" in the feedback function. The loop, in turn, may be explained in terms of positive-feedback effects, resulting (presumably) from inward bowing attitudes, or from strongly delayed negative-feedback effects, resulting from the expansion of control-rod shafts.

If the data are verified by repeating the experiments and various corrections applied, an attempt will be made to separate the feedback function into individual time-dependent components, each of which may be identified with specific physical feedback processes.

Fig. II.E.2. Feedback, H, 22.5 MW, 100% Flow, Run 26B, Oct. 23, 1967

Fig. II.E.3. Feedback, H, 30.0 MW, 100% Flow, Run 26B, Oct. 17, 1967

Fig. II.E.4. Feedback, H, 41.5 MW, 100% Flow, Run 26B, Oct. 18, 1967

(e) Analysis of EBR-II Rod-drop Test Results. To determine the time dependencies and amplitudes of prompt or relatively prompt feedback components, a substantial number of rod-drop tests were conducted in both Runs 25 and 26A. In brief, the method consists of removing reactivity rapidly by dropping a special stainless steel rod from the core. The resulting power decrease is monitored with a magnetic tape recorder, digitalized, and used as input for a variety of digital programs.

The first step in the analysis consisted of calibrating the stainless steel drop rod. By solving the inverse kinetics equations for a drop at zero power (actually 500 kW), a program is used to compute the reactivity worth of the rod as a function of position. To obtain the results illustrated in Fig. II.E.5, the rod worth-position curve actually used was based on an average of the results of three different calibrations conducted at zero power.

Fig. II.E.5. Results of EBR-II Rod-drop Experiment at 45 MW

Since noise causes an error of $\pm 0.15\%$ in the calculated worth of the rod from a single drop (total worth 4.0%), there is an advantage in averaging to reduce its effects. The fact that feedback recovery amounts to only 0.5% at the end of the first second following drop and only 1.0% at the end of 10 sec indicates more clearly the influence of noise. The possibility of reducing error through the use of smoothed data curves, which may reduce the error to as little as $\pm 0.01\%$, is being considered.

After the rod-reactivity-worth-position calibration is established, the inverse kinetics equations for the 45-MW rod-drop data are solved and the feedback recovery determined by subtracting the worth of the drop rod from the gross reactivity.

In Fig. II.E.5, the results for the feedback recovery represent the averaging of three different sets of data taken at 45 MW. The power trace, on the other hand, is given for one drop only. In general, the feedback is characterized by a relatively smooth dependence on time; however, at about 3.8 sec, a slight dip may be noted. Although the reason for the dip is not evident, the fact that a power variation of only 0.1% could account for its existence must be emphasized. A dip of this magnitude could also be caused by a slight change in the temperature of the inlet coolant.

If negative feedback is considered to be shown along the positive abscissa in Fig. II.E.5, the feedback appears to be positive for a few tenths of a second after the rod drop. The feedback for the first few tenths of a second was negative, however, in a similar set of computations carried out for a rod bank of 13.00 in. The short time behavior for the data illustrated in Fig. II.E.5 may be explained as the result of a small, prompt, positive-feedback component or as the result of drifts in power or coolant inlet temperature.

Attempts to fit the feedback recovery curve to one or more simple power coefficient terms are continuing.

(iii) Thermal and Hydraulic Analysis

(a) Heat-transfer Calculations for Blanket Subassemblies.

Calculations to determine the temperature distribution in the 10th row (depleted uranium blanket) subassemblies were completed, using the HECTIC-II code. These calculations are complementary to those for the 7th, 8th, and 9th rows reported last month. Although the calculations indicate that a relatively flat radial temperature distribution across the 7th-10th rows is obtainable through reorificing (reducing) the flow in the 7th and 8th row stainless steel reflector subassemblies, a question has arisen concerning the validity of the calculations. The heat-transfer calculations were based on a calculated radial gamma energy distribution. Preliminary results from an ORNL experiment carried out in July at the end of Run 25 indicate that the calculated gamma energy production in the 7th row may have been high by a factor of about three. The discrepancy is being investigated, and a more rigorous calculation of the gamma energy distribution will be carried out at ORNL based on the Run 25 core and blanket.

(iv) Safety

(a) Postirradiation Inspection of Fuel Subassemblies

Discharged from Run 25. Still regarded as a possible explanation for the decrease in power coefficient noted in Run 25 is a mechanism based on

the inward compression of core subassemblies from a reverse bowing action in the stainless steel reflector. To ascertain whether the postulated bowing causes permanent deformation of core subassemblies, postirradiation examinations are being conducted on all fuel and control-rod subassemblies discharged at the end of Run 25. Three types of measurements are being performed: (a) straightness, (b) twist, and (c) dimensions across the spacer buttons.

In the straightness inspection the subassembly is supported at the lower end of the bottom adapter and clamped at the upper and lower ends of the hexagonal tubing. Six dial indicators are positioned along the length of the hexagon at the following distances from the top of the hexagonal tubing. Six dial indicators are positioned along the length of the hexagon at the following distances from the top of the hexagonal tube: 6, 19.25, 32.5, 45.88, 59.13, and 72.5 in. Indicator readings are taken, after which clamp pressure is released and the subassembly rotated about its central axis. Indicator readings are taken for each flat so any significant deviation from straightness would be indicated by respective high and low readings across a set of flats.

The twist of the hexagonal tube is also established through dial indicator measurements. Two readings are taken at the bottom, and two at the top, of a given flat. Differences in the readings may be interpreted directly in terms of angular twist.

The dimensions across the spacer buttons are measured with a snap gauge equipped with a dial indicator readout. After the snap gauge is calibrated with a 2.318-in. standard, the actual dimensions across the buttons on three opposite sets of flats are established.

Although the straightness test is initially performed on a completed (loaded) subassembly, preirradiation button and twist measurements are made on the empty hexagonal tubes. All postirradiation measurements are taken on complete subassemblies.

Small deviations from specifications were noted for button dimension measurements. The results for the eight subassemblies examined thus far are summarized in Table II.E.2.

Specifications for the dimensions across spacer buttons are defined as 2.317 ± 0.001 in., and reproducibility of measurements has been shown to be ± 0.001 in. The results in Table II.E.2 indicate that small changes in button dimensions may have occurred on some subassemblies.

The distance across flats on the hexagonal cans of subassemblies C-273 and C-274 was measured at intervals of 1, 3, 6, 9, and 12 in. above and below the spacer buttons. The dimensions were

essentially all within or above the original dimensions, indicating that no permanent compression of the hexagonal cans occurred, even though the button dimensions may have been reduced.

TABLE II.E.2. Summary of Spacer-button Dimension Measurements

Subassembly	Grid Location	Flat Position	Preirradiation Results	Postirradiation Results (in.)
C-271	5B2	1-4	In spec.	2.319
		2-5	In spec.	2.315
		3-6	In spec.	2.317
C-272	4D1	1-4	In spec.	2.317
		2-5	In spec.	2.316
		3-6	In spec.	2.322
C-273	5C2	1-4	One set 2.316,	2.316
		2-5	others in spec.	2.309
		3-6		2.311
C-274	5C4	1-4	One set 2.316,	2.312
		2-5	others in spec.	2.324
		3-6		2.310
C-275	5E4	1-4	One set 2.314,	2.311
		2-5	others in spec.	2.321
		3-6		2.320
C-277	5B4	1-4	One set 2.315,	2.312
		2-5	others in spec.	2.316
		3-6		2.315
C-257	3B2	1-4	In spec.	2.320
		2-5	In spec.	2.319
		3-6	In spec.	2.317
B-344	6A2	1-4	One set 2.323,	2.316
		2-5	others in spec.	2.321
		3-6		2.318

An attempt has been made to correlate the extent of subassembly compression or expansion with the orientation of the subassembly with respect to core center. However, no entirely consistent correlation has been found.

No special significance can be attached to the results cited in Table II.E.2. Changes in the button dimensions may have occurred, but, since comparable postirradiation tests were not conducted on previously discharged fuel, such changes cannot be attributed to features unique to Run 25. Internal stress relief effects from the immersion in $\approx 770^\circ\text{F}$ sodium may be responsible for the dimensional changes.

c. Nuclear Analysis Methods Development (P. J. Persiani)

Last Reported: ANL-7382, pp. 19-24 (Sept 1967).

(i) EBR-II Physics Analysis. Various subassembly worths in Run 25D were studied, using the SNARG-2D XY twofold symmetric configuration presented in Figs. II.E.6 and II.E.7. Figure II.E.6 presents the hexagonal array, with individual assembly location identifiers for the EBR-II core, with the loading of Run 25D included. Figure II.E.7 presents the equivalent representation in terms of rectangles, as used in SNARG-2D. Table II.E.3 compares the results and available experimental data. The calculated values of worth for replacement in the core or fueled control subassemblies by their follower regions are all approximately 10% high; the calculated value for the replacement of the dummy steel subassembly in 6B1 by driver fuel is about 15% low. All Run 25D SNARG-2D calculations have been performed with the simplifying assumptions that all driver-fuel subassemblies have the same composition and that all experimental plus half-loaded subassemblies also have a single composition. Neither of these assumptions is made in the current series of Run 26 calculations, and with the availability of an improved six-group cross-section set, even better agreement with experiment is expected for Run 26.

The effect of replacing the stainless steel reflector with the depleted-uranium blanket subassemblies in Rows 7 and 8 for the Run 25D configuration was calculated, and the resulting fission-density distributions, normalized to 45 MW, are tabulated in Fig. II.E.7. Here, each square represents a subassembly; the analytic mesh size is one-quarter or one-half of a single subassembly, as shown on the diagram. Only typical core positions are shown. Of particular interest is the effect resulting from the insertion of the uranium blanket in Rows 7 and 8. The fission densities in the experimental subassemblies for the depleted-uranium blanket case are from 2 to 5% lower than in the loading having stainless steel subassemblies in Rows 7 and 8.

This replacement of the reflector produced a k_{eff} of 0.994, i.e., $\Delta k = -260$ lh. The further replacement of driver fuel for half-loaded fuel in Assemblies 4A3 and 4B3 (and also in the mirror-image positions below the centerline; see Fig. II.E.6) resulted in $k_{eff} = 1.012$. The replacement of the full-loaded for half-loaded driver fuel subassemblies averages $\Delta k = 0.0045$ (+195 lh) per subassembly. The system resulting from replacement of reflector by blanket in Rows 7 and 8 of Run 25D may be brought to criticality by the additional replacement of one or two half-loaded driver subassemblies by full driver subassemblies.

(ii) New Multigroup for Cross-section Sets for EBR-II Analysis. A new 22-group set (Set 238) and a new 6-group set (Set 23806) have been developed recently for analysis of EBR-II, which serve as replacements for the previously used cross-section sets, Sets 236 and 23606. (See Progress Report for January 1967, ANL-7302, pp. 6-10.) A new MC² program, which

Fig. II.E.6. Segment of EBR-II Experimental Loading Run 25D

TABLE II.E.3. EBR-II Run 25D Subassembly Worths

$$430 \text{ Ih} = 1\% \Delta k/k$$

Replacement	Calculated Δk (Ih)	Experimental Δk (Ih)
Foll. for 5A1(FC)	-149	-130.0
Foll. for 5A3(FC)	-174	-162.5
Foll. for 5B1(FC)	-155	-142.5
Foll. for 5B3(FC)	-182	-160.0
D for 6B1(SB)	+109	-
MR for 6D1(SB)	-16	-
D for 6B1(MR)	+125	+150 $\pm$ 5
Na for 2D1(X)	-1016	-
Na for 2A1(D)	-1234	-
X for 6A3(D)	-93	-
D for 6B5(X)	+93	-

Subassembly Types:

D	Driver fuel
X	Experimental
FC	Fueled control
SB	Steel blanket (reflector), (19 v/o Na, 81 v/o SS)
MR	Modified reflector (dummy), (33.3 v/o Na, 66.7 v/o SS)
Na	100 v/o Na
Foll.	FC follower region

employs an improved method of treating inelastic scattering, was used for Sets 238 and 23806. Set 238 contains nuclides weighted with spectra characteristic of the core, reflector, and blanket of EBR-II, whereas Set 236 contained only the core and blanket weighted nuclides. The same MC² library tape was used in these calculations as was used in the development of Set 236. It is planned to develop new cross sections based on a library tape containing the ENDF-B data when this becomes fully available. The 22-group cross sections from the MC² program were converted into MACH and XLIBIT formats and the data written on magnetic tape. The 22-group MACH cross sections were then collapsed to 6-group cross sections, using the same energy structure that was used in collapsing Set 236 to Set 23606. The neutron spectrum used in collapsing these cross sections was that generated by the MACH program for a spherical reactor having a core, reflector, and blanket similar in compositions to those found in EBR-II. The 6-group cross-section sets were written in MACH and XLIBIT formats and stored on magnetic tape.

(a) Comparison of Sets 238 and 23806 with Sets 236 and 23606. A series of criticality calculations using diffusion and various orders of transport approximations was used to compare the new cross sections

with those previously developed. Table II.E.4 presents the series of calculations, along with the values of k_{eff} . The calculations were all performed on the spherical representation of EBR-II used in collapsing Set 238 to Set 23806. The values of k_{eff} are normalized; therefore the S_8 calculation using Set 238 has a k_{eff} of unity. This case was selected for normalization because it was considered to be the most accurate of the reactivity calculations in Table II.E.4. The influence of the neutronic model (diffusion or various orders of transport theory) on the value of the reactivity is about the same for all cases. Comparison of Cases 1 and 2, and Cases 5 and 6 shows that the use of reflector cross sections in the reflector region results in a ~1% lower value of k_{eff} . The effect of the improved treatment of the inelastic scattering on k_{eff} is seen in the difference in k_{eff} between Cases 2 and 3 and between Cases 6 and 7. Comparison of Cases 1 and 5, and Cases 2 and 6 shows the excellent agreement between the 22-group results and the 6-group results for Sets 238 and 23806. However, the spectrum used to collapse the 22 groups to 6 groups is identical to the spectrum of the system whose reactivities are calculated in Table II.E.4. An examination of Cases 5 and 7 shows that for an S_2 calculation the use of a 23806 set should yield a value of reactivity about 1.2% lower than that calculated with the 23606 set. These results are encouraging because the calculated reactivity of EBR-II with Set 23606 has been greater than the measured value.

TABLE II.E.4. Value of k_{eff} of Spherical Representation of EBR-II for Various Cross-section Sets, Cross Sections in The Reflector Region, and Neutronic Models

Cross-section Set	Weighting Spectrum for Cross Sections in Reflector Region	Diffusion	S_2	S_4	S_8
238	Reflector	0.9747	1.0231	1.0019	1.0000
238	Blanket	0.9848	1.0331	1.0122	1.0102
236	Blanket	0.9858	1.0276	1.0069	1.0050
236	Core	0.9855	1.0274	1.0066	1.0047
23806	Reflector	0.9758	1.0250	1.0035	1.0016
23806	Blanket	0.9866	1.0351	1.0138	1.0119
23606	Blanket	0.9894	1.0374	1.0164	1.0144
23606	Core	0.9886	1.0367	1.0155	1.0135

(b) Analysis of Cross Sections in the Reflector Region.

A perturbation analysis was conducted to determine the cause of the lower value of the reactivity calculated with the new cross-section sets. The reactivity effects produced by exchanging blanket and reflector spectrum weighted cross sections in the reflector region were calculated for the individual and total components of $\Delta k/k$. The results of the calculations, given in Table II.E.5, reveal that the leakage component makes the largest contribution

to the difference in the reactivity. The exchange of the reflector and blanket spectrum weighted cross sections of iron accounts for 89% of the total difference. The absolute values for the total $\Delta k/k$ (9.41×10^{-3} and 11.26×10^{-3}) for the two substitutions do not agree. This disagreement is due to the different spectrum generated by the cross sections, the difference not being taken into account by a first-order perturbation analysis. The findings of the perturbation analysis led to a very detailed study of the method employed in the MC² code for calculating the transport cross section. The final conclusion of the study is that the composition used to generate the weighting spectrum may, in certain cases, greatly affect the calculated value of the transport cross section.

TABLE II.E.5. Component and Total Values of $\Delta k/k$ for the Exchange of Reflector Weighted Spectrum Cross Sections in the Reflector Region

Nuclide	Capture	Radial Leakage	Elastic Scattering	Inelastic Scattering	Total $\Delta k/k$
Substituting Blanket for Reflector Weighted Cross Sections in Case 1 MACH Calculation					
Fe	-3.189×10^{-4}	8.374×10^{-3}	1.439×10^{-4}	-1.612×10^{-6}	8.197×10^{-3}
Ni	-2.609×10^{-5}	-4.203×10^{-6}	-5.094×10^{-5}	-8.70×10^{-7}	-8.210×10^{-5}
Cr	5.478×10^{-6}	1.191×10^{-3}	3.143×10^{-5}	-2.77×10^{-6}	1.226×10^{-3}
Na	3.251×10^{-6}	5.851×10^{-5}	7.997×10^{-6}	$+2.59 \times 10^{-7}$	7.002×10^{-5}
Total for Mixture	-3.363×10^{-4}	9.619×10^{-3}	1.324×10^{-4}	-4.993×10^{-6}	9.411×10^{-3}
Substituting Blanket for Reflector Weighted Cross Sections in Case 2 MACH Calculation					
Fe	2.242×10^{-4}	-1.002×10^{-2}	-2.359×10^{-4}	$+6.7 \times 10^{-7}$	-1.003×10^{-2}
Ni	2.841×10^{-5}	-1.113×10^{-5}	$+2.981 \times 10^{-5}$	7.865×10^{-7}	4.787×10^{-5}
Cr	-6.694×10^{-6}	-1.178×10^{-3}	-3.320×10^{-5}	2.191×10^{-6}	-1.215×10^{-3}
Na	-3.081×10^{-6}	-5.056×10^{-5}	-1.064×10^{-5}	-3.32×10^{-7}	-6.461×10^{-5}
Total for Mixture	2.428×10^{-4}	-1.126×10^{-2}	-2.499×10^{-4}	-3.316×10^{-6}	-1.1262×10^{-2}

Preliminary calculations using Sets 238 and 23806 have been encouraging. The value of the reactivity of EBR-II, Run 25, at 45 MW had previously been calculated to be 1.0574 with Set 23606. Changing only the cross-section set from 23606 to 23806 produced a value of 1.038. Reactivity calculations of Run 26A have just started. Using Set 23806 and new data on the composition of EBR-II, the SNARG (Sn = 2) Code gives a k_{eff} of 1.0294. Subtracting $0.025 \Delta k/k$ to correct for the Sn = 2 approximation (see Table II.E.4) and $0.0047 \Delta k/k$ (or 200 lh) to correct for the initial excess reactivity of Run 26 yields 0.9997. Examination of the input and output data is continuing.

d. Reactor System Testing, Surveillance, and Evaluation (B. C. Cerutti)

Last Reported: ANL-7382, pp. 24-25 (Sept 1967).

(i) Repair of Mark-II Oscillator Rod Drive. Special tools for the installation of the oscillator-rod drive were designed and fabricated, and

detailed instructions for the entire replacement procedure were written. The oscillator driveshaft bearing carrier tube was also reassembled.

The major steps in the oscillator-rod installation are as follows:

- (1) The control-rod drive was removed from position No. 8.
- (2) The modified single-pin oscillator rod was placed in the empty control-rod position using a special manual gripper tool.
- (3) The oscillator driveshaft was installed using the control-rod-drive pulling pipe to purge the gripper bellows area with argon.
- (4) The driveshaft was lowered onto the oscillator rod and the gripper jaws were closed.
- (5) The assembled bearing carrier was lowered in place over the driveshaft and threaded onto a small rotating plug sleeve.
- (6) The upper drive assembly was assembled and realigned.

Operational checkout of the oscillator in the "fuel-handling" mode and "reactor operation or rotation" mode was completed satisfactorily. The running torque applied to the shaft at zero reactor power was very low (approx 10 in.-lb). The drive system was tested from zero to 9 Hz (540 rpm) with no indications of noise or harmonic vibration such as experienced with the previous oscillator rod.

The reactor was then brought up to power, and the oscillator-rod experiments were started. The rubbing problem encountered with the previous oscillator rod was still evident above 25 MWt reactor power at low speeds. Rubbing becomes more severe with increasing reactor coolant inlet and outlet differential temperatures and at decreasing rotational speeds. This problem is being investigated.

e. Higher Power Operation (R. E. Rice)

Last Reported: ANL-7382, p. 25 (Sept 1967).

Engineering work on the reorificing of fuel subassemblies in Rows 4, 5, and 6 continues. Flow calibrations were made for a prototype sixth-row driver subassembly with the trifluted axial blanket. For a standard Row 6 driver with six 0.259-in. inlet holes, the flow rate of 800°F sodium was 66.5 gpm at 41 psi. For a configuration with six 0.314-in. inlet holes, the flow rate is 84 gpm at 41 psi, which is the flow rate for the prototype subassembly B382 now installed in reactor position 6C4. Subassembly

outlet temperature data have been taken at 45 MW with full flow and at 22.5 MW with flow rates from 100 to 50% of full flow for Run 26. These data, with similar data from a standard driver in Run 25, are shown in Table II.E.6 to compare the characteristics of subassembly B382 with a standard Row 6 driver subassembly. As a result of this test, the required inlet hole size for a 78-gpm Row 6 driver flow will be specified and flow-tested.

TABLE II.E.6. Sodium Outlet Temperatures, Core Position 6C4

Power (MW)	Flow (%)	Flow Ratio: B382 to Standard Driver	Outlet Temp (°F)		Temp Rise Ratio: Standard Driver to B382	Temp Rise Ratio: 22 MW, 100% Flow, to 22 MW, 55% Flow	
			Standard Driver	B382		Standard Driver	B382
45	100	1.26	872	854	1.12		
22.5	100	1.26	800	776	1.32		
22.5	55	1.26	860	840	1.14	0.625	0.544

f. Fuel Swelling and Driver Surveillance (F. G. Foote)

(i) Mark IA--Anomalous Fuel Swelling Investigation

Last Reported: ANL-7382, pp. 26-29 (Sept 1967).

The nature of the problem was described in the Progress Report for March 1967, ANL-7317, pp. 8-12. The total problem has been split into 11 subproblems and will be reported in this format.

(a) Extended Burnup of MC-S Type Fuel Pins. Burnup is being accumulated during Reactor Run No. 26.

(b) Capsulated Irradiations of SL Type Fuel in EBR-II. Burnup is being accumulated.

(c) Effect of the Change to Mark-IA Design upon Fuel Swelling. Burnup is being accumulated.

(d) Effect of Enrichment Increase from 48% (Mark I) to 52% (Mark IA). Burnup is being accumulated during Reactor Run No. 26. Subassembly C-283 in reactor grid position 1-A-1 will reach target burnup about midway through Run No. 26; assuming steady operation of the reactor, this will be during the first week of November 1967. C-283 will be the first of the special subassemblies to reach the target burnup. Fuel surveillance should start late in November or early in December 1967.

(e) Effect of Position and Orientation in the Casting Bundle upon Fuel Swelling. Burnup is being accumulated.

(f) Effect of Trace Element Impurities upon Fuel Swelling

(1) Subassembly C-2077. Burnup is being accumulated in reactor grid position 3-B-2.

(2) FCF-produced Melts. Melt identifications and trace-element compositions are given in Table II.E.7. The alloying elements (with the exception of zirconium) and the isotopic compositions are all within the standard specifications for EBR-II fuel and are not included in the table. With the exception of Melt No. I, all the ingots were remelted, in some cases more than once, to achieve the specified compositions. There appears to have been no great difficulty in introducing iron and aluminum to the melts, but the introduction of the required amounts of silicon appears to be much more difficult. This is unfortunate since silicon appears to be particularly effective in reducing fuel-pin swelling and in view of a recent recommendation that all EBR-II fuel contain a minimum of 250 weight ppm silicon. The problem of how to best introduce controlled amounts of silicon is under intensive study by Idaho personnel, with assistance from Illinois. In any case, it is now doubtful that these melts adequately fulfill the requirement of this experiment.

TABLE II.E.7. Melt Identifications and Trace-element Composition--FCF-produced Melts

Melt No.	Ingot	Casting Batch	Trace Elements (ppm by weight)			
			Carbon	Silicon	Aluminum	Iron
I	(Specification)		a	a	a	a
	EUf-63S	0448S	248 ^b	120		171 227
II	(Specification)		250	300	a	a
	EUf-58S			140	75	189
	EUf-58SR		720	250		
	EUf-58SR-II	0450SR	307 ^b	325 210		145 241
III	(Specification)		250	300	250	750
	EUf-42S			120	205	591
	EUf-42SR		415	220	126	750
		0453SR		170		736
IV	(Specification)		250	300	750	250
	EUf-46S				787	153
	EUf-46SR		380	190		
		0454SR	259 ^b	130		254
V	(Specification)		a	300	a	a
	EUf-59S			160	262	123
	EUf-59SR		200 ^b	220	290 ^b	149
		0452S			151	172
		0452SR	675 ^b	140 ^b		165

^aNo deliberate addition of trace elements made.

^bTentative analytical value.

While the results of chemical analysis were awaited, the pins were processed, sealed into fuel elements, and the elements assembled into subassemblies. The S-number identifications used in the September report can now be replaced by actual subassembly numbers as follows:

S-1 $\simeq$ C-2110;

S-2 $\simeq$ C-2111;

S-3 $\simeq$ C-2113;

S-4 $\simeq$ C-2112.

In view of the low silicon content, it may be desirable to consider these subassemblies as regular driver fuel (not requiring the extensive postirradiation examination previously planned) and to explore alternate ways of achieving the objective of the experiment.

(g) Effect of Pressure upon Fuel Swelling. The P-1 capsule of six specimens, the first of a series of three, has progressed through Item (9) of the following sequence of postirradiation measurements; P-2, a similar capsule, has progressed through Item (5).

- (1) Neutron radiography of whole capsule.
- (2) Decanning of outer capsule.
- (3) Identification of specimens.
- (4) Photography of assembly and individual specimens.
- (5) Neutron radiography of individual specimens.
- (6) Determination of specimen density (jackets included).
- (7) Determination of specimen lengths and diameters (jackets included).
- (8) Measurement of residual pressure in jackets and taking of samples for gas analysis.
- (9) Decanning of each specimen.
- (10) Photography of fuel pins.
- (11) Determination of pin density,
- (12) Determination of pin lengths and diameters.
- (13) Sectioning of pin for burnup-analysis sample.
- (14) Sectioning of pin for metallographic sample.
- (15) Replication of metallographic sample for electron-microscopic examination.

In capsule P-1, the cladding was ruptured on two specimens. All specimens were swelled to some degree, promoting a belief that burnup may have exceeded the targeted 1.5 a/o at 410 MWd.

Partial data for P-1 pressure specimens are given in Table II.E.8. The four intact specimens have provided some information in advance of direct swelling measurements, which are expected later. Qualitative comparison of appearance of the pins in radiographs has suggested some measure of inhibition of swelling resulting from the higher pressures. Also, the sealing method seems to be reliable up to 1600 psi. The calculated volume differences of contained gas before and after irradiation are of the same order as those predicted for fission-gas release, consistent with precision of measurement. No significant dimensional or density changes were detected on the specimens as jacketed.

TABLE II.E.8. Partial Data for P-1 Pressure Specimens

Spec No.	Cast Pin No.	Melt No.	Mean In-pile Fuel Temp (°C)	Mean In-pile Pressure ^a (psia)	Filling Pressure (psia)	Residual Pressure (psia)	Gas Volume Increase ^b (cc)
1	53-5	409	560	54	39	Ruptured	-
2	-11	409	560	160	64	89	2.3
3	-1	409	560	213	137	Ruptured	-
4	-7	409	560	486	259	290	3.4
5	-3	409	560	876	502	530	3.8
6	-9	409	560	1599	991	975	1.4

^aAssumed no fuel swelling or gas evolution.

^bContains instrument error of about same order of magnitude.

The P-2 capsule was removed from the CP-5 reactor on September 14, 1967, after 223 MWd, for neutron radiography. After the radiographs had been examined, the test was terminated because a suitable spectrum of fuel-swelling values appeared to exist. One specimen, loaded at 325 psig, showed a cladding failure.

The P-3 capsule has been subjected to neutron radiography (at 103 MWd) during the reactor shutdown, but has been returned for additional exposure.

(h) Effect of Heat Treatment upon Fuel Swelling. Six pins from each of the injection-casting batches 0450 and 0452 (Melts II and V made from EUF-58S and EUF-59S, respectively) have been received at ANL, Illinois (see Table II.E.2, Item 6). The pins were only slightly radioactive and can be handled in gloveboxes rather than in shielded hot laboratories. Metallographic examinations and heat-treatment studies have been started. However, the below-specifications silicon contents (detailed in Item 6) made it questionable whether these pins are suitable stock for this rather detailed study. Alternate sources of material are being sought.

(i) Hot-laboratory Examination of Irradiated Fuel

(1) Chemical Analysis. Available and forthcoming chemical analyses of trace-element impurities in EBR-II, Mark IA, fuel materials are given in Table II.E.9. The table is more extensive than usual in several respects: (1) All available analyses from Idaho, ANL-Illinois, and Oak Ridge are included; and (2) the identification of fuel materials for which swelling results exist, and which have been or will be submitted for chemical analysis, are included. This feature is intended to give some feeling for what additional analytical results will become available in the next few months. When analyses of more than one pin of a given casting batch are available, the average of the results is given.

TABLE II.E.9. Trace-element Analyses--EBR-II, Mark IA, Fuel

Casting Batch and Pin No.		Melt Refining No.	Subassembly and Element No.		Trace Elements (ppm by weight)				Analyst
					Carbon	Silicon	Aluminum	Iron	
4001	1	22	C-184	87	107	88	295	470	ANL and OR
4002	9	23	C-180	60	132	94	140		ANL and OR
4002	14	23	C-180	57	121	88			ANL
4002	24	23	C-184	32	139	57	170	190	ANL and OR
4002	79	23	C-184	61		53	168	100	ID
4002	82	23	C-184	60	146	89	115	160	ANL and OR
4002	-	23	Average		135	76	148	150	
4003	3	24	C-186	53	200	162 ^a	100	435	ANL and OR
4003	5	24	C-186	52	199	165 ^a			ANL
4003	19	24	C-186	46	198	290 ^a			ANL
4003	48	24	C-184	57	192	217	200	520	ANL and OR
4003	71	24	C-180	32	193	203			ANL
4003	86	24	C-179	82	214	181	135	420	ANL and OR
4003	90	24	C-179	81	211	216	100	465	ANL and OR
4003	91	24	C-179	80		211	137	280	ID
4003	92	24	C-179	79	189	226	190	440	ANL and OR
4003	-	24	Average		200	208	144	427	
4004	9	25	C-184	7	108	140	125	435	ANL and OR
4008	19	30	C-196	31	137	71	205	155	ANL and OR
4008	30	30	C-197	20	131	92			ANL
4008	69	30	C-196	85	124	97			ANL
4008	-	30	Average		130	87	205	155	
4010	9	C-26	B-331	35	243	260	296	330	ID
							138		
4010	-	C-26	Average		243	200	217	330	
4011	7	C-29	C-196	89	191			240	ID
4011	8	C-29	C-196	88	250	132	220	300	ANL and OR
4011	33	C-29	C-197	56			198	292	ID
4011	43	C-29	C-197	47		151		?	ID
4011	45	C-29	C-197	45	237	122			ANL
4011	-	C-29	Average		221	134	209	277	
4020	40	39	C-199	69	264	127			ANL
4020	92	39	C-198	9	283	139	60	150	ANL and OR
4020	-	39	Average		274	133	60	150	
4022	13	C-30	C-199	90		145			ID
4022	33	C-30	C-217	3	187	180 ^a	215	340	ANL and OR
4022	83	C-30	C-198	87			220	243	ID
4022	95	C-30	C-198	63	240	106 ^a			ANL
4022	-	C-30	Average		218	144	221	243	
4023	56	33	C-199	26	162	132	50	170	ANL and OR
4023	60	33	C-199	23	176				ANL
4023	-	33	Average		169	132	50	170	

TABLE II.E.9. (Contd.)

Casting Batch and Pin No.	Melt Refining No.	Subassembly and Element No.	Trace Elements (ppm by weight)				Analyst
			Carbon	Silicon	Aluminum	Iron	
4024	15	28 C-218 33	207	106	180	180	ANL and OR
4024	69	28 C-218 35		70			ID
4024	82	28 C-218 34	160	100			ANL
4024	97	28 C-217 16			229	238	ID
4024	100	28 C-217 14	233				ID
4024	-	28 Average	200	92	205	209	
4025	2	C-33 C-217 76					ID
4025	4	C-33 C-218 75		174			ID
4025	24	C-33 C-218 1	257	207	105	240	ANL and OR
4025	35	C-33 C-217 84				282	ID
4025	44	C-33 C-217 78	262	213	210	240	ANL and OR
4025	-	C-33 Average	260	198	158	254	
4026	2	C-32 C-218 20		172			ID
4026	7	C-32 C-218 15		175			ID
4026	21	C-32 C-217 57	224	219	165	425	ANL and OR
4026	47	C-32 C-217 38			273	500	ID
4026	54	C-33 C-217 32	248	231	220	440	ANL and OR
4026	-	C-33 Average	236	199	219	455	
4027	9	46 C-217 68		205		285	ID
4027	16	46 C-218 64	206	170	120	140	ANL and OR
4027	21	46 C-217 61	187		77		ID
4027	24	46 C-217 60	229	195	50	135	ANL and OR
4027	-	46 Average	207	190	82	187	
4031	78	C-35 C-229 16		340			ID
4031	88	C-35 C-229 9	210		234	440	ID
4031	-	C-35 Average	210	340	234	440	
4032	11	C-36 C-229 37	207	120	275	385	ID
4032	85	C-36 C-229 33			139		ID
4032	85	C-36 C-229 33					ANL
4032	102	C-36 C-231 56	268	181	69	265	ID
4032	-	C-36 Average	238	151	161	375	
4033	46	C-34 C-231 6		243	233 ^a	365	ID
4033	51	C-34 C-231 5			117		ID
4033	62	C-34 C-229 45	204		110		ID
4033	-	C-34 Average	204	243	153	365	
4034	13	44 C-231 12		102	51	294	ID
4034	19	44 C-230 49	161		229		ID
4034	-	44 Average	161	102	229	294	
4035	14	43 C-231 20		250	78	665	ID
4035	91	43 C-233 13	178		163		ID
4035	-	43 Average	178	250	121	665	
4036	66	C-40 C-233 20		270	20		ID
4036	67	C-40 C-233 19	188			330	ID
4036	-	C-40 Average	188	270	20	330	
4037	65	45 C-233 36	215		187	350	ID
4037	?	45 C-233 ?		160			ID
4037	-	45 Average	215	160	187	350	
4049		C-45 C-247 4	267				ANL
4050		C-37 C-247 23	310				ANL
4051		C-44 S-610 10					ANL
4052	54	C-247 10	228				ANL
4053		C-46 C-247 43					ANL
4054		C-47 S-610 34					ANL
4057	55	C-249 67	265				ANL
4058	C-50	C-249 32					ANL
4059	56	C-249 59	230				ANL

TABLE II.E.9. (Contd.)

Casting Batch and Pin No.	Melt Refining No.	Subassembly and Element No.	Trace Elements (ppm by weight)				Analyst
			Carbon	Silicon	Aluminum	Iron	
4060		C-51					ANL
4061		58					ANL
4062		C-253					ANL
4062	37	60	209	220	263	230	ID
4062	-	Average					
4063		C-249					ANL
4064		C-52					ANL
4065		57					ANL
4066		59					ANL
4067		C-49					ANL
4068		63					ANL
4068	79	63	222	170	325	265	ID
4068	-	Average					
4069		C-53					ANL
4070		C-54					ANL
4071		C-55					ANL
4072		65					ANL
4073	39	66	167	160	412	290	ID
4075		70					ANL
4076		67					ANL
4077		68					ANL
4078		C-57					ANL
4079		64B					ANL
4080		72					ANL
4081		69					ANL
4103	84	84	284	170	223	330	ID
4124	-	89			150	500	ID
		Cast Pin					
SL-1	23	-	151	84			ANL
SL-1	44	-	189	77			ANL
SL-1	-	Average	170	81			
SL-2	27	-	163	84			ANL
SL-3	8	-	167	87			ANL
SL-3	29	-	141	90			ANL
SL-3	-	Average	154	94			
SL-4	21	-	138	81			ANL
SL-5	45	-	194	95			ANL
SL-5	-	-	146	104	35	90	
SL-5	-	Average	170	100			
SL-6	50	-	127	75			ANL
SL-6	-	-	141	126	35	70	ANL
SL-6	-	Average	134	101			
SL-12	48	-	168	115			ANL
SL-12	-	-	-	102	25	60	ANL
SL-12	-	Average	168	109			
SL-14	14	-	154	64			ANL
SL-16	26	-	181	70			ANL
SL-17	47	-	134	65			ANL
SL-17	-	-	-	54	25	60	ANL
SL-17	-	Average	134	60			

TABLE II.E.9. (Contd.)

Casting Batch and Pin No.		Melt Refining No.	Subassembly and Element No.		Trace Elements (ppm by weight)				Analyst
					Carbon	Silicon	Aluminum	Iron	
SL-19	14	-	C-185	48	174	76			ANL
SL-19	46	-	C-179	67	170	96			ANL
SL-19	-	-	Average		172	86			
SL-20	16	-	C-186	32	152	110			ANL
SL-20	40	-	C-185	2	149	85	175	150	ANL and OR
SL-20	52	-	C-184	36	143	90	275	145	ANL and OR
SL-20	-	-	Average		148	95	225	148	
SL-21	20	-	C-186	60	153	87	200	155	ANL and OR
SL-21	-	-	Cast Pin			70	20 ^a	30 ^a	ANL
SL-21	-	-	Average		153	79	200	155	
SL-29	32	-	C-179	13	208	110			ANL
SL-29	51	-	C-184	43	157	102	225	200	ANL and OR
SL-29	68	-	C-179	52	138	116	95	105	ANL and OR
SL-29	-	-	Average		168	109	160	153	
SL-30	8	-	C-184	37	217	108	140	255	ANL and OR
SL-31	-	-	Cast Pin		204	87	80	43	ANL
SL-32	64	-	C-197	72	225	74			ANL
SL-32	-	-	Cast Pin		201	124	97	37	ANL
SL-32	-	-	Average		213	99	97	37	
SL-33	-	-	Cast Pin		166	71	63	27	
SL-35	-	-	B-343						
SL-36	-	-	B-343						
SL-38	-	-	B-343						
SL-39	-	-	B-342						
SL-39	-	-	B-343						
SL-40	-	-	B-343						
SL-42	41	-	C-213	25		78	244	193	ID

? Questionable value, not included in averages.

a Pin to be resampled and reanalyzed.

ANL Illinois analyses.

ANL and OR Carbon and silicon by Illinois, aluminum and iron by Oak Ridge.

ID Idaho analyses.

(j) Characterization of Unirradiated Fuel Pins. As indicated for Item 8, six pins have been obtained from each of injection casting batches 0450 and 0452 for preirradiation examination and for heat-treatment studies. We had planned a detailed examination of these materials in an effort to locate the silicon impurity and to identify its effect, if any, upon the microstructure. However, the relatively low silicon content of these alloys makes the examination of questionable value.

Approximately 150 pins of Melts SL-49 and SL-50 are stored in the Idaho vault and have not been fabricated into fuel elements. These melts were the final two in the SL series and consisted of the scrap accumulated during previous melts. To some extent, they bear the same relationship to the rest of the SL melts as the MR-C and MR-S melts do to the regular MR melts made in FCF. Specimens of SL-49 and SL-50 have been submitted for chemical analysis.

(k) Analysis of EBR-II Fuel-pin Swelling. Values of the fuel-pin temperature, calculated at Idaho by use of the HECTIC-II code, are now being included in the swelling analysis. The simplest method to include temperature in the analysis is to consider the swelling to be a function of the product of temperature and burnup. Machine plots of Y ($\% \Delta V/V$) against $Bu \times T$ (where Bu = average Bu , and T = volume-averaged temperature in $^{\circ}F$) give smoother curves with less scatter and fewer irregularities. These plots are now being analyzed in greater detail.

The average values of the trace-element impurities (Table II.E.9) are retabulated in Table II.E.10 with two different figures of

TABLE II.E.10. Figures of Merit and Trace-element Analyses

Casting Batch No.	Melt Refining No.	Trace Elements (ppm by weight)				$Y/Bu_{\max}^2$ All Data	Y/Bu_{avg} $Bu \approx 0.975$
		Carbon	Silicon	Aluminum	Iron		
4001	22	107	88	295	470	8.46	11.38
4002	23	135	76	148	150	9.89	13.18
4003	24	200	208	144	427	6.17	7.90
4004	25	108	140	125	435	9.60	12.66
4008	30	131	87	205	155	9.89	11.72
4010	C-26	243	200	217	330	3.95	4.50
4011	C-29	221	134	209	277	11.25	13.83
4020	39	274	133	60	150	11.33	14.06
4022	C-30	218	144	221	243	9.15	13.11
4023	33	169	132	50	170	11.15	14.39
4024	28	200	92	205	209	10.12	13.46
4025	C-33	260	198	158	254	5.14	5.52
4026	C-32	236	199	219	455	5.95	7.91
4027	46	207	190	82	187	10.18	13.77
4031	C-35	210	340	234	440	5.23	6.87
4032	C-36	238	151	161	375	6.16	7.97
4033	C-34	204	243	153	365	4.34	5.50
4034	44	161	102	229	294	10.79	14.17
4035	43	178	250	121	665	4.36	5.60
4036	C-40	188	270	20	330	4.33	4.68
4037	45	215	160	187	350	12.31	13.45
4062	60	209	220	263	230	3.60	4.59
4068	79	222	170	325	265	5.10	6.28
4073	66	167	160	412	290	3.85	-
4103	84	284	170	223	330	6.23	-
SL-1	-	170	81			10.62	-
SL-2	-	163	84			10.04	-
SL-3	-	154	94			6.52	-
SL-4	-	138	81			5.37	-
SL-5	-	170	100			9.34	12.78
SL-6	-	135	101			8.85	12.06
SL-12	-		109			10.55	14.37
SL-14	-	154	64			9.91	14.06
SL-16	-	181	70			6.80	13.72
SL-17	-	134	60			10.46	14.29
SL-19	-	172	86			7.83	13.02
SL-20	-	148	95	225	148	8.20	11.26
SL-21	-	153	79	200	155	8.62	14.06
SL-29	-	148	109	160	153	9.86	13.19
SL-30	-	217	108	140	255	10.60	14.53
SL-31	-	204	87			10.50	13.70
SL-32	-	213	99			11.98	15.02
SL-33	-	166	71			7.40	-
SL-42	-		78	244	193	11.58	-

merit to characterize the swelling behavior. The first figure of merit ($\% \Delta V/V/Bu_{\max}^2$) has been used previously and was developed to use all available swelling data, including those at relatively low burnup. More extensive analytical data have been obtained for material that has been carried to higher burnups. Here the distinction between low- and high-swelling fuel becomes more pronounced. A second figure of merit (Y/Bu_{avg}) has been calculated on the basis of only swelling data for which $Bu_{\text{avg}} \geq 0.975$ (equivalent to maximum burnup ≥ 1.09). These two figures of merit are plotted against the silicon content in Figs. II.E.8 and II.E.9.

Fig. II.E.8. Swelling/Max Bu^2 as a Function of Silicon Content

Fig. II.E.9. Swelling/Avg Bu as a Function of Silicon Content

The two plots do not differ significantly, although the second more definitely distinguishes high- from low-swelling material. In general, the SL-type fuel is indistinguishable from the high-swelling 4000 series fuel produced in FCF. With the exception of two casting batches (4032 and 4027, which are being reanalyzed at ANL, Illinois), there is no overlap between high- and low-swelling material. A rather sharp break occurs at about 170 weight ppm silicon. If casting batches 4032 and 4027 are included, there is a region of overlap (marked on Fig. II.E.9) extending from about 150 to 190 weight ppm silicon, with four casting batches involved in the overlap.

The unit swelling can also be plotted against various combinations of trace-element impurities; one such plot is shown in Fig. II.E.10, in which the figure of merit is plotted against the sum of the carbon and silicon contents. The region of overlap is relatively narrow, but five melts are involved.

Fig. II.E.10. Swelling/Avg Bu as a Function of Sum of Carbon and Silicon Contents

g. Mark-II Driver Fuel-element Development(i) Element Irradiation Tests (J. H. Kittel)

Last Reported: ANL-7382, pp. 29-32 (Sept 1967).

(a) Mark-II Fuel-capsule Irradiation Experiments. In July 1967, 37 encapsulated Mark-II fuel elements were shipped to Idaho to make up a subassembly for irradiation in EBR-II up to and including 6 a/o burnup. Details of plans and purposes of this experiment have been reported (Progress Report for August 1967, ANL-7371, pp. 27-29). In the interim, Idaho personnel conducted a series of inspection tests on the capsules. X-ray inspection of the closure welds of the end plugs of the capsules led to the rejection of three capsules for lack of acceptable penetration of these welds (see Table II.E.11). This lack of penetration is being construed as hazardous to the operation of the reactor, and the capsules are being returned to Illinois for repair or replacement. If the defective capsules cannot be repaired by rewelding, they will be replaced.

TABLE II.E.11. Weld Defects in Mark-II Capsules

Rejected Capsule No.	Defect Location	Allowable Defect Size (in.)	Size of Defect (in.)	Wall Thickness (in.)	Penetration (%)
204	Bottom Weld	0.0020	0.0056	0.020	72
209	Top Weld	0.0020	0.0036	0.020	82
214	Bottom Weld	0.0020	0.0036	0.020	82

Several tests were conducted on four sodium-bonded Group M-2 capsules to determine if certain cooling rates could produce a pore-free sodium bond at room temperature. The capsules were subjected to the cooling rates indicated in Table II.E.12. These capsules were reheated

TABLE II.E.12. Sodium-bond Tests; First-run Results

Capsule No.	Cooling Method	Results
227	End immersed to a depth of 3 in. in flowing water.	Reject
236S	Furnace cool.	Reject
273	Insertion into flowing water at 6 in./min.	Acceptable
286S	Insertion into flowing water at 3 in./min.	Reject

above the melting point of sodium, vibrated, and then cooled in flowing water as shown in Table II.E.13. The results are erratic and do not indicate any clear trends. This may be the result of many previous repeated heating and cooling cycles imposed on these capsules.

TABLE II.E.13. Sodium-bond Tests; Second-run Results

Capsule No.	Cooling Rate (in./min)	Results		
		X-ray (Room Temp)	Eddy Current (Room Temp)	Eddy Current (150°C)
227	9	Acceptable	Reject	Reject
236S	6	Reject	Reject	Acceptable
273	3	Acceptable	Reject	Acceptable
286S	12	Reject	Reject	Acceptable

Plans call for the mockup of ten dummy capsules to evaluate two methods of sodium filling and to determine the effects of cooling rates on sodium-bond integrity.

(ii) Subassembly Proof Tests. The objective of this subtask is to characterize the performance of the Mark-II fuel with selected design specifications, under conditions amenable to statistical treatment, particularly in the establishment of confidence levels relating to fuel performance.

The second phase of the evaluation of the in-reactor performance of the Mark-II element consists of irradiations of unencapsulated elements in complete Mark-II 91-element subassemblies. These subassemblies will therefore function in EBR-II as driver fuel subassemblies. They will be exposed directly to sodium flow, so that any effects due to the primary coolant can be determined.

The insertion of five such subassemblies will be timed so that they do not accumulate more than 0.5 a/o burnup before the encapsulated elements with 2 a/o burnup have been removed and examined. The irradiations for the subassembly proof tests are scheduled to be in the range of 2 to 5 a/o burnup.

Selection of the preferred axial fuel restrainer design and the gas-plenum volume for the subassembly irradiations will be based on results of examinations of the irradiations of encapsulated elements at 1 and 2 a/o burnup.

The EBR-II Irradiation Evaluation Committee has approved, in principle, the irradiation of the five Mark-II subassemblies. Final Mark-II element and subassembly drawings have been completed. This task group is presently evaluating the Mark-II subassembly design preparatory to final submittal for approval by AEC-RDT.

(iii) TREAT Experiments (P. J. Persiani)

Performance of this task will require experimental apparatus capable of safely containing Mark-II fuel pins during transient temperature excursions in TREAT. Of equal importance is the requirement that transient instrumentation be as complete and reliable as possible. Apparatus developed for the fast-reactor safety program is being used to supply the needs for the EBR-II Mark-II development effort.

Apparatus to be used in this task includes the Mark-II transparent capsule* and the Mark-II integral sodium loop.** Shipping containers developed for these designs will also be used.

h. Equipment--Fuel Related (E. Hutter)

(i) Improved Gripper and Holddown Force-limit Devices

Last Reported: ANL-7382, p. 33 (Sept 1967).

Assembly of the electrical controls is approximately 70% complete. The drive motor was returned, but is still not suitable and cannot be used with its present mounting. The factory has been requested to rectify the problem.

Components for modifying the gripper mechanism have been fabricated and will be evaluated with the complete test device.

(ii) New Control-rod Subassemblies

Last Reported: ANL-7382, p. 33 (Sept 1967).

(a) Control Rod of Greater Worth. Two approaches are being considered in the continuing design study. One approach employs a modified fuel rod with an absorber follower; there would be a boron-carbide absorber section above a standard 61-fuel-element bundle. As the fuel section is lowered out of the core region, the absorber section moves in to reduce reactivity further. This combination control rod is a desirable configuration from a physics standpoint, but limitations of rod travel and available space introduce design difficulties. The possibility of alleviating these difficulties is being studied.

The other approach involves a control rod that has an absorber section with gas-collecting chambers above and below the absorber, but no fuel section. Extensive studies of this approach have resulted in three concepts. Layout drawings of each concept have been completed and are being evaluated.

*Reactor Development Program Progress Report, July 1967, ANL-7357, p. 130.

**Reactor Development Program Progress Report, March 1967, ANL-7317, p. 106.

(1) Type A. This concept features a boron-carbide core with a hollow center for coolant circulation. A 12-in. gas chamber above, and a 19-in. gas chamber below, the absorber section provide space for helium generated by boron carbide burnup. The principal problem is the thick section of B_4C , which produces temperatures as high as $3500^\circ F$, leading to increased helium generation. Therefore, a method for decreasing the B_4C thickness was developed for the Type B layout.

(2) Type B. The boron carbide is contained in a seven-tube bundle and is connected to the upper and lower gas chambers by drilled passages in the tube support grids. The volume percent of B_4C , sodium, and stainless steel are approximately the same as in Type A. To reduce the complexity and cost of the gas chambers, support members, and manifolds, the Type C concept was developed.

(3) Type C. The seven-tube-bundle array used in Type B was retained, but instead of terminating the tubes at the ends of the B_4C section, the tubes extend the full length of the hexagonal control-rod tube. The B_4C material is held in place by steel balls tightly fitted and plug-welded to the tube walls. The remainder of each tube is used as gas space, except for sections in which shield pieces (steel cylinders) are plug-welded to the tube wall.

Studies are being made to find a material that has the proper porosity to permit helium to pass through and out of the subassembly, and yet not allow sodium to enter the B_4C section. Thoria yttria, which is being tested, has been relatively unaffected by exposure to $900^\circ F$ sodium for 6 months. Preliminary tests show it permits helium to pass through a 1/16-in.-thick section at an acceptable rate. Further leakrate tests in water and sodium are planned.

The compilation of information on the performance of boron carbide as control-rod material in other experimental and operational reactors has been completed. The operating temperatures were not as great as expected for EBR-II operation; therefore, the applicability of these data to EBR-II is questionable.

(iii) Oscillator Rod--Mark II

Last Reported: ANL-7382, pp. 33, 34 (Sept 1967).

A proposal for the new oscillator rod and associated components was prepared and is being reviewed. The modifications used in developing the Mark-IIA oscillator rod were further evaluated; detailed calculations are being made of temperature relationships in the Mark-IIA design.

In addition, other concepts are being investigated as alternatives to the existing Mark-IIA design. One would use a uranium-oxide capsule to balance the heat generated by the boron carbide capsule. Another would use tubes to enclose individual heat-generating material capsules and provide a spiral coolant flow around the boron carbide capsule and the balancing aluminum oxide capsules.

(iv) Engineering Consultation (E. Hutter)

Last Reported: ANL-7382, p. 34 (Sept 1967).

(a) Control-rod Bushings. Additional evaluations were conducted to confirm evidence that aluminum-bronze is a more desirable material for the guide bushings than the beryllium-copper now in use. A single aluminum-bronze bushing is preferred to four individual rings. Further development and fabrication await approvals.

(b) Mark A Irradiation Subassembly Bypass Flow. Proposed modifications for operation of the Mark A irradiation assembly at a higher than normal irradiation temperature were evaluated. The modifications would reduce the temperature difference of the mixing streams and increase the mixing space; they also would increase, by an insignificant amount, the number of fatigue cycles to failure. The evaluation indicates that the desired capability cannot be achieved with a Mark A subassembly unless major redesign is undertaken.

(c) Power Coefficient. During the initial approach to power in Run 25, the power coefficients were significantly less negative than during previous runs. The change is believed to be due primarily to the replacement, between Runs 24 and 25, of the natural-uranium blanket subassemblies in Rows 7 and 8 with stainless steel reflector subassemblies. An explanation of the mechanisms has been evolved that supports the observed changes in power coefficient; analyses of the proposed mechanisms support the explanation.

(1) Hypothesis. During previous runs, the steady decrease of power density with reactor radius caused all blanket subassemblies to be hotter on their inboard faces than on their outboard faces. As a result, these subassemblies tended to bow, so that their upper ends moved away from the core as power was increased. Thus the upper ends of the Row 7 subassemblies did not reach obstructions (the adjacent blanket subassemblies) until relatively high powers, around 20 MWt, were attained. Only then did additional bowing of the Row 7 subassemblies (limited then to inward movement at the core centerline) cause the core radius to begin to decrease, introducing a positive component into the power coefficient. Because considerable thermal expansion of the core subassemblies had already occurred at that power, relatively little decrease in core radius could occur before the core became fully compacted and resisted further reduction.

In Run 25, the substitution of stainless steel for uranium caused less heating; therefore the coolant temperatures were lower in Rows 7 and 8 than before. Apparently, heat transfer from Row 9 caused the outboard faces of the Row 8 subassemblies to be hotter than the inboard faces. This forced the Row 8 subassemblies to bow so that their upper ends moved toward the core as power increased. As a result, the upper ends of the Row 7 subassemblies reached obstructions at a power closer to 5 than to 20 MWt. Less core-subassembly thermal expansion occurred at that lower power; so core radius decreased more, as power increased, before the core compacted enough to resist further reduction. This is believed to be the reason why the regime of reduced negative power coefficient, due to decreasing core radius, starts at a lower power and extends over a wider power range for Run 25 than for previous runs.

The change in power coefficient may also be affected by the greater stiffness of the stainless steel subassemblies. Once the core has compacted, it tends to expand thermally as power increases, but this expansion is resisted by the Row 7 subassemblies, which continue to bow inward at the core centerline. The core radius thus depends on the relative stiffness of the core and Row 7 subassemblies; therefore the stainless steel subassemblies may be more effective in resisting core expansion than the less-stiff uranium subassemblies.

Detailed analyses are being made of these and other hypotheses.

i. Reactor and Primary Coolant System (B. C. Cerutti)

Last Reported: ANL-7382, pp. 34-36 (Sept 1967).

(i) Sodium Sampling Pump and Filter

Last Reported: ANL-7342, p. 11 (May 1967).

Parts for the pump and filter have been fabricated; the shield plug is being filled with high-density concrete. Most of the special fabricated parts, including the ball nut and screw assembly, have been received. The water-test rig has been designed and is being fabricated.

(ii) Design for Sodium Drain Tank (B. C. Cerutti)

The design of the primary sodium storage facility is in progress. A study indicates that the primary tank can be drained by pressurizing to 5 psi and using a vacuum assist. This procedure would eliminate the need for a costly annular EM pump.

(iii) Primary-tank Pressure-vacuum Relief (B. C. Cerutti)

A final draft of the Engineering Submittal for modification of the primary-tank pressure-vacuum relief was completed. This system was described in detail in the Progress Report for August 1967 (ANL-7371). Final drawings are nearing completion. Because of space limitations, the system argon storage tank was reduced 1 ft in height to 15 ft. This reduces the stored volume of argon to about 172 ft³.

(iv) Engineering Consultation (B. C. Cerutti)

The unusually high incidence of incoming power failures to the EBR-II facility was studied. During the past summer, these failures have been characterized by periods of instability preceding total failure. These failures have caused the reactor to scram even when the EBR-II turbogenerator is in operation; although, in theory, the reactor need not scram on incoming power failure when the turbogenerator is supplying local power needs. The study resulted in a recommendation to install monitoring and trip devices which will disconnect the EBR-II site from the outside power system when the incoming power shows signs of instability and the turbogenerator is operating in parallel with the incoming power.

The increased incidence of incoming power instability has been traced to the expansion of the Northwest Power Pool to include ties to other power pools in the United States and Canada and hence is beyond local control.

A mechanical review of the Mark-IIA oscillator rod, special control-rod thimble, storage basket thimble, and the associated handling tools is in progress. This review will be completed before fabrication of the parts is started.

(v) Intermediate Heat-exchanger Seal

Last Reported: ANL-7357, p. 8 (July 1967).

A test rig that simulates the curvature of the intermediate heat exchanger and the preloaded spring segments (baffles) have been completed. Preliminary tests indicate that the baffles fabricated from Inconel-X in 0.025- and 0.015-in. thicknesses are best suited for this application.

A new set of baffles in these thicknesses is being fabricated with increased free length and height. The greater height will compensate for manufacturing-tolerance differences between the heat exchanger and nozzle and will engage the baffle with the inside wall of the nozzle more positively; the result will be better restriction of natural-convection sodium flow in the annular gap between the nozzle and the heat-exchanger shell. The longer baffles will better engage the baffle retainer within the shell.

(vi) General Improvements (B. C. Cerutti)

(a) Purification-cell and Sampling-cell Ventilation. The temporary ventilation supply duct to the sampling cell was removed following testing which showed that a separate ventilation supply duct to this space is not needed. Removal of this duct, which was routed through the doorway, allowed reinstallation of the cell door, and the exhaust system now maintains a slight negative pressure. This will help to confine smoke in case of a sodium fire and prevent its spread out of this cell. Sufficient supply air is provided through the louvered door panels, around the instrumentation panel, and through the ceiling penetrations. Five of the ceiling penetrations have been plugged to prevent an outflow through the purification-system EM pump rectifier blower.

(b) FERD Loop Plugging Meter and Sampling System. The instrumentation and control components for monitoring of temperature and flow and for control of heater power and flow have been installed. Operational tests are in progress.

Initially, the piping loops were heated dry to check the heater control systems and to balance heater power to the piping sections. Temperatures of the various piping sections were monitored during heater power adjustment. Heater control was proved adequate for any desired piping-loop temperature up to 800°F. The piping loops were then filled, and the heater controls were rechecked. Flow was established in the plugging-meter loop and checks made for stability of control and for variance of control over the pump's operating range. All the system protective devices such as leak detectors, high-temperature cutouts, and heater-power limiters received an operational checkout. Flow has not yet been established in the sampling system, and an effort to make the system operational is in progress. Resolution of the problem may not be possible until the next shutdown. With the exception of some cleanup work, the installation is complete.

(c) New Plugging Meter for Primary and Secondary Purification Systems. Installation drawings for the new plugging meter are being prepared.

(d) Blanket-gas Monitor. Specifications for the three single-channel gamma analyzers required for the isotope monitor have been circulated to prospective bidders. Components necessary for transmitting the readings from the gas chromatograph located in the reactor building to the control room are on order.

(e) Primary-tank Bulk-sodium Temperatures. The cold side temperature signal used for obtaining a readout of reactor coolant ΔT is a conditioned signal obtained by cascading amplifiers. The base amplifier

for the system is located in the reactor building in an area subject to both electrostatic and electromagnetic interferences; which result in a noisy signal. Various schemes of shielding, cable routing, etc., had in part corrected for the adverse conditions; but because of the following cascade circuits amplifying the remaining circuit noise, the readout signal was never satisfactory. The noise path was found to be the feedback path between the input and output of the amplifier; therefore this amplifier was replaced by a new amplifier designed to isolate the input from the output. System checks have shown that this change in amplifiers has reduced noise effects by at least one decade (3.0 to 0.3 degrees) at the readout instrumentation.

(f) Surge Tank for Primary Sodium-purification System.

A final draft of the Engineering Submittal modification of the surge tank for the primary sodium-purification system has been completed. Final review of the drawings has been completed, and corrections have been initiated.

j. Secondary Sodium and Power Systems (B. C. Cerutti)

Last Reported: ANL-7371, p. 36 (Aug 1967).

(i) Feedwater System. A study is in progress for the selection of a replacement for the bypass feedwater control valve, SF-3, which is giving trouble in service.

(ii) Turbine Generator. With the exception of the initial pressure governor, the turbine generator is operating satisfactorily. Changes in electrical load are requiring abnormally large changes in steam pressure. The problem will be investigated during the next plant shutdown.

(iii) Sodium Boiler. Information to be used in the selection of replacement safety valves for the steam drum is being received and studied.

k. Instrumented Subassembly (E. Hutter)

Last Reported: ANL-7382, pp. 37,38 (Sept 1967).

(i) Design

(a) Subassembly Attachment and Lead Severance. Bending and free-play tests were performed on the recently completed modified top-end-fixture coupling (axially free without spring-loading) (see Progress Report for July 1967, ANL-7357, pp. 9-12). Free play in three axial planes normal to the faces of the hexagonal lower tube measured an average of 0°23' for the three positions. Slight additional distortion was apparent as the load was increased up to a moment of 2,500 in.-lb, where the test was

stopped, as were previous tests. When the load was released to zero, this coupling returned to its original configuration, indicating elastic behavior.

Torsional tests were completed on specimens of the following couplings: (1) hexagonal support, (2) internal bayonet latch, and (3) modified top-end fixture.

The test of the hexagonal-support coupling resulted in considerable damage where the wedge rods engaged mating surfaces. However, after being loaded to 200 ft-lb in 20-ft-lb increments and then released to zero, the coupling returned to near its original alignment. This coupling, because of its experimental configuration, could be loaded in only one direction.

The internal-bayonet-latch coupling was loaded in both directions to 200 ft-lb in 20-ft-lb increments. A "set" load of 100 ft-lb was applied in each direction before applying the test load. Except for a small discrepancy, this coupling returned to its original alignment upon release of the load to zero from either direction. No damage to the coupling was apparent.

The modified top-end-fixture coupling was also loaded in 20-ft-lb increments up to 200 ft-lb in both directions. Before the test load was applied, a small torque was exerted to "set" the coupling. In the counterclockwise test, recovery to original alignment was complete when the load was released to zero. In the clockwise direction, a small angular discrepancy remained because of inherent motion between corresponding parts.

1. Packaged Loop (B. C. Cerutti)

Last Reported: ANL-7382, pp. 38,39 (Sept 1967).

The feasibility study of the packaged loop is in progress. Technical literature is being searched for applicable heat-transfer information. A search is in progress for materials that will withstand the conditions to which the loop will be subjected. Some detailed flow calculations were made, but no firm conclusions have been reached.

m. Process Chemistry (D. W. Cissel)

Last Reported: ANL-7382, pp. 39-45 (Sept 1967).

(i) Sodium Coolant Quality and Control (D. W. Cissel)(a) Primary Sodium

(1) Carbon Analysis. Table II.E.14 lists the results of analyses for carbon in the primary sodium. Partial results on two of these samples were reported in the Progress Report for September 1967. All analysis results are shown here for clarity. The decreasing trend of results is not considered significant.

TABLE II.E.14. Carbon Analyses on EBR-II Primary Sodium

Sample	Aliquot	Carbon (ppm)	Remarks
8-24-67	1	7.3	Sample slow-cooled, 1 hr
	2	7.0	
	3	8.9	
	4	8.1	
	5	7.4	
	6	4.7	
	7	5.5	
	8	7.5	
	9	4.0	
	10	6.9	
	11	10.4	
	12	7.4	
Avg: 7.0 ± 1.7 at $1\sigma^a$			
9-20-67	1	6.0	Sample quick-chilled, 35 sec
	2	5.1	
	3	5.1	
	4	6.7	
	5	5.1	
Avg: 5.6 ± 0.7 at 1σ			
9-21-67	1	5.6	Sample slow-cooled, 1 hr
	2	4.6	
	3	4.2	
	4	5.9	
Avg: 5.0 ± 0.8 at 1σ			
9-21-67	1	5.3	Sample quick-chilled, 25 sec
	2	3.6	
	3	5.6	
	4	5.1	
	5	4.2	
Avg: 4.8 ± 0.8 at 1σ			

Overall value for 26 analyses: 6.0 ± 1.6 ppm at 1σ .^a 1σ signifies one standard deviation.

Recovery tests have been made to check the performance of the analytical equipment. Results of these tests are shown in Table II.E.15.

TABLE II.E.15. Carbon Recovery Test No. 2, Sodium Present

Run No.	Weight of Sodium Sample (g)	Weight of Carbon Present (μ g)	Amount of Carbon Added (μ g)	Total Carbon Present (μ g)	Total Carbon Found (μ g)	Percent Recovery
1	0.76	3.6 ^a	152.3	155.9	156.0	100.0
2	0.80	3.8 ^a	152.3	156.2	150.0	96.3
3	0.76	3.6 ^a	152.3	155.9	155.6	99.8
4	0.79	3.6 ^b	152.3	155.9	157.4	101.0
						99.2 (Avg)

^aCalculated on the basis of 4.7 ppm carbon in sodium.

^bCalculated on the basis of 4.5 ppm carbon in sodium.

The excellent results of recovery tests, and the very close agreement between all aliquots analyzed thus far, strongly support the validity of the analytical and sampling techniques. Samples are taken by flowing through the sample vessel, then freezing the sample with the outlet line molten and back pressure on the sample. This method of sampling ensures good flushing of the vessel and prevents shrinkage voids from forming during freezing. Aliquots are extruded directly from the sample vessel into the analysis vessel in a dry box, thus minimizing contamination problems.

The only phenomenon that would invalidate the results of this carbon analysis is radial segregation. If a substantial quantity of carbon migrates to within 0.002 in. of the vessel wall, the results of the analysis will indicate a carbon concentration less than that actually in the sample. Clearance between wall and extrusion ram is 0.002 in. A continuing effort is being made to study impurity segregation in sodium samples. (Refer to previous Progress Reports and Cesium Segregation Studies below.)

(2) Cesium Segregation Studies. The mechanism by which cesium segregates to interfaces in sodium samples is under investigation. As reported in the Progress Report for September 1967, rapid cooling of the sample does not reduce the quantity of cesium that segregates at the vessel wall. A test was made which shows that segregation occurs either while sodium is flowing through the vessel or while it is stagnant after flow is stopped but before the sample is cooled. A sample vessel with a screen liner next to the wall was filled with sodium. The flow was stopped

for a few minutes, and then the vessel was drained (all at fairly constant temperature). The ratio of ^{137}Cs to ^{22}Na in sodium remaining on the screen was comparable to that previously seen at the wall of sample vessels.

Two tests were made to examine the theory that segregation could be prevented if the samples were stirred or agitated while freezing.

In the first test, a sample was quick-frozen in an extrusion vessel without stopping sodium flow prior to cooling. In the second test, the sample was quick-frozen in an extrusion vessel while being mixed with a magnetic stirring apparatus. Both samples were analyzed by the method reported in the Progress Report for June 1967, ANL-7349, and the results are shown in Table II.E.16. Also tabulated in Table II.E.16 for comparison are results of a slow-cooled and a fast-cooled (no mixing) sample, which were reported in the Progress Report for September 1967.

TABLE II.E.16. Ratios of $^{137}\text{Cs}/^{22}\text{Na}$ for Extruder-partitioned EBR-II Primary Sodium Samples

Aliquot No.	$^{137}\text{Cs}/^{22}\text{Na}$	$^{137}\text{Cs}/^{22}\text{Na}$	$^{137}\text{Cs}/^{22}\text{Na}$	$^{137}\text{Cs}/^{22}\text{Na}$
	Slow Chill (60 min), No Mixing	Quick Chill (<40 sec), Flowing Sodium	Quick Chill (<40 sec), Magnetic Mix	Quick Chill (<40 sec), No Mixing
1	0.06	0.15	0.10	0.06
2	0.08	0.17	0.17	0.05
3	0.09	0.17	0.15	0.07
4	0.09	0.15	0.16	0.07
5	0.07	0.16	0.15	0.07
6	0.07	0.10	0.12	0.07
7	0.07	0.06	0.09	0.06
8	0.04	0.23	0.05	0.03
Bottom	0.02	0.20	0.13	0.15
Vessel Wall	12.9	14.3	10.3	12.7

Segregation was not prevented by mixing the sample, but the degree of segregation was reduced. These results seem to indicate that segregation can be prevented or reduced to insignificant levels if the sample is vigorously mixed while freezing.

For comparison purposes, the ratio of $^{137}\text{Cs}/^{22}\text{Na}$ for a nonpartitioned bulk sample is 0.26; i.e., if there were no segregation of the ^{137}Cs , the ratio 0.26 would be observed for each aliquot of a partitioned sample.

(3) Analysis of Tin and Bismuth. Chemical-analysis data for tin and bismuth contents in primary sodium from 1963 through the present are listed in Table II.E.17. The tin values are also shown in Fig. II.E.11.

As noted in the table, the values for historical samples were obtained by analysis of portions (aliquots) of sodium taken from aluminum containers. Current samples (September and October 1967) were taken in 10 ml Pyrex beakers and each total sample was dissolved for analysis.

No significant axial segregation of tin was found in the historical samples, but axial segregation of bismuth was found in the historical samples, with the bismuth concentrated toward the bottom of the sample tube (see results for October 18, 1965). Analysis of portions from the top, middle, and bottom of a sample taken in December 1966 gave values of 4, 20, and 44 ppm bismuth, respectively (not shown in Table II.E.17). These results, obtained by independent work in the ANL Chemical Engineering Division, are comparable with the results obtained in Idaho. Apparently segregation and precipitation of the bismuth occurred during cooling of the historical samples.

TABLE II.E.17. Tin and Bismuth in Primary Sodium

Date	Sample Container	Sample Analyzed	Tin (ppm)	Bismuth (ppm)	Remarks
<u>Historical</u>					
10/2/63	1-in. Al tube	Top aliquot	2.9	-	Bismuth not determined
3/11/65	1-in. Al tube	Top aliquot	2.5	<0.5	
6/28/65	1-in. Al tube	Top aliquot	4.0	-	Bismuth not determined
7/9/65	1-in. Al tube	Top aliquot	12	1	
7/26/65	1-in. Al tube	Top aliquot	10	8	
10/18/65	1-in. Al tube	Top aliquot	16	3	Sodium removed mechanically
	1-in. Al tube	~1 in. below top		6	Sodium melted from sample tube.
	1-in. Al tube	Bottom aliquot		45	
6/16/66	1-in. Al tube	Top aliquot	16	3	
8/4/66	1-in. Al tube	Top aliquot	15	5	
11/10/66	1-in. Al tube	Top aliquot	16	2	
5/11/67	1-in. Al tube	Top aliquot	19	1	
<u>Current</u>					
9/22/67	Pyrex beaker	Total		2.3	Tin not determined
9/29/67	Pyrex beaker	Total		2.6	Tin not determined
10/3/67	Pyrex beaker	Total	17	2.5	
10/4/67	Pyrex beaker	Total	18	2.2	
10/10/67	Pyrex beaker	Total	16	2.2	Cold-trap influent
10/10/67	Pyrex beaker	Total	15	1.0	Cold-trap effluent

NOTE: Bismuth values on aliquots of historical samples are not representative of bismuth content of primary sodium because of segregation and settling of bismuth.

Fig. II.E.11. Analysis Results for Tin in Primary Sodium

Assuming a linear gradient of bismuth concentration from top to bottom of the sample tube (as indicated by the results reported in the preceding paragraph), the average bismuth content of the primary sodium in October 1965 through December of 1966 was 22 to 26 ppm. The tin content of the October 1965 sample was 16 ppm. Current primary sodium samples (total sample analyzed) show a bismuth content of ~2.5 ppm, indicating that cold-trapping has removed most of the bismuth from the primary sodium since December 1966. The fact that there was only one day of cold-trap operation between June 28 and October 18, 1965, and 46 days of operation through December 1966, may account for the high bismuth content during that period. The tin concentration, however, remained at a nearly constant level. The data indicate that a sharp increase in tin and bismuth concentrations occurred between June 28, 1965 and July 9, 1965. A further increase, although of lesser magnitude, is also indicated between July 26, 1965 and October 18, 1965.

The source of this increase is believed to be the eutectic tin-bismuth alloy from the rotating plug seal trough(s). During these periods, difficulties were experienced in obtaining indication of satisfactory temperature distributions in the seal troughs and in initiating rotation of the plugs, especially the large plug. These difficulties were summarized in the Progress Report for July 1965 (ANL-7082, pp. 1-2).

Samples of alloy and dross were taken from the seal troughs on both sides (air and argon) of the dip rings during the period July 13-17, 1965. Additional sampling and cleaning operations, involving only the air sides of the troughs, were carried out between July 27, 1965 and September 28, 1965. Seal alloy sampling operations preceding the first indicated increase in tin and bismuth concentrations were carried out on January 18 and 19, 1965, and on May 5, 1965. Both of these operations involved sampling from the argon sides of the troughs.

Although the possibility that the sampling operations were somehow involved must be acknowledged, a review of these operations has produced no reason to suspect that alloy was lost from the troughs as a result. A review of plant operating conditions during the periods of interest is continuing in an attempt to find an explanation for the increased tin and bismuth concentrations.

(4) Other Analysis of Primary Sodium. Results of copper and uranium analysis on primary sodium samples are shown in Table II.E.18.

A primary sodium sample taken September 25, 1967, was analyzed by gamma spectrometry for radionuclides. Results are shown in Table II.E.19.

TABLE II.E.18. Copper and Uranium
in Primary Sodium

Date	Element	Concentration (ppm)
8/27	Uranium	<0.05
9/11	Copper	0.30
9/25	Copper	0.40

TABLE II.E.19. Radioactivity
in Primary Sodium

Nuclide	$\mu\text{c/g}$
^{22}Na	1.8×10^{-2}
^{137}Cs	1.5×10^{-2}

(b) Secondary Sodium

(1) Oxygen Meter Loop. An economizer was installed in the secondary oxygen meter loop, but installation of heaters, insulation, controls and instrumentation has been delayed due to shortage of personnel and higher priority of other plant modifications.

(c) Expanded Sampling Capabilities for Primary and Secondary Sodium

(1) Primary Sodium. An improved sampling system for primary sodium is being designed by the EBR-II Engineering Group. Proposed improvements listed in the Progress Report for September 1967 are being incorporated in the design.

(2) Secondary Sodium. Design of a sampling equipment test loop for the secondary sodium system continued. Piping and heater/thermocouple drawings were completed. The loop, when installed and operational, will provide a versatile system for routine sampling and sample equipment testing in the secondary system.

(d) Effect of Recycling Type 304L Stainless Steel Tubing.

To determine how storage in either normal or dry air affects the sensitization of Type 304L stainless steel tubing that previously had been exposed 7 days to 1100°F sodium and then cleaned (see Progress Report for May 1967, ANL-7342, pp. 20-22), samples of such tubing were subjected to Strauss reagent for accelerated corrosion and their electrical resistivities were measured.

Fig. II.E.12. Strauss Reagent Progressively Raised Resistivity of Unstored Specimens but, after First 1-2 hr, Did Not Continue to Affect Specimens That Had Been Stored 120 days in Normal Air

(1) Normal-air Storage.

Figure II.E.12 compares the resistivity changes of specimens that had been stored for 120 days in normal air with those of specimens that were tested immediately after being exposed to the sodium and cleaned. For the stored specimens, Strauss-reagent attack stabilized after 1-2 hr, but the specimens that had no significant air storage continued to be

attacked during the 5.7 hr in Strauss reagent. A possible explanation relates to the assumption, based on earlier data (see Progress Report for January 1967, ANL-7302, p. 43), that sodium penetrates subsurface grain boundaries and is not removed by the usual cleaning procedures. Thus, the residual grain-boundary sodium would be free to form chemical compounds (oxide or hydroxide) with the air during the 120-day storage, and may then react with the Strauss reagent to only a limited extent.

Figure II.E.13 compares attack of specimens with 120-day air exposure with attack of specimens that had been exposed to air for 10 days (see Progress Report for July 1967, ANL-7357, p. 51). These data indicate that 10-day normal-air storage is not long enough for subsurface sodium to react with air to an equilibrium point, at which point the effective electrical resistivity of the 120-day-stored samples would increase from the as-received value (70 microhm-cm) to a slightly higher but constant value (78 microhm-cm).

Fig. II.E.13.

Storage in Normal Air for 10 days Is Not Long Enough for Subsurface Sodium to React with the Air Sufficiently to Prevent Continued Attack by Strauss Solution. As-received sample was not exposed to sodium.

(2) Dry-air Storage. The data in Figs. II.E.14 and II.E.15 were generated under exactly the same conditions of sodium exposure, sample cleaning, and reagent exposure as those in the normal-air storage tests (Figs. II.E.12 and II.E.13), except that the samples were stored in dry, CaSO_4 -desiccated air. Initially the Strauss reagent rapidly attacks all the specimens that had been exposed to sodium, regardless of storage time in dry air; then the attack plateaus, and then it occurs again from 2.5 hr on. The rapid initial attack and the renewed attack after 2.5 hr on the dry-air stored samples indicate a significant difference in the phenomena related to dry- and normal-air storages. Possibly, in the dry air, hydroxides of sodium could form until the dry-air humidity is further reduced. Then, because of the excess of oxygen in the dry-air medium, oxides could form to constitute an oxide-hydroxide seal in, or associated with, the metal grain boundary. Further reaction of excess oxygen with subsurface entrapped sodium would be inhibited. The initial plateau of 120-day dry-air-stored specimens would be similar to processes described for the normal-air-stored specimens, whereby the Strauss reagent reacts for the first 2.5 hr with sodium plus sodium oxide and sodium hydroxide. After 2.5 hr in Strauss solution, the sodium oxide-hydroxide products would be reduced, leaving the stainless steel with some free sodium remaining in the grain boundary. Comparison of the slopes of the unstored and 10-day dry-air-stored samples with the slopes of the 120-day dry-air-stored specimens between 2.5 and 5.7 hr indicates similarity of reaction. We may infer, then, that between 2.5 and 5.7 hr exposure the Strauss reagent is reacting with grain-boundary-entrapped sodium.

Fig. II.E.14. Attack of Dry-air-stored Specimens by Strauss Solution Starts Rapidly, Plateaus, Then Renews after 2.5 hr

Fig. II.E.15. Comparison of Reagent Attack on Specimens That Had Been Stored in Dry Air for 10 days and for 120 days after Single Exposure to Sodium. As-received sample was not exposed to sodium.

All measurements of effective electrical resistivity have been correlated with photomicrographic inspection. The effects of 120-day normal- or dry-air storage are limited to about 0.002-0.004-in. depths, principally on the inner surface of the tubes.

The effects of a second sodium exposure after 120 days of dry- and normal-air storage are being determined.

n. Experimental Irradiation and Testing (D. W. Cissel)

Last Reported: ANL-7382, pp. 45-51 (Sept 1967).

(i) Experimental Irradiations (D. W. Cissel)

(a) Status of Experiments in EBR-II. The status of all experimental irradiations in EBR-II as of October 19 is shown in Table II.E.20.

TABLE II.E.20. Status of EBR-II Experimental Irradiations, October 19, 1967

Subassembly	Date Loaded	Capsule Content and Number of Capsules ()	Experimenter	Approx Accum Exp (MWd)	Goal Exp (MWd)
XG02	7/16/67	UO ₂ -20 w/o PuO ₂ (1) Stainless Dummies (18)	GE	11,534	13,650
XG03	7/16/67	UO ₂ -20 w/o PuO ₂ (2) Stainless Dummies (17)	GE	11,534	19,450
XG04	7/16/67	UO ₂ -20 w/o PuO ₂ (2) Stainless Dummies (17)	GE	11,534	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	10,957	14,750
XA08	12/13/65	UC-20 w/o PuC (8) Structural (11)	ANL	9,135	19,600
XO10	3/24/66	UO ₂ -20 w/o PuO ₂ (4) Structural (11) Structural (4)	GE ANL PNWL	9,135	19,600
XO12	8/10/66	UO ₂ -20 w/o PuO ₂ (19)	NUMEC	7,765	20,600
XO15	11/15/66	PuO ₂ -UO ₂ (11)	NUMEC	3,110	11,000
XO16	1/13/67	Structural (9) Structural (10)	ANL GE	1,790	5,400
XO17	11/15/66	PuO ₂ -UO ₂ (11) (PuU)C (3) Mk-1A (Metal) (5)	NUMEC UNC ANL	3,110	6,500
XO18	12/6/66	Structural (3) Structural (1) Structural (2) Structural and Heavy Metal Fission Yield Samples (1)	GE PNWL ANL ANL	2,420	21,300
XO19	1/13/67	UO ₂ -20 PuO ₂ (7) (U ₈ Pu ₂)C (3) Structural (8) Graphite (1)	GE UNC PNWL PNWL	1,790	12,500
XO20	1/13/67	Mixed Oxides (9) Mixed Carbides (3) Structural (4) Structural (2) Graphite (1)	GE UNC PNWL ANL PNWL	1,799	25,000

TABLE II.E.20 (Contd.)

Subassembly	Date Loaded	Capsule Content and Number of Capsules ()		Experimenter	Approx Accum Exp (MWd)	Goal Exp (MWd)
XO21	2/27/67	Structural	(7)	PNWL	1,790	21,500
XO22	2/27/67	Structural	(7)	PNWL	1,790	18,000
XO25	10/10/67	Structural	(19)	GE		5,000 ^a
XO26	10/11/67	Structural	(7)	NRL		3,500
XO30	10/10/67	Thermocouple Matl.	(2)	PNWL		No limit
		Graphite	(3)	PNWL		
		Structural	(1)	PNWL		

^aNominal; actual exposure will be established on the basis of best interests of LMFBR program.

Subassemblies XO25, XO26, and XO30 were loaded into the grid at the start of Run 26. The capsules in Subassembly XO25 contain structural material capsules of the GE, L-14 program, and the capsules in Subassembly XO26 contain structural materials from the Naval Research Laboratory. Subassembly XO26 is a special design to permit reactor sodium to flow past the test samples. Subassembly XO30 contains proposed fast-reactor thermocouple materials and general-technology graphite samples, and was loaded as a space-available subassembly to be removed at a time that best serves the LMFBR irradiation program. Approvals-in-principle for the loadings were obtained from LDO on the following dates: June 7, 1967 for XO25; September 6, 1966 for XO26; and December 9, 1965 and July 25, 1966, respectively, for the PNL graphite and thermocouple samples in XO30. Operational approval was given by the Idaho Division Director in September 1967 at the recommendation of the Irradiations Review Committee.

Eleven experimental carbide capsules, part of the 19 capsules mentioned in the Progress Report for September 1967, were received from United Nuclear Corporation.

Twelve re-encapsulated graphite elements were received from Battelle Northwest Laboratory. These capsules will be irradiated on a space-available basis if approved for insertion in the reactor.

The 19 ANL M-3 series irradiation capsules were approved for insertion in the reactor and loaded into experimental Subassembly XO28.

(b) Procurement and Inventory Control of Experimental Hardware. Hardware for experimental capsules is being shipped from Argonne, Illinois to the Idaho Division as rapidly as possible. Capsule tubing, machined end fittings, and lower blanks have already been received. Remaining to be shipped is an inventory of bar stock from which additional end fittings can be machined. Additional supplies of 0.805 and 0.290-in.-OD Type 304 stainless steel tubing have been ordered to supplement the quantities received from Illinois.

(ii) Nondestructive Testing (D. W. Cissel)

(a) Capsule Examinations. X rays of the welds in the ANL Mark II fuel capsules indicated defects in three capsules, which will be returned to the experimenter for rework. All other capsules in the series were satisfactory.

The PNL graphite capsules were examined by radiography. The weld penetration on the upper end fittings was, in general, less than 90% of the capsule-wall thickness on nine of the twelve capsules. Since they contain no fuel material, these capsules will be held until an Irradiation Review Committee can determine their acceptability for insertion in the reactor.

Irradiation Subassembly XO27, containing the 16 GE ElH series of irradiation capsules, was disassembled. Each capsule was X rayed to determine if any changes had taken place while the subassembly was held in the storage basket and during handling operations. Each capsule was in satisfactory condition. Sodium levels and bond conditions were similar to those found in the initial examination. Two capsules were observed to have bond voids larger than 1/8 in. These two capsules were heated to 500°F and x-rayed hot. No voids of any consequence were seen in the hot x-rays.

Four UNC carbide capsules were examined for suitability for insertion in the reactor. Three were found acceptable; one was rejected for incomplete weld penetration of an end fitting weld.

(b) Other Nondestructive Testing. Ultrasonic equipment was used in a test arrangement to determine its value in locating sodium levels in capsules with narrow annuli. The sodium level in a special test capsule was quite apparent, but sodium levels in actual capsules were much more difficult to determine.

The eddy-current equipment for bond tests is near completion and requires only the coils to place it in service. Experimental work will be required to determine the best system and to establish standards for its use.

Fabrication and assembly of electronic equipment have begun on the eddy-current flaw detector which will be used for capsule tubing inspection.

A precision optical comparator was received and is being used for quantitative weld examination. Depending upon the quality of the X ray, measurements of 0.001-0.0005 in. can be made on weld and wall thickness.

The melt-wire temperature monitor, reported in the Progress Report for September 1967 under Reactor Analysis and Testing, Operational Data, was neutron-radiographed while positioned in a groove in the lower end of a subassembly triflute axial blanket assembly; just above the fuel section. In this location, the stainless steel weights were difficult to see through the large mass of stainless steel in the blanket section. Alloying with boron was suggested to make the weights more clearly visible.

Several casting batches of EBR-II Mark IA fuel produced in the cold line were x-rayed, but the results were not satisfactory because of the difficulty in penetrating the uranium. A radioisotope source was obtained from INC to complete the examination.

Several welds in the process piping in the EBR-II sampling system associated with the FERD loop were x-rayed.

(c) Antimony-Beryllium Neutron Source. Bids have been requested for a foil-positioning fixture for the FCF neutron radiography facility.

(iii) Handling and Examination (D. W. Cissel)

(a) Irradiated Capsules from Subassembly XO11. Capsules from Subassembly XO11, except capsules HOV-4, HOV-10, F4F and 5P-9, were washed with demineralized water in the IBC. A metallographic comparison will be made between the unwashed capsule walls of 5P-9 and the washed walls of 5P-12. The other three unwashed capsules contained failed fuel elements.

Cermet capsules 5P-9 and 5P-12 were returned to PNL for examination. Capsule F4D was returned to GE for the same purpose. At the request of ANL, part of the examination of the foregoing capsules will be concerned with the condition of the capsule wall (see Progress Report for September 1967).

Capsules HOV-4, HOV-10, and F4F were examined visually and by a dye-penetrant technique at the TAN RML hot cells. Defects that might breach the capsule walls of HOV-4 and HOV-10 were found, but no such defects were found on F4F.

Capsules HOV-4 and HOV-10 were returned to ANL-MET in Illinois. Further testing on these capsules has been requested to identify and characterize the failures. Capsule F4F will be returned to GE for the same purpose.

(b) Testing of Subassembly Sodium-removal Procedures

(1) Metallographic Examination of Capsule-wall Material (Type 304 Stainless Steel) from Capsules Irradiated in Subassembly XG06. GE has agreed to perform a metallography examination program on selected samples of capsule-wall material from Subassembly XG06, but work has been delayed for the past two months. GE now reports that the material to be investigated has been moved into the hot cells and the work should be mostly completed during November 1967.

(2) Cyclical Testing of Type 304L and 304 Stainless Steel Tubing Exposed at 1100°F, Washed in Water, and Stored in Air. The Reactor Engineering Division is nearing completion of Phase IV of this program, including 120-day air-storage periods.

Comparison of data for the 120-day air-storage period, in both dry and moist air, with the corresponding data from the 10-day air-storage period of Phase II indicates insignificant changes in microstructure. There was, however, a noticeable lessening of corrosion by Strauss reagent for the "120-day stored" steel compared to the "10-day stored" steel of Phase II. This reduction in corrosion appeared more pronounced for the normal air storage than for the dry air storage. When samples were resoaked in 1100°F sodium after the first 120-day storage period, the intergranular attack by Strauss reagent was greater on specimens stored in dry air than on those stored in moist air.

This cyclical testing program will be terminated after the investigation of effects from the second 120-day air-storage period.

(3) Cyclical Testing of Stressed Capsules (Type 304 Stainless Steel) at EBR-II. Subassembly X900 containing the pressure-stressed tubing samples was removed for the second time. Another set of seven stressed samples was removed. The subassembly is to be returned to the storage basket for the third cycle of sodium soaking and cleaning. Soaking time for the second cycle was extended, as was that for the first cycle, because of a period of reduced ($<700^{\circ}\text{F}$) primary sodium temperature.

Apparatus for measuring and depressurizing the helium gas used to stress the capsules did not function properly, necessitating minor modifications and checkout tests. Until the measuring equipment performance is satisfactory, both sets of samples are being stored in air.

(4) EBR-II Subassembly Recycle Test. The sodium-exposure, water-wash recycle test was delayed because none of the irradiated subassemblies selected for the test was removed from the reactor. Plans and schedules are now being formulated, and a subassembly has recently been made available.

(c) Sodium-bond Integrity Demonstration. Data are being accumulated in Idaho on the condition of capsule sodium bonds after the bonds have been melted (as by immersion in the primary sodium), then frozen during standard handling procedures. These data will be combined with the previous data reported by ANL-MET in order to support plans for reirradiation of experimental capsules containing sodium-bonded fuel elements.

(iv) Safety Review of Experiments (D. W. Cissel)

The proposal for experimental Subassembly XO28 containing 15 test capsules of ternary metal alloy, U-15 Pu-10 Zr, and four capsules of Mark IA fuel was reviewed from safety and operational aspects, and approval was recommended.

The proposed criteria for irradiations of unencapsulated fuel elements were revised on the basis of comments received from AEC-RDT, ANL-LDO, and others.

o. FCF Process Analysis and Testing (M. J. Feldman)

Last Reported: ANL-7382, pp. 51-52 (Sept 1967).

(i) Operational Analysis (M. J. Feldman)

The computer program for preirradiation fuel-pin file has been finished and is being placed into operation. FCF is obtaining two magnetic tapes for use with the IBM-1620 computer. As soon as these tapes are received, the preirradiation file data will be transferred from the fuel-pin identification cards to the magnetic tapes. The primary purpose of the file will be to identify other computer tapes that contain specific scientific data pertaining to fuel pins of interest. The EBR-II computer center is continuing to program the scientific data tapes.

(ii) Test and Analytical Methods (M. J. Feldman)

The accuracy of, and precision for, analysis of fission constituents in uranium-fission alloy has been compared by atomic absorption versus wet-chemical spectrophotometric methods. Six samples of depleted uranium-fission alloy from the "cold line" were analyzed in triplicate by atomic absorption without prior separations and by the "standard colorimetric" methods employed to date in the ID laboratory for these elements. Atomic-absorption calibration curves were prepared from solutions containing depleted uranium, in addition to the element sought. No other special parameters were required; however, colorimetric standards and atomic-absorption standards were analyzed by both methods and agreed within the limits of the analytical methods. The data obtained are summarized as follows:

Element	Atomic Absorption w/o + 1 σ	Colorimetric w/o + 1 σ
Mo	3.57 $\pm$ 0.05	3.61 $\pm$ 0.08
Ru	3.03 $\pm$ 0.06	2.94 $\pm$ 0.03
Rh	0.454 $\pm$ 0.008	0.460 $\pm$ 0.007
Pd	0.301 $\pm$ 0.006	0.298 $\pm$ 0.010

Atomic-absorption analysis for fission constituents in uranium-fission alloy will be used on all cold-line samples to replace the time-consuming wet-chemistry methods, since the precision and accuracy of the methods are the same and the atomic-absorption method is a great deal faster.

An investigation has been started of the condition of the sodium bond in irradiated driver-fuel elements that have been removed from the reactor and dismantled in the Vertical Assembler-Dismantler (VAD). The parameter being explored is the effect of maintaining the elements in the vertical attitude at all times during removal from the reactor, transfer to the FCF, and dismantling in the air cell. Previous operations required that the subassembly be turned to the horizontal position for dismantling. Bond tests conducted on the "as-received" elements dismantled in the previous fashion nearly always indicated disruptions in the sodium bond. These disruptions were indicated in tests conducted at elevated temperature (135°C) as well as in those performed at cell temperature. In the current program, fuel elements from selected subassemblies are subjected to eddy-current bond-testing at ambient temperatures and temperatures above the melting point of sodium. These elements are then rebonded and again bond-tested. No data are yet available.

The results of this investigation should be pertinent to future driver-fuel-element or experimental irradiation programs which require reinsertion of the experiments in the reactor following removal for examinations, etc.

(iii) Advanced Process Methods (M. J. Feldman)

To date, efforts in this subtask have consisted of familiarization of the FCF staff with the advanced processes being developed by the Chemical Engineering Division for potential pilot operation in Idaho.

The use of a portion of the FCF (after a vendor fuel supply has been established) to accomplish those test capabilities deleted from the present HFEF plan, will in the future use a major portion of the manpower assigned to this subtask. This will represent a change in scope for this subtask.

p. FCF Equipment Improvement (M. J. Feldman)

Last Reported: ANL-7382, pp. 52-53 (Sept 1967).

(i) Auxiliary Equipment (M. J. Feldman)

The bottom-loading and -unloading transfer cask, which is being designed for the interim subassembly storage area, will use an in-cask subassembly grapple system. Construction of a model of one proposed grapple system is proceeding satisfactorily. The model is expected to be tested in November.

q. Hot Fuel Examination Facility (HFEF)--Feasibility and Cost Study (N. J. Swanson)

Last Reported: ANL-7382, pp. 20-21 (Sept 1967).

Contract negotiations and technical meetings have been scheduled with the architect-engineer (Norman Engineering Company of Los Angeles). The initial effort of the architect-engineer will be to review the preliminary cost estimates being prepared by the Laboratory with the assistance of a consulting engineering firm (F. C. Torkelson of Salt Lake City). These estimates are based on the recent reduction in scope of HFEF. Conceptual drawings, preliminary design criteria, and a systems outline for this concept have been completed.

r. Superheater and EM Pump Study and Test

(i) Electromagnetic Pump Vibration Test (R. A. Jaross)

Vibrations in the ac electromagnetic (EM) pump presently installed in the secondary system, and in the new (replacement) pump, are to be measured. Normal and detrimental vibrations of the pump duct are to be distinguished to ensure satisfactory operation.

The pump vendor has outlined a three-phase test program: (1) Investigate pump-duct vibration signals, (2) design and install vibration-monitoring system on presently installed ac-EM pump, and (3) design and install vibration-monitoring equipment on the replacement pump. The first phase would take 17 weeks, the second would entail 14 weeks, and the third would require 11 weeks. A significant portion of the work would require that EBR-II be placed in standby condition.

(ii) Superheater Vibration Study (R. A. Jaross)

Although previous design analyses and experience with the superheaters at 45-MWt operation indicate that the probability of detrimental vibration is very low, the units are being evaluated further to ascertain the

likelihood of future failure due to wear or fatigue caused by flow-induced vibrations. The analyses consider cross-component, parallel-component, and steam flow, thermal stresses, impact velocity, and rotary motion. Fatigue also will be evaluated.

The superheater vibration study has been initiated. The design specifications and existing stress analyses of the superheater are being reviewed; particular attention is being given to features that can influence vibrational characteristics, for example, initial preload and clearances. In addition, literature is being reviewed for applicable failure experience in operational heat exchangers and for current methods for analyzing tube vibrations.

Knowledge of structural natural frequencies and mode shapes is basic to any vibration analysis and fatigue study. The baffle plates provide for lateral restriction of tube motion within the limits provided by the clearance. In the calculation of tube natural frequencies, we assume initially that the baffles act as internal supports restricting the tubes to no motion at these "supports."

The duplex tube is approximated as a uniform beam (this neglects the influence of the shock tube) with three internal supports. The method of receptances is well suited to calculating natural frequencies of vibrating systems of this type. The frequency equation is obtained as the determinant of a three-by-three matrix of interior receptances of the beam. These interior receptances are tabulated* for any combination of free, clamped, pinned, or sliding end conditions. With the assumption of clamped-end conditions, a computer code has been written to evaluate the eigenvalues (roots of the frequency equation). The computer results will allow us to compute the first few natural frequencies of tube vibration for the two support configurations resulting from the different baffle spacings obtained when the shell is considered in halves.

s. Reactor Improvements, Nuclear Instrument Test Facility Study
(B. C. Cerutti)

Last Reported: ANL-7382, p. 54 (Sept 1967).

The test equipment to be used for the flux and gamma mapping of the 01 thimble has been assembled. Shop calibration, alignment of components, and general preparation of equipment are in progress. With the exception of the gamma detector, scheduled for delivery later this year, the equipment required for flux mapping is on hand.

*Gladwell, G. M. L., and Bishop, R. E. D., Interior Receptances of Beams, J. Mech. Eng. Sci. 2, No. 1 (1960).

t. Feasibility Study of Fuel Failure Detection--Chemical and Mechanical Methods (C. E. Crouthamel and F. A. Cafasso)

The main objective of the program on fuel failure detection is to develop chemical methods and instrumentation for identifying the position of fuel failure in an operating reactor. The program has as its basis the use of specific radiochemical or isotopic tags for individual subassemblies; in the event of fuel-element failure, the tag would be released to the sodium coolant, and its detection in the sodium would identify the position of fuel failure. The tag elements would be present in the sodium in concentrations of the order of a nanogram per gram of sodium. For stable tags, added specificity can be obtained by using isotopic compositions that are different from natural compositions. In this way, inadvertent contamination of the sodium coolant by the selected tag element would not be mistaken for a fuel failure.

(i) Trace Elements Analytical Techniques. To ensure a reliable identification of the faulty subassembly, it will be necessary to select applicable tags, evaluate their detection sensitivity in sodium, and develop a suitable method of placing the tag in the fuel pin so that it remains available for dispersion into the sodium during a long in-pile irradiation period. An attempt will be made to select and test up to 100 unique tags so that each driver subassembly in a reactor core such as EBR-II can be identified. As part of this program, the trace-element content of the EBR-II coolant will be thoroughly surveyed, and the changes that occur under operating conditions will be determined.

Initial work will be concerned with developing analytical techniques to measure the trace-element content of the sodium coolant. Spark-source mass spectrometry has been selected as the principal method for trace-element analysis. Other techniques that will be evaluated are surface-ion emission mass spectrometry and activation analysis.

Spark-source mass spectrometry will provide high sensitivity and rapid multielement analysis. However, problems of the relatively high volatility and low ionization potential of sodium vapor must be controlled before the full capabilities of the instrument can be realized.

To minimize the sodium vapor problem, consideration is being given to the use of matrix isolation techniques to transfer the trace element of interest from the bulk-sodium phase into a phase (e.g., lithium metal) that has both lower volatility and higher ionization potential. Radioactive tellurium is being used to test this technique. Na_2Te dissolved in sodium metal will be contacted with lithium metal. Free-energy data favor the reaction

by about 15 kcal/mole. Thus the trace element of interest should transfer into the lithium phase and present an ideal sample for mass-spectrometric analysis.

Shipment of a spark-source mass spectrometer for use in this program is scheduled for the first of November 1967. (This spectrometer will also be used in a separate program on the chemistry of irradiated fuels.) The instrument is an Associated Electronic Industries MS-7, made in Great Britain, and modified according to our design for use with radioactive materials. The source unit is one designed for ready interchangeability and decontamination, a feature that is necessary in sodium work because of the "memory" effect of slightly volatile materials. The source unit is also equipped with a liquid-nitrogen-cooled sample holder for work with such materials.

The facility for the mass spectrometer is under construction.

(ii) Tag-confirmation Study. The preliminary selection of tags and development of sensitive analytical techniques will require confirmatory studies of the stability of the tag in the sodium-coolant stream and the continued availability of the tag in the fuel pin during an extended irradiation period.

The major effort thus far has been devoted to the design of a small-scale experimental loop for circulating liquid sodium. This design work is nearing completion. The loop will provide facilities for (a) testing and comparing various sodium sampling techniques, and (b) evaluating the behavior of very low concentrations of radiochemical or stable isotopes (approximate levels, nanogram per gram of sodium) in circulating sodium.

Conceptual designs of the loop facility are also nearing completion. Features of the system include (a) an attached inert atmosphere glovebox in which isotopes can be added to, and samples can be extracted from, circulating sodium; (b) equipment for reduction of sodium impurity levels by cold trapping; (c) equipment for evaluation of the plugging temperature of the loop sodium; (d) apparatus to permit separation of less-volatile impurities from sampled sodium by distillation; and (e) a gas-purification unit (recirculation type), that will maintain a high-purity inert atmosphere over exposed sodium samples.

A cost estimate of the experimental loop has been completed, and a tentative sequence for component fabrication and equipment ordering has been formulated. The loop is to be constructed of Type 304 stainless steel.

Operational parameters for the loop are being calculated.

(iii) Mechanical Failed-fuel Locator (E. Hutter)

As an interim alternative to chemical-tagging methods that sample the total coolant from the reactor to detect fission-product gases, and thus determine which subassembly's cladding is leaking, mechanical methods for sampling coolant flowing from a single subassembly are being investigated. The initial engineering study has defined four approaches:

(1) Place a suspect subassembly in the fuel-unloading machine, reduce argon (coolant) flow to the minimum so that the subassembly will heat and emit fission gases, and then monitor the argon gas for fission products.

(2) Position a suspect subassembly under the fuel-transfer port, use a special gripper to secure a hollow tube assembly over the subassembly adapter, and then raise the subassembly out of the sodium into the argon blanket, where it would heat and give off fission gas, which would be detected in a sample of the argon.

(3) Place a closed-base tube in the sodium in the catch basin, put the suspect subassembly into it, cap the tube, and then depressurize the tube so that the pressure differential would cause fission gas to emerge through the cladding flaw and rise to the top of the tube, where there would be an inert gas that could be sampled and analyzed for fission gas.

(4) Place a sampling mechanism in the reactor in place of a control rod, rotate a nozzle on the mechanism over the adjacent subassemblies to sample sodium flow, and monitor the sodium for fission-gas release during reactor operation.

Study indicates that the fourth concept is the most sensitive because monitoring would occur during reactor operation. The sampling device would fit atop a dummy control-rod subassembly and hold it in place. When the open lower end of the retractable sampling tube is extended over the coolant exit of a neighboring fuel subassembly, differential pressure would drive the coolant sodium up to an annular space approximately 1 ft above the bottom of the rotating plug. From the annular space around the sampling tube, the sodium would flow downward by gravity into the bulk sodium in the primary tank; hopefully, turbulence caused by the change in flow direction would allow the entrapped fission gases to escape from the sodium and rise through a collector and condenser. The separation might be facilitated by introducing argon into the sodium stream before it leaves the sampling tube. The sampling device could be rotated 360° so that sodium samples could be taken from any of the six surrounding fuel subassemblies.

A preliminary layout has been prepared. Methods for measuring sodium flow, monitoring temperature, and collecting sodium vapor are being studied. The possibility of using the same device to sample liquid sodium will be investigated. Principles will be verified experimentally so that feasibility can be determined.

u. EBR-II Materials--Coolant Compatibility (J. E. Draley)

Last Reported: (Sept 1967).

All unirradiated and irradiated tensile specimens (except the 18-4-1 tool steel specimens) for the first surveillance subassembly from EBR-II have been pulled in the D-301 facilities. Data are being interpreted.

In consideration of the amounts of tin and bismuth that have been present in EBR-II primary sodium, specimens of stainless steel irradiation capsules are being cross sectioned and analyzed for these two elements by microprobe trace. This analysis will reveal any substantial grain-boundary or other local penetration if it has occurred.

2. Outside Fuel Procurement (C. E. Stevenson)

Last Reported: ANL-7382, pp. 11, 12 (Sept 1967).

a. Mark IA-IB Fuel Procurement

(i) Fuel Elements. Aerojet has completed the modification of facilities for the production of EBR-II fuel elements, and has commenced the installation of equipment. Restrainers, tips, and tubing for cladding fabrication are being produced by subcontractors, and two lots of restrainers have been received. Receipt of the first lot of tubing is expected by the end of October.

Pulsed-eddy-current equipment for receiving inspection has been set up and put into operation; this equipment is presently being used to detect natural defects in tubing supplied by ANL and to begin preparation of defect standards.

The initial batch of wire procured for wrapping the cladding tubing did not meet specifications because of marks and kinks, and was rejected. The order was cancelled, and a new order is being placed with another supplier. The welding process is being developed for the wire-to-tube weld.

Analytical methods were developed and analytical equipment was put into operation for analyses of stainless steel and fissium-alloy components. Equipment for inert-atmosphere and sodium analyses was set up.

Process equipment design was substantially completed, and most equipment components are under construction. The method planned for pin-element volume measurement was reviewed in detail and appears to be satisfactory. The wire-wrapping and -welding machine is substantially completed. The inert-atmosphere glovebox is scheduled for delivery in December 1967.

ANL is considering rewriting contracts to include tensile-strength and burst-test specification limits for fuel-element tubing. (The supplier is now required only to furnish test data.) Data are being obtained by ANL on previously procured tubing, including that now in use for FCF production, to provide a basis for setting such specifications.

(ii) Subassembly Hardware. A further meeting was held with a vendor interested in negotiating a contract to supply 500 sets of core, inner blanket core-type, control, and safety-rod preassemblies.

3. Operations--Reactor Plant (G. K. Whitham)

Last Reported: ANL-7382, pp. 2-5 (Sept. 1967).

a. Operations. The reactor was operated for 90 MWdt during Run 26A in the performance of tests and measurements designed to determine its kinetic behavior in a core loading similar to that of Run 25. The reactor was then shut down and the primary sodium tank temperature reduced to 600°F for replacement of one of the control rods and its drive with the reactor oscillator. Installation of the Fuel Element Rupture Detector (FERD) loop plugging meter and sampling system was completed, and normal FERD loop operation was verified.

The gripper jaws of the control-rod drive in reactor position No. 7 had been experiencing some sticking during fuel-handling operation for the past several months. Each time the jaws were closed a manual assist was required to get through midtravel. Since the oscillator rod was to be installed in position No. 8, the control-rod drive was removed from position No. 7 and replaced with the control-rod drive from position No. 8, which operates satisfactorily. The oscillator rod was then in position No. 8. The malfunctioning unit from position No. 7 is presently stored in the reactor building and will be disassembled for inspection and repair.

Following heatup of the primary system to 700°F, reactor loading adjustments originally planned for Run 26 were completed.

Run 26B was then started, control rods Nos. 5 and 10 were intercalibrated, the reactivity worth of the oscillator rod was calibrated, and low-power oscillator experiments were performed. Power-coefficient measurements up to 45 MWt were taken. At each 5-MWt increment, the torque required to manually turn the oscillator rod was measured. At 30 MWt the torque requirement increased, indicating possible bowing and rubbing of the oscillator rod. Because of the increasing torque necessary to turn the rod, measurements were discontinued above 35 MWt.

Further oscillator experiments (at frequencies from 0.0025 to 8.8 Hz) and rod drops were conducted at 500 kW and 22.5 MWt. At 30 and 41.5 MWt, tests were performed at a minimum frequency of 0.1 and 0.3 Hz, respectively, because of rod rubbing below these values.

Reduced flow experiments at 22.5 MWt and 90, 75, 60, and 54% flow were conducted. Between physics program experiments, the reactor was operated at 45 MWt.

Reactor loading changes made for Run 26B included the installation of three experimental subassemblies, XO25, XO26, and XO30, in Rows 4, 6, and 7. Reactivity adjustments were required to provide sufficient

reactivity for operation until the scheduled intermediate shutdown (1100 MWd). Two fueled subassemblies were installed in Row 6 in place of subassemblies loaded with stainless steel. Additional changes included the substitution of a standard C type for a 1/2 worth in Row 4 and a 1/2 worth for a standard B type in Row 6.

b. Operational Support and Maintenance

(i) Spare Parts. A spare air operator for the main feedwater pump pressure control valve (VC-596) was received, but the piston stop for the operator was broken on its arrival. The supplier is being notified of this condition.

No major purchase orders were issued for inventory additions to spare parts.

(ii) Maintenance. Piping for the secondary sodium oxygen analyzer has been installed.

The thrust bearing in the Fuel Unloading Machine (FUM) argon-cooling system 10-hp turbo compressor motor failed. A spare motor was installed.

The FUM gripper was removed, cleaned, and reinstalled because of difficulty with its operation attributed to sodium oxide accumulation over a 2-month period.

A bellows failed in the throttle valve for the primary purification plugging indicator loop, necessitating replacement of the valve.

The No. 2 primary pump motor generator eddy-current coupling was replaced with a spare coupling to allow for the 5-year inspection and reconditioning of the original unit in accordance with the manufacturer's recommendation.

The water-level sight glass on the No. 4 feedwater heater was repaired, and the safety valve was replaced on this heater.

c. Operator Training. Revision of the training material for the Power Plant qualification area continued. All lesson-plan material for P-10 (Power Plant Operator Procedures) are now completed and available. Illustrative and descriptive training material for the FUM were revised to describe the new Mark III gripper and its operation.

The training group is currently providing orientation material and short lectures for the SEFOR supervisory group from the General Electric Company.

d. Operating Manual. Operating Manual revision bulletins Nos. 60 and 61, containing revisions to the Fuel Handling division of the Manual were issued. Bulletin No. 60 covers the Restricted Operation and Special Procedures sections, and Bulletin No. 61 covers the General and Unrestricted Operation sections.

The rough draft for the new section covering operation of the Fuel Element Rupture Detector Loop has been completed.

A new section covering operation of the Fission Gas Monitor is being drafted, and the Normal Operating Procedures section of Division II is being revised.

The Special Operating Procedure entitled "Emergency Procedure for Fuel Element Rupture" was revised to apply to the current reactor run.

e. Planning and Scheduling. The plant operating schedule was revised to reflect the changing schedule from shutdown, to operation, and back to shutdown. Bar charts and network schedules were prepared for completion of the sample loop and plugging loop, oscillator-rod installation, and physics testing for Run 26. Daily surveillance was maintained on the critical path items.

Preparations are continuing for early issuance of the long-range plan for operation, testing, and maintenance.

4. Operations--Fuel Cycle Facility (FCF) (M. J. Feldman)

Last Reported: ANL-7382, pp. 5-11 (Sept 1967).

a. Hot-line Operation. Production schedules were reduced during the past month to accommodate the surveillance of subassemblies from reactor Run 25 and as a result of the delay in the reactor startup and subsequent delay in discharge of Run 25 fuel for reprocessing. Subassemblies from this run were measured to determine (a) overall subassembly straightness, (b) twist, (c) width across buttons, and (d) hexagonal tube width directly above and below the buttons. Experimental support was provided for the examination of capsules from XO11 and X900.

Approximately 17 kg of iron-contaminated fuel was shipped to ICPP for recovery.

Table II.E.21 summarizes the production activities for the month.

TABLE II.E.21. Production Summary

	10/1/67 through 10/21/67	Total This Year
1. Subassemblies Received:		
Core, Control, Safety	10	54
Other	2	124
2. Subassemblies Dismantled (for processing)	12	60
3. Subassemblies Dismantled (for examination, etc.)	2	37
4. Subassemblies Fabricated (includes seven made with unirradiated fuel and two in cold line)	12	147
5. Subassemblies Transferred to Reactor	9	84
Subassemblies Stored in L&O Vault and Interbuilding Corridor	4	28
6. Elements Decanned:		
From Irradiated Subassemblies	524	4,921
Rejects	104	876
Other	0	0
Total Decanned	629	5,797
<u>Melt Refining</u>		
	<u>Irradiated Fuel</u>	<u>Recycle Material</u> <u>New Fuel</u>
7. Number of Runs	5	4 4
8. Average Pour Yield, %	92.3	94.1 94.6
1967 to date (Total)	40	68 52
<u>Processing</u>		
	<u>10/1/67 through 10/21/67</u>	<u>Total This Year</u>
9. Injection-casting Runs (Total Number)	9	148
10. Elements Processed:		
Accepted	742	13,066
Rejected	145	1,743
11. Elements Welded	746	12,935
Rewelded	4	17
12. Elements Leak-tested:		
Accepted	883	12,384
Rejected	22	261
13. Elements Bonded (including recycle)	1676	15,882
14. Elements Bond-tested:		
Accepted	985	12,784
Rejected	61	776
15. Elements to Surveillance	205	612
Number of Subassemblies	7	47
<u>Waste Shipments</u>		
16. Cans to Burial Ground	2	87
17. Oxide and Glass Scrap to ICPP	1	51

b. Cold-line Operation. Four runs were made in the alloy-preparation furnace: two runs to produce depleted uranium-fissium alloy to be used as standards for chemical analysis (5 and 2.5% fissium); and two enriched production runs. During the second of the enriched runs, the crucible cracked, allowing a small amount (300 g) of alloy to contact the graphite susceptor. This was the second use of this crucible.

A successful pour was obtained, but the susceptor had to be removed to free the crucible and recover the uranium. The furnace has been rebuilt and is again operable. A double susceptor was used in the rebuilt furnace. If crucible cracking reoccurs, the inner susceptor can be replaced without disturbing the remainder of the furnace.

Work has continued on refining the operating parameters of the injection-casting furnace. Five depleted and two enriched uranium-fissium runs were made. Rods from these castings were X rayed to aid in evaluating the data produced by the eddy-current portions of the pin processing equipment.

The subassembly transfer coffin (for transfer of subassemblies to storage at Idaho Chemical Processing Plant) has been received in Idaho and is being leak-tested and checked for alignment with FCF transfer equipment.

c. Maintenance and Repair. The bridge drive on the air-cell crane continued to fail intermittently. The bus bars that supply power to the air-cell crane bridge have been coated with a conductive lubricant. The lubricant does not appear to have alleviated the problem. Other possible solutions to the problem are being investigated.

Three electromechanical manipulators incurred roller failures on the main telescoping-tube system. Repair has been achieved, and probable causes are being investigated.

d. Operator Training. On-the-job training continued for technicians who had been given new work assignments.

As part of the FCF safety program, introductory and refresher courses in the use of the Scott Air Pak were given to all members of the FCF operating organization.

e. Operating Manuals and Reports. The following work was done on the FCF Operating Manual: Five sections or parts of sections were being drafted; four new sections and two revised sections were completed and submitted for review; eleven sections or parts of sections were approved for issuance; and four sections, revisions of sections, or parts of sections were issued as accepted portions of the Manual.

f. Analyses

(i) Chemical Analyses. The number of fuel production analyses on cast-pin and ingot samples, together with the average values and range, are tabulated below:

Production Fuel			
Analyzed for	Number	Average Value	Range
U (total and isotopic)	12	95.16 w/o	94.90-95.75
²³⁵ U		52.21 w/o	52.13-52.31
Pu (total and isotopic)	4		
Ru	12	1.90 w/o	1.81-2.09
Pd	9	0.182 w/o	0.18-0.19
Rh	11	0.295 w/o	0.276-0.318
Zr	15	0.053 w/o	0.032-0.090
Nb	5	0.021 w/o	0.020-0.024
Fe	28	324 ppm	114-736
Al	4	180 ppm	150-205
Si	8	180 ppm	120-300

In addition to production analyses, top, center, and bottom samples from two cast pins of depleted uranium-8% fissium alloy were analyzed in triplicate to determine homogeneity of the fissium constituents. No significant variation between samples was observed. The results are tabulated below:

Average of Three Analyses						
Element	Pin No. 1			Pin No. 2		
	Top, w/o + 1	Center, w/o	Bottom, w/o	Top, w/o	Center, w/o	Bottom, w/o
Mo	3.66 ± 0.08	3.76	3.59	3.52	3.51	3.51
Ru	2.93 ± 0.08	2.93	2.92	3.03	2.97	3.03
Pd	0.306 ± 0.010	0.310	0.284	0.293	0.303	0.293
Rh	0.454 ± 0.008	0.467	0.456	0.451	0.465	0.463
Zr	0.120 ± 0.016	0.108	0.144	0.126	0.109	0.172
Nb	0.035 ± 0.014	0.020	0.024	0.018	0.017	0.020

(ii) Postirradiation Analysis of EBR-II Fuel. Selected driver-fuel elements from subassemblies discharged from EBR-II at the end of Run 25 are being collected in the FCF air cell. These elements will be subjected to tests to determine the magnitude of irradiation swelling in the uranium-5% fissium alloy. No data are yet available.

The ICPP is conducting a corrosion testing program to determine whether the stainless steel cladding of EBR-II fuel elements that had been exposed to reactor sodium would become defective as a result of being stored in a water storage basin. After three months' storage in water, five stainless steel-clad EBR-II fuel pins continue to show no penetration of the cladding.

The ICPP is also conducting a leach test in which a small piece of de-clad EBR-II fuel is being exposed to storage basin water. Initial results show that, excluding an initial large removal of cesium-137 [$13.2 \mu\text{Ci}$ of $^{137}\text{Cs}/(\text{cm}^2)(\text{day})$], the leach rate during a subsequent 16-day exposure average $0.19 \mu\text{Ci}$ of $^{137}\text{Cs}/(\text{cm}^2)(\text{day})$. (This latter rate is only $1/2000$ of that obtained in earlier work by ICPP for an exposed uranium-aluminum-alloy fuel element.) Two other radioactive species ($\text{Zr-}^{95}\text{Nb}$ and ^{144}Ce) have been identified. However, their amounts have been decreasing; this indicates the possibility that these two species are either being plated out on the test vessel or precipitated as a sludge. The amount of leaching of these species will be determined when the test vessel is cleaned after completion of the test.

PUBLICATIONS

LIQUID-METAL FAST BREEDER REACTORS--CIVILIAN

Compatibility of Type 304 Stainless Steel with Sodium Paste Reactor Fuels

M. G. Chasanov

Nucl. Sci. Eng. 30, 310-311 (Nov 1967) Note

Methods for the Numerical Evaluation of Fourier Integrals

R. Gold, C. E. Cohn, and I. Olson

J. Comput. Phys. 1(4), 536-540 (June 1967)

Corrosion of Vanadium-Base Alloys in Sodium at 550° to 750°C

S. Greenberg, W. E. Ruther, and H. A. Levin

Alkali Metal Coolants, Proc. IAEA Symp. on Alkali Metal Coolants, Corrosion Studies, and Systems Operating Experience, Vienna, November 28-December 2, 1966. Intern. Atomic Energy Agency, Vienna, 1967, pp. 63-84

Examination Techniques for High Burnup U-Pu Fuel Elements for Fast-Breeder Reactors

B. J. Koprowski and F. L. Brown

Proc. 15th Conf. on Remote Systems Technol. Am. Nucl. Soc., Hinsdale, Illinois, 1967, pp. 63-67

Monte Carlo Analysis of Reactivity Coefficients in Fast Reactors. General Theory and Applications

L. B. Miller

ANL-7307 (March 1967)

Microanalysis of Solid Fission Products in Mixed-Oxide Fuel Irradiated in EBR-II

D. R. O'Boyle, F. L. Brown, and J. E. Sanecki

Trans. Am. Nucl. Soc. 10(2), 462 (Nov 1967) Abstract

Addenda to the Hazards Summary Report on the Oxide Critical Experiments

K. E. Plumlee, D. S. Craig, L. R. Dates, J. O. Juliano, C. N. Kelber, and D. H. Lennox

ANL-5715 Addenda (March 1967)

A Wide-range Counting-Campbell Neutron-flux Detection System

G. F. Popper, W. C. Lipinski, and J. M. Harrer

ANL-7224 (April 1967)

650°C Sodium Free-surface, Stress, and Wear Effects on Reactor Component Metals

F. A. Smith, E. S. Sowa, E. L. Kimont, and C. R. Sutton

Alkali Metal Coolants, Proc. IAEA Symp. on Alkali Metal Coolants, Corrosion Studies, and Systems Operating Experience, Vienna, November 28-December 2, 1966. Intern. Atomic Energy Agency, Vienna, 1967, pp. 247-260

III. GENERAL REACTOR TECHNOLOGY

A. Applied and Reactor Physics Development-- Research and Development

1. Theoretical Reactor Physics

a. Cross-section Data Evaluation (C. N. Kelber)

Last Reported: ANL-7382, p. 95 (Sept 1967).

The FORTRAN programs used in handling ENDF/B data are being converted from the CDC-3600 to the IBM system 360. The programs CHECKER and CRECT have been compiled on the system 360, and several test problems have been run successfully. Program DAMMET has been compiled and is currently being checked out. Little difficulty was encountered in compiling the programs, but much difficulty arose in determining the appropriate job-control language for successful manipulation of the tapes involved.

An ANL topical report is being written describing the compilation of ENDF/B data for magnesium, titanium, vanadium, molybdenum, and gadolinium. Listings and graphs of the data are to be included.

b. Reactor Computations and Code Development (B. J. Toppel)

Since delivery of the IBM/360 Model 75 in July, effort has been expended in acquiring familiarity and facility with regard to use of the IBM System/360 50-75 computer complex. Although substantial progress in the ARC system development had been made on the IBM/360 Model 50 during the previous year, acquisition of the Model 75 and operation of the computer within the ASP environment has resulted in many of the problems engendered by new computer acquisition. This initial shake-down period now appears to be nearing a successful conclusion. Future work on the ARC system* is to be concentrated on implementation of four general computational areas. These are:

- (1) One-dimensional diffusion theory.
- (2) One-dimensional S_n transport theory.
- (3) Fuel cycle.
- (4) Cross-section preparation.

An ANL topical report** describing the motivation and initial configuration of the ARC system has been completed and is being prepared for publication.

* Reactor Development Program Progress Report, ANL-7286 (Dec 1966).

** Toppel, B. J., The Argonne Reactor Computation System, ARC, ANL-7332 (to be published).

An important area of study required to extend the computational capability of the ARC system is that of flux synthesis. The methods of variational flux synthesis have been successfully used during the past few years in the calculation of multidimensional flux shapes for complex reactor configuration with acceptable accuracy and at considerable savings in computer time. The need for time-saving approximation techniques is even more pressing in the field of fast-reactor design, where it is usually necessary to obtain solutions to the neutron-diffusion equations using many energy groups.

A survey of the work published in this field has been conducted. The idea of using discontinuous trial functions in conjunction with an appropriate variational principle appears to be the one with the better potential for accuracy and economy. Most of the work reported is still at the experimental stage. An effort has been initiated to specify a consistent variational formulation allowing the use of trial functions which exhibit quite general discontinuities.

2. Experimental Reactor Physics

Last Reported: ANL-7357, p. 96 (July 1967).

a. Basic Reactor Data (R. Gold)

(i) Systematic Error Due to Angular Correlation in Fission-counter Coincidence Measurements. Large discrepancies exist in ν and σ_f measurements.* One source of these differences for experiments that use coincidence counting as a central feature of the calibration is the strong angular dependence between fission fragment and emitted neutrons.** In the past it was assumed that, as the fission-fragment counter efficiency approaches unity, this effect rapidly vanishes. In practice, the surrounding neutron detector rarely has true 4π detection efficiency, a deficiency that arises because a beam hole is usually situated along the high fragment efficiency axis of a parallel-plate fission counter.†

A thin coating of ^{252}Cf (about 10^5 fission/sec) was prepared by vacuum evaporation and incorporated into a fission counter which could be calibrated by Hornyak button prompt-neutron coincidence counting. The parallel plate fission detector was rotated about an axis running through the foil, producing the distribution shown in Fig. III.A.1. Even though the fission counter has $99.0 \pm 0.5\%$ fission fragment efficiency, the deviation due to orientation extends to 1% for a neutron detector intersecting 60% of a 2π solid angle. A front/back asymmetry of 1/3% is also noticeable.

*Westcott, C. H., et al., Atomic Energy Rev. 3(2), 3 (1965); also, Proc. Conf. Nuclear Data, Paris, IAEA (Oct 1966); also De Volpi, A., submitted to Power Reactor Technology.

**Maslin, E. E., et al., Phys. Rev. 139, B852 (1965); also, De Volpi, A., and Porges, A., Proc. Conf. Nuclear Data, Paris, IAEA (Oct 1966).

†Porges, K., and De Volpi, A., Proc. Symp. Standardization of Radionuclides, Vienna, IAEA (Oct 1966).

Fig. III.A.1. Fission Rate as a Function of Angle between Plane of Fission Foil and Hornyak Button Fast-neutron Detector

3. Nuclear Data--Cross-section Measurements

a. Fast-neutron Cross Sections (A. B. Smith)

(i) Elastic and Inelastic Neutron Scattering from Deformed-Even-Heavy Nuclei. A detailed experimental study of elastic and inelastic neutron scattering from Sm, Hf, Gd, and Yb was carried out for incident neutron energies of 0.3 to 0.8 MeV. The results define the elastic cross sections for each of these elements in detail and provide the inelastic excitation cross sections pertaining to the low-lying rotational states. The measured data are nearly fully processed and appear to be of excellent quality. The coming program of measurements will (1) extend the above results to incident neutron energies of 1.5 MeV, (2) define the inelastic excitation of high (less than 1.2 MeV)-lying states, and (3) provide detailed high-resolution information pertinent to the excited structure of the respective isotopes.

(ii) Elastic and Inelastic Scattering from Medium-weight Nuclei. Studies of neutron scattering from germanium have been completed to incident neutron energies of 1.5 MeV. The elastic scattering cross sections show intermediate resonance structure correlated with that previously

observed in the total neutron cross sections. The inelastic excitation cross sections of states at 560, 590, 690, 835, and 1035 keV have been accurately determined. Calculations are now in progress with the objective of relating the experimental results to the optical potential. The germanium results are requisite to the use of Ge(Li) gamma-ray detectors in the determination of high-priority gamma-ray production cross sections.

The measurement of elastic and inelastic scattering (resulting in the excitation of the 0.34 MeV state) cross sections of vanadium has been completed. The data are being analyzed in an attempt to relate the observed structure to the dynamics of the compound nucleus formation.

(iii) Scattering from Light Nuclei. Efforts in this area have been devoted to the study of elastic and inelastic neutron scattering from fluorine. Results recently obtained provide a good knowledge of these scattering processes to an energy of < 1.5 MeV. The data are now being interpreted in terms of an R-matrix formalism and intermediate structure. The future program will extend the previous high-resolution total neutron cross-section measurements from 0.6 to 1.5 MeV and will provide the high-resolution information necessary for a detailed resonance-fitting procedure.

(iv) Capture Cross-section Measurement of ^{236}U . The neutron capture cross section of ^{236}U was measured by means of the activation method. This cross section is of interest in reactor calculations since ^{236}U occurs in fuel elements due to the ^{235}U (n, γ) process and is in specific response to requested measurement from DRDT. An absolute measurement was made at 30-keV neutron energy applying the associated activity method; measurements were also carried out at some energy points between 150 and 1000 keV relative to the ^{235}U fission cross section. Counting equipment for relative γ -counting (of ^{237}U) and absolute α -counting (of ^{241}Pu , which decays to ^{237}U) is being built.

(v) Fission Cross-section Measurements in the keV Energy Range. The ^{235}U fission cross section, important for both fast reactor design and neutron cross-section experiments, is being measured. The measurement will cover the 10-1000-keV energy range in the first stage of the experiment with an accuracy of about 2%. A gas scintillation counter was built for the detection of the fission fragments, and a "beam-catcher" type neutron detector was designed for the neutron intensity measurements. Extensions of the energy range are under study, as is the application of the experimental equipment to other fissile nuclei and to the determination of the cross sections for the reactions ^{10}B (n, α , γ) and ^6Li (n, α).

(vi) Standard Measurement of Total Neutron Cross Section of ^7Li . Preliminary measurements on the total neutron cross section for ^7Li were made in 1-keV intervals between 300 and 400 keV. The ^7Li sample used is the thickest of two new samples recently obtained from Los Alamos.

These measurements are being made in an effort to clarify fine structure originally found on the large resonance at 260 keV. The two new samples are thin enough to permit a more reasonable transmission at the high cross sections near and on the resonance. Special care was taken in the sample fabrication to eliminate any possible contaminants. The preliminary measurements were not detailed enough to evaluate the presence of fine structure, but did duplicate the average cross section found previously in this interval. Additional measurements are planned.

(vii) Elastic Neutron Scattering and Polarization Measurements.

An important function of any neutron cross-section program is the determination of nuclear constants and cross sections for use in the reactor-physics field. In many instances, the cross sections desired by the reactor physicist have not been measured, either because the materials are not available or because the measurement may be difficult and/or expensive. To fill this "gap" a sufficiently good understanding of the basic physical processes must be achieved so that nuclear models may be devised for calculating the unmeasured quantities.

The present program of elastic neutron scattering and polarization measurements is designed to help fill this need by permitting a better definition of the parameters of a nuclear model, e.g., the optical model. Optical model parameters are especially sensitive to the polarization, so that a simultaneous fit of the scattering cross section including the polarization should yield a good determination of the parameters.

For all the polarization measurements in this program, the ${}^7\text{Li}$ (p,n) reaction serves as the source of polarized neutrons; thus a knowledge of the polarization of the emitted neutrons is a basic requirement. To better define the polarization of the emitted neutrons from the ${}^7\text{Li}$ (p,n) reaction, we have made precision measurements of the scattering cross sections and polarizations of the neutrons elastically scattered from magnesium and silicon. The resonance structure observed in the cross sections was fitted to a set of phase shifts determined from R-matrix theory. Since the phase shifts determine the polarization of the scattered neutrons, it was possible to use both magnesium and silicon as polarization analyzers for the neutrons emitted from the ${}^7\text{Li}$ (p,n) reaction. Although the data are not completely analyzed in terms of the polarization of the source neutrons, the results do agree fairly well, at 1 MeV, with a very recent measurement at Duke University.

Scattering cross sections and polarization measurements are being performed for a number of nuclei throughout the periodic table. At present, partial results are in process for Al, S, Ni, Cu, Se, Y, Mo, and Sn.

(viii) Fast-neutron Reaction Cross Sections--Theory. The formula for the average fast-neutron cross section depends on optical-model transmission coefficients as well as on the distributions and correlations of resonance pole parameters.* The latter are under study by means of analytic and numerical methods.**

b. Fission Process Data (A. B. Smith).

(i) Correlation of Mass, Energy, and Angle in Neutron-induced Fission. To understand the mechanism of mass division in fission, its dependence on the levels at the saddle point must be known. The correlation of angular anisotropy and mass from fission at well-defined excitation energies gives information concerning their dependence.

The kinetic energies of pairs of fragments emitted at 0, 45, and 90° to the incident neutron beam were measured by back-to-back surface-barrier detector systems. About 100 $\mu\text{g}/\text{cm}^2$ of fissionable material is deposited on a 130- $\mu\text{g}/\text{cm}^2$ nickel foil and placed near a 4- cm^2 detector so that nearly all fissions are detected. Three additional detectors determine the complimentary fragments and define the respective angles. The fragment masses were calculated from the relation

$$\frac{M_1}{M_1 + M_2} = \frac{E_2}{E_1 + E_2}.$$

Measurements have been made for 6-MeV neutron-induced fission of ^{235}U and processing of the data has been completed. Table III.A.1 lists the average masses of the light and heavy peaks and their variances, the average total kinetic energy after neutron emission and its variance, and the peak-to-valley ratio of the mass distribution. Similar quantities are shown for thermal-neutron fission of ^{235}U obtained with the same experimental setup.

TABLE III.A.1. Summary of Mass-angle Fission-fragment Results for ^{235}U

Neutron Energy	Angle (deg)	$\bar{M}_L$	$\sigma_L^2 E$	$\bar{M}_H$	σ_H^2	$\bar{E}_T$	σ_E^2	$\frac{\text{Peak}}{\text{Valley}}$
Thermal	0	97.3	34.8	139.1	34.6	170.5	112.6	294
	45	97.2	33.5	139.1	33.6	170.4	112.7	333
	90	97.4	34.8	139.1	34.9	170.5	111.7	304
6.0 MeV	0	98.0	45.9	138.6	47.1	169.2	114.9	21.9
	45	98.0	45.1	138.6	46.2	169.0	115.8	22.4
	90	97.9	45.8	138.2	46.3	169.5	113.7	22.1

*Moldauer, P. A., Phys. Rev. 135, B642 (1964); Phys. Rev. Letters 18, 249 (1967).

**Earlier results in this program were reported in Moldauer, P. A., Phys. Rev. 157, 907 (1967); Phys. Rev. Letters (Oct. 30, 1967).

c. Instrumentation and Digital Techniques (A. B. Smith)

(i) Reactor Physics On-line Computer Planning. A two-day conference was held at Argonne between three representatives of the Idaho Division and two representatives of the Reactor Physics Division. The purpose of this conference was to assemble a set of specifications for three computer systems with specific degrees of compatibility and equipment sharing. As a result of this conference, a prospectus was set up for vendors to provide equipment meeting the system specifications. The sections of the prospectus dealing with the requirements for the Reactor Physics Division have been written (inclusive of diagrams) and are being integrated with the requirements of the Idaho Division.

d. Determination of Other Nuclear Constants (A. B. Smith)

(i) Electron-capture Branching Ratio of ^7Be . The knowledge of the branching ratio in the ^7Be -decay is important for the application of the associated activity method to highly accurate cross-section measurements at 30-keV neutron energy. The MnSO_4 -bath method and counting of the 469-keV γ -transition with a well-calibrated NaI detector will be applied to measure the branching ratio. The setup for this experiment was designed. A $4\pi\beta$ -proportional counter, which will be a part of the $4\pi\beta$ - γ -coincidence apparatus, was designed to measure absolute radioactive source strength needed for the calibration of the NaI detector.

B. Reactor Fuels and Materials Development

1. Research and Development-- Behavior of Reactor Materials

a. Irradiation Behavior of Advanced Ceramic Materials (L. C. Michels)

(i) Irradiation of High-temperature Materials

Last Reported: ANL-7382, pp. 98-99 (Sept 1967).

Ceramic fuel materials of interest for FBR's and other high-temperature reactor applications are being irradiated. The materials include UC-20 w/o PuC, PuC, and US. All materials are clad in Nb-1 w/o Zr. They are being irradiated in instrumented temperature-controlled capsules in MTR. The irradiations are summarized in Table III.B.1.

TABLE III.B.1. Status of Ceramic-fuel Irradiations in Progress in MTR

Capsule No.	Specimen No.	Design Parameters				Operating Conditions		
		Fuel Composition (w/o)	Effective Density (%)	Cladding OD (in.) ^a	Cladding Thickness (in.)	Max Cladding Temp (°C)	Estimated Burnup to Date	
							a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ ^b
56-11	MV-2	UC-20 PuC	79	0.281	0.012	470	8.8	23
56-8	MV-3	UC-20 PuC	81	0.281	0.012	715	9.2	25
56-8	MV-5	UC-20 PuC	80	0.281	0.012	705	8.9	24
56-11	MV-6	UC-20 PuC	80	0.281	0.012	480	9.3	24
56-8	S-7	US	80	0.281	0.012	535	7.5	15
56-8	S-8	US	89	0.281	0.012	725	9.5	19
56-8	S-9	US	76	0.281	0.012	750	9.5	18
56-8	S-10	US	91	0.281	0.012	690	9.5	21
56-11	S-15	US	82	0.281	0.012	380	6.4	13
56-11	S-16	US	90	0.281	0.012	510	8.7	19
56-11	S-17	US	88	0.281	0.012	500	7.2	16
56-11	S-18	US	77	0.281	0.012	610	9.1	17

^aAll cladding is Nb-1 w/o Zr alloy.

^bBased on effective density.

Neutron radiographs were taken of the ANL-56-13 capsule after its removal from MTR in September 1967. Fuel pin No. 1, containing PuC, appears to have failed during irradiation, whereas fuel pins No. 2, 3, and 4, containing UC-20 w/o PuC, appear to be intact. Pins No. 2, 3, and 4 appear to be highly porous with the porosity increasing in Nos. 2 and 4 toward the bottom of the pins, but uniform in No. 3. This may indicate the possibility of failure in Nos. 2 and 4 as well as in No. 1. More detailed analyses will be made when the capsule is disassembled.

b. Structures and Properties of Advanced Fuel Material

(i) Hot-pressing of Ceramic Fuel Materials (O. L. Kruger and J. B. Moser)

Last Reported: ANL-7349, pp. 96-99 (June 1967).

The hot-pressing characteristics of UC, UP, and US powders have recently been investigated since high-density specimens needed for the measurement of thermal properties can be obtained with this method

of fabrication. Material densified by this process usually has a fine grain size because grain growth does not occur at the relatively low temperatures and short times used with this technique. Rossi and Fulrath* have shown that the densification rate can be expressed by the equation

$$\frac{d\rho}{dt} = \frac{40D\Omega_0}{kTd^2} \sigma(bP),$$

if the strain rate for creep, as expressed by the Nabarro-Herring relationship, is assumed to be proportional to the shrinkage rate of the pressure-sintered specimen. The term (bP) is a stress-correction factor which takes into account the influence of die wall constraint on the creep rate and the effect of porosity on applied pressure. Since the stress-correction term is uncertain, even for well-characterized (spherical) powders, and the appropriate values have not been determined for the materials used in this investigation, the data on the uranium compounds were analyzed according to the above equation. This equation and the one derived from the Mackenzie-Shuttleworth model** differ only by a slight change in numerical constant and the correction factors normally used with the latter expression.

Curves of density as a function of time for pressure-sintered UC, UP, and US are shown in Figs. III.B.1, III.B.2, and III.B.3, respectively. In this preliminary study, the runs were made over a limited temperature range and for periods of one and one-half to two hours. A wider range of temperature and longer times are needed to obtain a better analysis of the pressure-sintering process; therefore further work is planned. At 1400°C the UC specimen attained a maximum density of 87% of theoretical. However, the final density at 1300°C was 81%, and at 1200°C it

Fig. III.B.1. Density of Pressure-sintered UC as a Function of Time

*Rossi, R. C., and Fulrath, R. M., J. Am. Ceram. Soc. **48**, 558 (1965).

** Mackenzie, J. K., and Shuttleworth, R., Proc. Phys. Soc. (London) **62**, 833 (1949).

Fig. III.B.2
Density of Pressure-sintered
UP as a Function of Time

Fig. III.B.3. Density of Pressure-sintered US as a Function of Time

was still lower at 63% of theoretical. Riley* has shown that densities of about 94% of theoretical can be obtained at 1400°C in 30 min with finely divided UC ($< 4 \mu$ -dia particle size). Uranium monophosphide, with a particle size of about 5μ , hot-pressed to a density of 90% of theoretical in 1 hr at 1200°C, whereas the same material with an average particle diameter of about 7μ attained a density of only 85% of theoretical at the same pressure

*Riley, B., United Kingdom Atomic Energy Report, AERE-R 4154 (1962).

and temperature. The above data show that the densification rate increases with decreasing particle size, all other factors being equal. This behavior suggests a diffusion-controlled process and encourages more recent investigators to invoke diffusion theory for the analysis of the hot-pressing data.

Density on a log scale is plotted against time in Figs. III.B.4, III.B.5, and III.B.6 for the various uranium compounds. The diffusion coefficients calculated from the slopes of these plots were consistent with the temperature of measurement, only for the range of porosity from about 10 to 20%. Diffusion coefficients were calculated for UC by use of the vacancy volume of $3.04 \times 10^{-23} \text{ cm}^3$ estimated by

Fig. III.B.4. Porosity (Log Scale) of Pressure-sintered UC as a Function of Time

Fig. III.B.5. Porosity (Log Scale) of Pressure-sintered UP as a Function of Time

Fig. III.B.6. Porosity (Log Scale) of Pressure-sintered US as a Function of Time

Lee and Barrett.* These data, given in Table III.B.2, are in excellent agreement with creep diffusion coefficients calculated by the same authors. Their calculations were based on measurements of compressive creep rates of UC by Norreys and Wheeler.** The creep diffusion coefficients were closer to the diffusion coefficients of carbon than those of uranium. Any reasonable stress-correction factor for the hot-pressing data would still leave our measurements in fairly close agreement with the creep data.

TABLE III.B.2. Calculated Diffusion Coefficients for Pressure-sintered UC, UP, and US

Compound	Temperature (°C)	Average Particle Diameter (μ)	Diffusion Coefficients ^a ($\text{cm}^2 \text{sec}^{-1}$)
UC	1300	25	1.6×10^{-10}
	1400	25	3.9×10^{-10}
UP	1200	~5	2.9×10^{-11}
	1200	~7	3.2×10^{-11}
	1300	~7	2.9×10^{-10}
	1400	~7	5.4×10^{-10}
US	1200	~15	1.3×10^{-10}
	1300	~15	2.6×10^{-10}
	1400	~15	6.3×10^{-10}

^aNo stress-correction factor was used for these calculations.

Diffusion coefficients for pressure-sintered UP and US are also given in Table III.B.2. The vacancy volume for UC was used for these calculations. The diffusion coefficients for UP and US at 1400°C are of the same order of magnitude as the value for UC. Since the rates of material transport are relatively equivalent, the compounds should have about the same plasticity up to 1400°C.

The plots of porosity versus time (Figs. III.B.4 to III.B.6) show that no end-point density was reached; therefore the data are consistent with a diffusion model for densification. The change in slope of the curves at densities greater than 90 to 95% of theoretical can be attributed to gas entrapped within the closed pores which cannot diffuse out rapidly enough to avoid decreasing the densification rate. This process has been described by Coble[†] for pressureless-sintered Al_2O_3 . The densification

* Lee, H. M., and Barrett, L. R., Trans. Brit. Ceram. Soc. 66, 159 (1967).

** Norreys, J. J., and Wheeler, M. J., Trans. Brit. Ceram. Soc. 62, 183 (1963).

† Coble, R. L., J. Am. Ceram. Soc. 45(3), 123 (1962).

rate above about 95% of theoretical density would be expected to be extremely slow for pressure-sintering studies conducted in argon. The curvatures of the density time plots are consistent with this predicted behavior; however, very fine powder of UC has been hot-pressed to a density greater than 98% of theoretical in the short time of 30 min.* These results suggest that in argon the densification rate in the closed pore stage depends on the pore distribution and perhaps on the pore configuration as well. Additional vacuum and argon hot-pressing data are needed to clarify the effect of gas entrapment on the end-point density.

(ii) U-S-O System (P. D. Shalek)

As part of an overall investigation of phase relations in the system U-S-O, a tentative phase diagram has been developed for the pseudo-binary system US- UO_2 . Compositions were prepared at 10 m/o intervals from mixtures of U, UO_2 , and US_2 powders and equilibrated at 1700°C in vacuum. Solidus and liquidus points were determined by observing, through a high-temperature microscope, the melting behavior of small sample particles resting in the "V" of a tungsten wedge ribbon furnace.

The system appears to be simple eutectic, with no indication that the region of liquid immiscibility reported for the U- UO_2 system** extends over to this portion of the U-S-O ternary system. The eutectic occurred at about 45 m/o UO_2 and 1910°C, with the melting points of US and UO_2 determined to be 2435 and 2810°C, respectively. The Clausius-Clapeyron equation for freezing-point depression was applied to the liquidus data to determine the heat of fusion of US and UO_2 . Assuming ideal solutions and no solid solubility, the integrated equation states

$$-\ln N_A = \frac{L_f}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right)$$

where L_f is the heat of fusion of component A, T_0 is the melting point of pure A, and T is the liquidus temperature at composition N_A . The slope of $\ln N_A$ plotted versus $(1/T - 1/T_0)$, where N_A approaches 1 (and the solution approaches ideality), yields L_f . The heat of fusion (and entropy of fusion derived therefrom) thus calculated for US and UO_2 is 20.8 kcal/mole and 7.68 cal/deg-mole, and 16.2 kcal/mole and 5.79 cal/deg-mole, respectively. This L_f value for UO_2 compares favorably with values obtained from other UO_2 eutectic diagrams, while the entropy values are in the ranges expected for these particular structures.

The eutectic occurs at about 150°C lower than would theoretically be expected from reapplication of the Clausius-Clapeyron equation using the calculated heats of fusion. One factor might be that the entropy of

*Riley, B., United Kingdom Atomic Energy Report, AERE-R 4154 (1962).

**Martin, A. E., and Edwards, R. K., J. Phys. Chem. 69, 1788 (1965).

mixing for the liquid solutions is too simply represented by the relation, $\Delta S_m = -R(X_{US} \ln X_{US} + X_{UO_2} \ln X_{UO_2})$. A more complicated relation, such as one assuming mixing of the three atomic species, would yield a higher value for ΔS_m and could account for the seemingly increased stability of the solutions toward the eutectic.

A tentative phase diagram has also been developed for the pseudobinary system UOS- UO_2 . As detailed in the July report, up to 70 m/o UO_2 was soluble in UOS at 1750°C. The liquidus over this solid-solution range dropped from 1975°C, the melting point of UOS, to a minimum of 1900°C at 12 m/o UO_2 , then rose to a maximum of 2350 to 2400°C at about 65 m/o UO_2 . The liquidus over the two-phase, terminal UOS_{SS}- UO_2 , area then rose, seemingly without a break, to 2810°C, the melting point of UO_2 . This suggests that the terminal UOS_{SS} terminates at a peritectic point at about 2350 to 2400°C and 70 m/o UO_2 . This conclusion should be reconfirmed, however, by running a more carefully characterized series of samples in the 60 to 100 m/o UO_2 compositional range.

That the minimum in the solid-solution liquidus is significant is indicated by the fact that two other measured physical properties were also found to peak at the 12 m/o UO_2 composition. A maximum in initial c-axis expansion (a linear contraction in the c-direction occurs with further solution of UO_2) and a minimum in the electrical resistivity were found to occur at this same composition.

Such a minimum in a solid-solution system only occurs when $\Delta H_m^S > \Delta H_m^L > 0$. Both a strain effect (large size difference between O^{-2} and S^{-2} ions) and a chemical effect (electronegativity difference between O and S) probably contribute to the positive heat of mixing for the solid solutions, while the chemical effect probably predominates in the liquid solutions. An area of solid immiscibility is to be expected in such a system, but an attempt to unmix the solid solutions at 1200°C for 48 hr proved unsuccessful.

c. Fundamentals of Corrosion in Liquid Metals (C. A. Youngdahl, J. Y. N. Wang, and S. Greenberg)

(i) Lithium Corrosion Studies at Elevated Temperatures

(a) Corrosion Inhibition of Lithium Attack on Tantalum

Last Reported: ANL-7371, p. 86 (Aug 1967).

When, as previously reported, silicon dissolves in lithium at 1200°C, interaction between silicon and tantalum occurs. A silicide layer, identified as Ta_2Si , forms on tantalum and is corrosion-inhibiting. The growth kinetics of the silicide phase at constant silicon concentration in lithium and constant temperature are being studied.

The solubility of iridium in lithium at elevated temperature has not been found in the literature, but recent work shows that iridium is readily soluble in lithium at 1200°C. In 9 days at this temperature, a solution of 1.5 a/o iridium in lithium forms a layer of intermetallic compound on the surface of a tantalum sample. The layer formed is 50 μ deep and protects the tantalum from penetrating-lithium attack.

The microhardness of the diffusion layer was measured by means of Knoop microhardness tester at a constant load of 50 g. The Knoop microhardness number of the layer was 300 to 1000 higher than that of tantalum matrix. The sharp increase in hardness of the layer was evidently associated with the tantalum there being alloyed with iridium.

A gray band exists between the outer layer and the tantalum matrix, as shown in Fig. III.B.7. An X-ray powder pattern revealed that the surface layer was predominantly TaIr_3 .

500X

Fig. III.B.7. Transverse Section of a Tantalum Specimen Exposed 9 days to 1200°C Lithium-1.5 a/o Iridium

The tantalum and iridium distribution of the layer, as a function of distance across the surface, was analyzed by an electron-microprobe technique. Results showed that there were three phases in the layer. The tantalum concentration in the outermost layer was about 29 w/o Ta, which corresponds approximately to the TaIr_3 phase; the intermediate layer had a concentration of about 52 w/o Ta, which closely approaches the TaIr phase; and the innermost layer, adjacent to the tantalum matrix, had a concentration of about 77 w/o Ta, and is believed to be the Ta_3Ir phase.

The existence of a tetragonal sigma phase (Ta_3Ir) was first identified by Nevitt and Downey* and later confirmed by Knapton.** The parameter of $TaIr_3$ in our finding is $a_0 = 3.88 \text{ \AA}$, which agrees with Dwight and Beck.† The $TaIr$ phase has not yet been confirmed in the literature. An attempt will be made to study the growth kinetics of these compounds to obtain information on the mechanism of interaction and to identify the rate-control process.

2. Chemistry of Irradiated Fuel Materials

a. Development of Analytical Facilities, Microstructure Sampling Techniques, and Analytical Procedures for Analysis of Irradiated Fuels (C. E. Crouthamel)

Last Reported: ANL-7382, pp. 102-103 (Sept 1967).

Construction has been started to convert a former storage area to an analytical facility for the study of the chemistry of irradiated fuels. The instruments to be housed in the facility include a spark-source mass spectrometer, a mass spectrometer for isotope-dilution analyses, and a shielded electron microprobe. Upon completion, the instrument facility will be used for microanalyses of samples prepared in the shielded helium-atmosphere facility now under construction in the Chemistry Division.

A spark-source mass spectrometer (Associated Electronic Industries Model MS-7) is scheduled for shipment on November 1. This spectrometer will be set up temporarily in a laboratory other than the new facility and then moved when the new facility is completed. The instrument will be used for the analysis of irradiated fuel samples and also, in a separate program on fuel-failure detection, for the analysis of sodium samples.

b. Postirradiation Studies of Reactor Fuel (M. D. Adams)

Last Reported: ANL-7371, pp. 86-87 (Aug 1967).

Preliminary examinations of laser-vaporized samples ($\sim 50\text{-}\mu$ diameter) of irradiated UO_2 were previously described (see Progress Report for August 1967, ANL-7371, p. 87). This fuel pin, which was 13% enriched $UO_2^{\dagger\dagger}$ irradiated to 6.8 a/o burnup, has now been examined further. The results are discussed below.

During irradiation, the temperature in the central area of the fuel pin was apparently high enough to effect complete isotopic exchange of

* Nevitt, M. V., and Downey, J. W., J. Metals (1957), Vol. 9, p. 1072.

** Knapton, A. G., J. Less-Common Metals (1960), Vol. 2, p. 113.

† Dwight, A. E., and Beck, P. A., Trans. Met. Soc. AIME (1959), Vol. 215, p. 976.

†† The desired enrichment was achieved by blending fully enriched and depleted UO_2 particles.

the fully enriched and depleted UO_2 . However, outside of this high-temperature area, numerous porous inclusions were observed. Electron-microprobe analyses have shown that these inclusions contain high concentrations of rare-earth fission products. Alpha spectrometric examination of the inclusions showed that they are depleted in ^{239}Pu in comparison to the adjoining matrix. This indicates that the inclusions must have consisted of highly enriched UO_2 particles, apparently the temperature of this area of the fuel pin during irradiation was not high enough to effect isotopic exchange.

3. Thermodynamics of Fuel Materials

a. Total Vapor Pressures and Carbon Potentials in the Ternary System U-C-Pu (P. E. Blackburn and M. Tetenbaum)

Last Reported: ANL-7382, pp. 103-104 (Sept 1967).

A transpiration method using H_2 - CH_4 mixtures as a carrier gas is being developed to measure the total vapor pressure and carbon activity of "monocarbide" compositions in the U-C-Pu system. The carbon activity of the condensed phase is calculated from the equilibrium methane concentration in the gas. Flame-ionization chromatography will probably be used to measure the methane concentration.

The carrier gas will be recycled to minimize plutonium transport outside the glovebox, and to ensure that the gas mixture is completely equilibrated with the condensed carbide. This technique differs from that used in the previous transpiration study on urania, in which a gas of fixed water composition was continuously passed over the sample, whose composition was altered until equilibrium with the gas was attained. To test the recycling concept, hydrogen was recirculated over the two-phase system U- UO_2 in the range 1970 to 2260°K. The composition of the gas mixture reached a reasonably constant $\text{H}_2/\text{H}_2\text{O}$ ratio, which demonstrated the feasibility of the recycling method. The oxygen and uranium activity data obtained from these experiments are discussed elsewhere in this report under "Total Vapor Pressure and Oxygen Potentials in the Ternary System U-Pu-O."

4. Techniques of Fabrication and Testing--Basic Fabricability--Research and Development

a. Development of Nondestructive Testing Techniques (H. Berger)

Last Reported: ANL-7382, p. 104 (Sept 1967).

(i) Determination of Elastic Moduli of High-temperature Materials by Ultrasonics. The elastic moduli of Type 304 stainless steel have been compiled from room temperature to 1300°C. This information was requested by Reactor Engineering on four samples cut from the 2-in. Type 304 stainless steel top plate of EBR-II. Two of the samples were cut

in a longitudinal direction, and the other two in a transverse direction with respect to the rolling direction in fabrication of the 2-in. plate. The compilation included information on Young's modulus, Poisson's ratio, and the shear modulus calculated from the data obtained.

A Type 316 stainless steel sample has been fabricated and data are now being taken to determine the elastic moduli of this material.

(ii) Ultrasonic Instrument and Transducer Development

Last Reported: ANL-7349, pp. 101-102 (June 1967).

The effect of the cone angle (defined as the angle from a line parallel to the front face and perpendicular to the tip) on the rear face of a backing member for ultrasonic transducer probes was studied. A 3-in.-long piece of aluminum rod was used in the study. The cone angle was varied from 0 through 80° by machining in 10° steps. Ultrasonic energy was transmitted into the aluminum specimen at each step by means of a 5-MHz fingertip transducer probe. Polaroid photographs of the resulting oscilloscope presentations were taken. Optimum cone angles in this material occur at 30 and 80°. Further studies on different backing member materials are contemplated.

(iii) Measurement of Thermal Conductivity of Irradiated Fuel as a Function of Burnup and Temperature

Last Reported: ANL-7382, p. 109, (Sept 1967).

Essentially all work on the postirradiation thermal conductivity of EBR-II driver fuel, Types MRC, MR, and SL is complete, and a final report is in preparation.

The present pulse-method setup for thermal-conductivity measurement has limited temperature capabilities. To enable the temperature range to be extended above the present limit of 600°C, a ruby laser is being installed to replace the xenon flash tube.

(iv) Development of a Nonreactor Neutron Source for Radiography

Last Reported: ANL-7382, p. 110 (Sept 1967).

The peak flux of the Am-Cm-Be neutron source-moderator has been determined to be 1.25×10^5 thermal neutrons/cm²-sec; the cadmium ratio (0.5-mm cadmium cover) is 6.7. Both measurements were made with gold foils.

Neutron beams have been brought out of the moderator by displacing water from the beam path. The beam collimator has originated

at the point of maximum thermal-neutron flux. A conical beam, about 2.5 cm in diameter at the source and 7.5 cm in diameter at the output end, has yielded a thermal-neutron intensity of $670 \text{ n/cm}^2\text{-sec}$ at the output end. The gamma intensity at that point is 200 mR/hr . This can be reduced by a factor of three with a 0.25-mm lead filter, but the $n\text{-}\gamma$ ratio is still unfavorable for direct-exposure neutron radiography. More filter studies are planned.

The BeO moderator study is progressing in that a source of coated BeO bricks has been located. Assembly of the moderator should be initiated next month.

b. Basic Fabricability of Nonfissile Metals and Alloys
(R. M. Mayfield)

(i) New Fabrication Concepts--ANL Three-roll Tube Reducer

Last Reported: ANL-7357, p. 11 (July 1967).

Operational tests on the tube-reducer plug feed mechanism have been essentially completed. An 8-in.-long straight cylindrical plug replaces the longer mandrels previously used. As tube reduction proceeds, the tube is moved over the plug in much the same manner as in the "Rockrite" tube-reduction method.

Except for a few required minor alterations, the device works quite well when the same operational parameters are used that have proven successful with the longer mandrels. A 23° rotation of the tube after each stroke produces good results. Feed rate (tube advance/stroke) varies with tube size, total reduction, and the reduction sequence being employed. No lubrication problems have been encountered.

(ii) Fabrication Development of Group VI Metals and Alloys

Last Reported: ANL-7342, pp. 92-94 (May 1967).

A fabricability evaluation of 3M-produced* tube blanks of tungsten, W-25 w/o Re, and W-30 a/o Re-30 a/o Mo is now under way. The 0.375-in.-OD by 0.285-in.-ID tungsten-alloy tube blanks were manufactured by a powder extrusion and sintering technique that yields a 93 to 96%-dense product with smooth surfaces. This method of tube-blank production may serve as an alternative to the presently used "filled-billet" extrusion technique. A pressed-and-sintered sleeve is the starting product for "filled-billet" extrusion, and the final extruded product frequently has extensive surface striations that can hinder subsequent secondary fabrication operations, particularly for unalloyed tungsten.

*Sowman, H. G., et al., Fabrication of Tungsten Alloy Tubing, COO-1518-4, Minnesota Mining and Manufacturing Co., St. Paul, Minn. (Dec. 23, 1966).

Sections of W-25 w/o Re and W-30 a/o Re-30 a/o Mo have been ductile-core and mandrel drawn to 0.265-in. OD by 0.208-in. ID and 0.251-in. OD by 0.192-in. ID, respectively, in accordance with the fabrication parameters previously outlined (see Progress Report for May 1967, ANL-7342, p. 93). No difficulties were experienced. Attempts to fabricate the tungsten tube blanks will get under way in the near future. (Experience indicates that some difficulties can be anticipated.) A metallographic study is planned for all the alloys, both before and after fabrication. Density values will also be determined in both conditions.

5. Engineering Properties of Reactor Materials--Research and Development

a. High-temperature Mechanical Properties of Fuel Oxides (G. D. White)

Last Reported: ANL-7249, pp. 52-53 (Aug 1966).

As an integral part of a study of the determination of the mechanical properties of polycrystalline nonmetallic materials, characterization and control of the test specimens are of prime importance. Shape and distribution of voids, shape and size of grains, and chemical makeup, especially when stoichiometry is a variable, affect the properties of the material. A study of these variables in the preparation of $U_{0.8}Pu_{0.2}O_2$ specimens has been initiated.

Depleted UO_2 powder was mixed with PuO_2 powder by ball milling for 4 hr in alcohol. Specimens pressed without binder were sintered at 1650°C for 4 hr in vacuum, helium, and helium-6 v/o hydrogen, respectively. Table III.B.3 lists data on the specimens for the three different atmospheres.

TABLE III.B.3. Data on $U_{0.8}Pu_{0.2}O_2$ Reacted and Sintered for 4 hr at 1650°C in Vacuum, Helium, and He-6 v/o H_2

Atmosphere	a_0 Å	O:M	
		Ox-Reduc	Chem
Vacuum (10^{-6} mm Hg)	5.459	1.977	1.987
He-6 v/o H_2	5.460	1.972	1.981
Helium	5.457	1.985	1.997

X-ray reflections were sharp with clean $\alpha 1$ - $\alpha 2$ separation at high angles. The specimen heated in He-6 v/o H_2 had slight broadening of the high-angle reflection as compared with the specimens heated in vacuum and helium. The unit-cell parameters in the table were determined by the Nelson-Riley method. The oxygen-to-metal ratios were determined by

oxidizing material in He-2 v/o O_2 for 2 hr at $850^\circ C$, followed by a reduction in 9 v/o CO-CO₂ at the same temperature for 3 hr, and determining stoichiometry from the weight changes. The chemical technique was the inert-gas fusion method, wherein the oxygen content was determined and the stoichiometries were calculated. Metallography has likewise been attempted on these specimens, but the structures were not well delineated. Development of a good etchant for PuO₂ or UO₂-PuO₂ is needed.

C. Engineering Development

1. Development of Master-Slave Manipulator Systems

a. Development of Manipulator Systems (R. C. Goertz)

Last Reported: ANL-7382, p. 111 (Sept 1967).

(i) Electric Master-Slave Manipulator, Mark E4A. The slave servo motors are cooled by having one fan force about 560 cfm of air past the motors. A simple cowling is used to guide the air, and the air is not filtered.

We are now studying designs which would include passing all cooling air or argon through a 95% efficient filter. The present large amount of cooling gas makes filtering difficult. Methods are being studied to reduce the amount of cooling gas needed. Fins are being added to some test motors to provide better heat exchange. So far, only one motor has been tested with fins. In a mockup, one motor required 8 cfm of air with fins and 24 cfm without fins. The motor (Diehl 25-frame) was excited with 150 V, 60 Hz, on each field and the stator copper temperature was held to not exceed 170°C (equivalent conditions for 50-lb load at the slave). The one motor will be tested further in both air and argon. Additional fins have been fabricated so that a complete servo gear drive unit can be tested.

One of the brakes on a servo drive unit has been cycled on and off. At 140,000 Hz, a pin failed. This was modified. At about 330,000 Hz, the modified pin and one of the slides in the solenoid failed. Considerable wear of the pawl and mating gear occurred after 240,000 Hz. Modifications and more tests will be made. We estimate that cycling over a 5-yr period in the manipulator would likely be between 100,000 and 500,000 Hz. We have set the no-failure goal at about 2 MHz.

(ii) Components and Tools

(a) Low-inertia AC Servo Motors for Manipulators. The copper sleeve on this motor was originally silver-soldered only at the ends. It has now been resoldered its full length to ensure better heat transfer from the copper to the iron core. This reduced the rapid drop in torque somewhat, but there still is about 30% drop in torque in about 90 sec after the motor is fully excited from a cold start (room temperature). The copper sleeve temperature at the hot end reaches 160°C in 90 sec. This change in temperature reduces its conductivity to 65% of its value at room temperature. We believe this is the principal reason for the rapid drop in torque. If we hold the current constant (in both phases) instead of holding the voltage constant, the torque increases about 10% in 90 sec. If we hold the current

constant in one phase and the voltage constant in the other, the torque drops about 10% in 90 sec. This indicates that there is a possibility of using impedance in the amplifier and/or fixed field to compensate for the change in torque as a function of heating.

This motor is cooled by forcing a gas through the rotor. We are presently using 11 cfm of air, which requires a pressure of about 27 in. of water. This cooling requires more power than we prefer, and, of course, much additional power will be needed for cooling with argon. More studies will be made on the motor to reduce the cooling problem.

(b) Low-inertia DC Motors for Manipulators. More studies and calculations have been made on shell-type dc motor designs. Our calculations indicate that motors with much lower inertia, lower electrical time constant, and fewer cooling problems can be developed. Commutator wear does, however, present a problem. One method of circumventing this problem is to enclose all the motors and keep a moist gas around them. Brush friction is another problem. Dither, force feedback, or some other method could be used to reduce its effect. We have ordered some permanent magnets, Alnico-9, to be used in experimental dc motors.

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

a. High-temperature Boiling-sodium Experiments (J. V. Tokar)

Last Reported: ANL-7382, pp. 112, 113 (Sept 1967).

(i) Niobium-1% Zirconium Loop. Instrumentation has been installed for continuously recording flow, pressure, and temperature during the initiation of boiling. A boiling run to check the operation of this equipment showed that the system is operating satisfactorily. Several weeks of 24-hr/day operation at 15-70 psia are planned.

b. Heat Transfer to Liquid-metal Heat Exchangers (R. P. Stein)

Last Reported: ANL-7382, pp. 113, 114 (Sept 1967).

(i) Nonsymmetrical-duct Heat Exchangers. Construction of the major portions of the sodium heat exchanger loop is essentially complete. The repaired electrical sodium heaters have been reinstalled in the loop. All thermocouples have been attached at various circumferential and axial positions along the outer wall of the first test section, which is a double-pipe heat exchanger with continuously variable eccentric placement of the inner tube. Simpler test sections with fixed eccentric placement, but with different effective thermal resistance of the circumferential heat-flow paths are being built; two of these are nearly completed. The objective of

the experiments is to obtain data on how eccentric placement and circumferential thermal resistance in both the inner and outer walls affect the overall rates of heat exchange at various operating conditions. The data will be used to test various methods of predicting overall heat-transfer rates in nonsymmetrical-duct heat exchangers.

(ii) Liquid-metal-heated Steam Generators. Design of the loop, with which experiments on simple simulations of liquid-metal-heated steam generators are to be performed, has been completed. Fabrication and purchase of equipment have begun. The experiments will accurately measure actual vaporization rates as influenced by operating conditions. Then the data will be compared with predictions based on current engineering methods of design and with more detailed analyses.

c. Heat Transfer in Liquid-metal-cooled Reactor Channels
(R. P. Stein)

Last Reported: ANL-7357, p. 116 (July 1967).

Studies are concentrating on predictions of forced-convection liquid-metal heat transfer during heat-flux transients; in particular, the applicability of a new "engineering-type" relationship for flux transients is being explored in detail. The use of this relationship, as well as more accurate predictions, requires values of certain coefficients that can be computed from the appropriate eigenvalues and eigenfunctions of the "Fundamental Sturm-Liouville Problem for Heated Ducts." These, in turn, depend on duct geometry and on the relationships used to compute turbulent-velocity and eddy-diffusivity distributions. Because the computation of these quantities occurs very frequently for a variety of studies, and is not generally available for liquid-metal flows, the formulation of a general computer subroutine for eigenvalues and eigenfunctions has been started. In addition to other general features, such as general boundary conditions and geometry, the subroutine will provide for use of velocity and eddy-diffusivity distributions as arbitrary functions from other subroutines. This will allow efficient exploration of the effect of the particular functional representations used for these distributions, an aspect considered especially important with liquid metals.

d. Fog-flow Heat Transfer and Fluid Flow (R. P. Stein)

Last Reported: ANL-7371, p. 91 (Aug 1967).

The initial literature survey has been revised to emphasize aspects immediately related to planning detailed experimental and theoretical research on turbulent-diffusion of fine droplets and droplet deposition rates at duct walls.

3. Liquid-metal Linear Generator

a. Effect of Interfacial Transport Processes on Condensation (J. H. Linehan)

Last Reported: ANL-7371, p. 92 (Aug 1967).

Data taken for turbulent-laminar and turbulent-turbulent conditions of vapor-liquid flow include variation of film subcooling from 150 to approximately 0°F. The data are being reduced.

4. Engineering Mechanics

a. Anisotropic Thermoelasticity (R. A. Valentin)

Last Reported: ANL-7382, p. 114 (Sept 1967).

Figure III.C.1 shows a typical transient-stress component of the quasi-static isotropic thermoelasticity problem that was chosen to illustrate the behavior of the stress field caused by sudden initiation of internal heat generation (see Progress Report for July 1967, ANL-7357, p. 121). Shown is the dimensionless stress component $\sigma_{\theta\theta}$ at the surface ($\rho = 1$) and the centerline ($\rho = 0$) of an infinite, traction-free, circular cylinder cooled at the surface through a temperature-independent boundary conductance specified by the Biot number $m = 0.1$, which results when, at dimensionless time $\tau = 0$, the axial segment between $\xi = \beta$ and $\xi = -\beta$ (where $\beta = 4$) begins uniform internal heat generation while the remainder of the cylinder does not generate heat.

Fig. III.C.1. Typical Transient Circumferential-stress Component of Quasi-static Isotropic Thermoelasticity Problem

Among the more important qualitative properties of the stress field illustrated, the transient stress levels never exceed in magnitude the steady-state values, $\tau = \infty$. This is in marked contrast

to the behavior observed in problems with abrupt time changes in temperature, in which case the thermal stresses may reach several times the

steady-state values. Also, in this case, the circumferential stress never exceeds the plane-strain, steady-state value and, hence, the latter provides a quick, conservative estimate of the maximum circumferential stress levels even when the heat-generation rate has step changes both in time and in the axial-space coordinate. This is also true of plane estimates of the radial stress, but does not hold for the axial stress or for the shear.

b. Core Structural Dynamics Studies (M. W. Wambsganss)

Last Reported: ANL-7371, p. 92 (Aug 1967).

In the experimental study of parallel-flow-induced vibration of cylindrical rods, testing has been resumed with brass tubing instead of the former plastic rods (used to produce easier study by means of exaggerated motion). The brass tubing permits the study of the experimental approach for a rod whose stiffness approaches that of a single fuel rod. Displacement data have been recorded for a tube with clamped end conditions and with flow rates up to the 20 ft/sec provided by a new pump and an acoustic damper to smooth the pump output (see Progress Report for June 1967, ANL-7349, p. 105). The resultant pressure fields (pressure variation with respect to time and position) in the test section have also been recorded. Reduction and analysis of this data are incomplete because of the breakdown of the magnetic tape recorder. The data processed before breakdown do not indicate an obvious trend (as exhibited by the data from the previously tested plastic rods). Further data treatment will determine where the fault lies or if a statistically significant trend does exist.

Because of priorities for vibration analyses of the EBR-II steam generators and superheaters, the rod vibration analysis may be delayed. The rod experimental program continues with the development of the small transducers to be mounted directly to the rods. A 1/2-in. solid stainless steel rod is instrumented with two 0.060-in.-dia piezoelectric transducers. A calibration chamber suitable for calibrating rod-mounted transducers is being built; it will be used for this rod. These transducers represent a first step in obtaining the desired 0.020-in.-dia transducers. Clinch* showed that units of this size were feasible; earlier attempts at assembling 0.020-in. dummy transducers demonstrated that their construction is within our capabilities.

*Clinch, J. M., Study of Vibrations Induced in Thin-walled Pipes by Fluid Flow, IITRI-N6095 (Sept. 24, 1965).

D. Chemistry and Chemical Separations

1. Fluoride Volatility Process

a. Fluorination Chemistry and Procedures (M. J. Steindler)

Last Reported: ANL-7357, pp. 125-127 (July 1967).

(i) Fluorination of UO_2 - PuO_2 -fission-product Pellets--2-in.-dia Fluid-bed Reactor. A 2-in.-dia fluid-bed reactor is being used for developmental studies to establish reaction conditions that result in maximum volatilization of plutonium and minimize residual plutonium in the final fluid bed. Uranium dioxide-plutonium oxide pellets containing non-radioactive fission-product oxides are the feed material. Pellets are first oxidized to U_3O_8 - PuO_2 fines by reaction with oxygen; then fluorination with BrF_5 forms nonvolatile PuF_4 and volatile UF_6 ; in the final step, reaction of PuF_4 with fluorine forms volatile PuF_6 .

Studied during the past month were (1) the behavior of fission-product compounds during processing, (2) the use of a bed containing prefluorinated alumina fines in a scheme to lower residual plutonium concentration in the final alumina bed, and (3) the effect of a short recycle-fluorination sequence on plutonium removal from the alumina bed. The data reported here include additional data from earlier alumina reuse experiments, runs Purse-20, -21, and -22 (see Progress Reports for June and July 1967: ANL-7349, pp. 107, 108, and ANL-7357, pp. 125, 126), and from runs Purse-13 and -15 (see Progress Reports for December 1966 and February 1967: ANL-7286, p. 51, and ANL-7308, pp. 71-73). Results for two new runs, Purse-23 and -24, are also reported.

Additional data have been obtained on ruthenium behavior in the alumina reuse experiments. Before the start of run Purse-22, ^{106}Ru tracer was added to the alumina bed to monitor the distribution of ruthenium during the various processing steps. Most of the ruthenium volatilized during fluorination with BrF_5 at 300°C , and only a small amount volatilized during the subsequent recycle-fluorination step with fluorine at 300 to 550°C .

The behavior of fission products during processing has also been examined for two other runs (Purse-13 and -15). A spark-source mass-spectrometric analysis has been performed* on alumina and sodium fluoride samples from these runs (in which the BrF_5 fluorination step was carried out at either 250 or 350°C). Samples obtained from the alumina bed after each step and from the NaF trap (PuF_6 product collector) after the fluorine step were analyzed for ruthenium, molybdenum, and rhodium. In the spark-source spectrometer analyses, analyses of replicate samples

*The Dow Chemical Co., Rocky Flats Plant, Golden, Colorado.

were found to vary by as much as a factor of three, necessitating that samples be analyzed a number of times. Within this limitation, the spark source data indicate that 70 to 80% of the ruthenium and more than 95% of the molybdenum was volatilized from the fluid bed during the BrF_5 step. During the fluorine step, additional molybdenum was volatilized and was collected at 100°C on NaF ; however, only a small portion of the ruthenium remaining on the alumina bed at the beginning of the fluorine step was fluorinated by fluorine, and the volatile ruthenium species formed was not completely removed from the gas stream by the NaF trap. The spark source analyses for rhodium do not indicate any fluorination of this element; rhodium concentrations in alumina bed samples did not vary significantly during the run.

During the report period, run Purse-23 was performed to evaluate a technique for decreasing plutonium retention in the fines. These are recovered after each run by rapping and brushing the upper section of the reactor (which has a lower temperature during a run than the reaction zone). The scheme was to use a bed containing 10 w/o alumina fines (the remainder of the bed being -40 +170 mesh alumina). It was anticipated that the fines would provide a precoat of the fluid-bed filters before plutonium-bearing solids would collect there and would also provide a vehicle for returning these solids to the reaction zone of the reactor when the filter was blown back.

Prior to run Purse-23, the entire alumina bed was fluorinated with 90 v/o F_2 for 5 hr at 550°C . At the start of this run, ~645 g of UO_2 - PuO_2 -fission-product pellets containing 2.78 g of plutonium was added to an 1100-g bed of alumina containing 0.6 g of CsF , 0.16 g of RbF , and 0.51 g of NpO_2 . After a 4-hr oxidation with 20 v/o O_2 at 450°C , fluorination with 9-11 v/o BrF_5 at 300°C proceeded for 2 hr; the final step was recycle-fluorination with 90 v/o F_2 at 300 - 550°C for 8 hr and at 550°C for 3 hr. During the run, a 31-g agglomerate formed in the upper section of the reactor. The agglomerate contained 0.08 w/o of the plutonium charged. After the bed was removed from the reactor, 13.6 g of fines containing 1.56% of the plutonium charged was collected from the upper section of the reactor by brushing and rapping.

The final dumped bed contained 0.0015 w/o plutonium or 0.67% of the plutonium charged--the lowest residual plutonium level obtained in the experiments with the 2-in.-dia reactor. With additional work, prefluorination of alumina and other processing techniques developed in the 2-in.-dia reactor can probably be used to achieve low plutonium levels in the fluid bed with only a single use of the alumina bed.

Recoveries of neptunium and uranium were determined for each of three runs (Purse-18, -19, and -23) in which the initial beds contained 10% fines. The final beds contained less than the detectable amount of neptunium (0.003 w/o) and less than 0.1% of the uranium charged.

In run Purse-24, a recycle-fluorination sequence of shorter duration was studied. Graphs showing the progress of plutonium fluorination in earlier runs indicated that residual plutonium in the final bed would not be affected by shortening the recycle-fluorination period. Duration of the recycle-fluorination was reduced from 11 hr (normally used) to 6 hr (1 hr each at 300, 350, 400, 450, 500, and 550°C). The final bed from run Purse-24 contained 0.018 w/o U, <0.003 w/o Np, and 0.009 w/o Pu. These results are similar to those for runs Purse-18 and -19, which used the longer recycle-fluorination sequence. Run Purse-24 thus demonstrated that effective plutonium removal from the alumina bed can be obtained using a shortened recycle-fluorination period.

b. Fission-product Fluoride Chemistry (R. L. Jarry)

Last Reported: ANL-7382, pp. 115, 116 (Sept 1967).

(i) Chemistry of Ruthenium. Four laboratory-scale experiments have been conducted in which a mixture of RuO_2 and alumina was contacted first with BrF_5 at 250 or 300°C and then with fluorine at 300-550°C. In the BrF_5 step, the concentrations of BrF_5 in nitrogen for the four runs were 2, 4, 10, and 11 v/o. Since the RuO_2 was tagged with ^{106}Ru , the progress of the reaction could be monitored by measuring the activity in the sample-containing boat, and in a NaF trap and a soda lime trap where volatile ruthenium species were collected. Gamma-count data indicated that ~70% of the RuO_2 reacted in the BrF_5 step and ~85% had reacted after completion of the fluorine step. Neither the higher temperature nor a higher concentration of BrF_5 increased the ultimate conversion of RuO_2 to a volatile species. However, the reaction rate constant increased from 0.00672 min^{-1} at a BrF_5 concentration of 2 v/o to 0.0212 min^{-1} at a BrF_5 concentration of 11 v/o.

c. Neptunium Fluoride Chemistry (L. E. Trevorow)

Last Reported: ANL-7357, p. 127 (July 1967).

Work is being conducted in a flow system to match more closely the conditions under which the NpF_6 -NaF reaction would occur in a process gas stream. The NpF_6 -NaF reaction was carried out by circulating 10 v/o NpF_6 -90 v/o F_2 over a ~0.002 mole sample of solid NaF heated to 250°C and contained in a small nickel vessel. On the basis of weight-change measurements only, the product had a Na/Np atom ratio of 4.

The solid reaction product was then fluorinated at 400°C, using 100% fluorine at a pressure of ~1 atm. A cold trap at -78°C removed the volatile product (presumably NpF_6) from the circulating gas stream. Weight-change data indicated that 80.5% of the neptunium was removed from

the product during a 7.5-hr fluorination. A second experiment was carried out using the same procedure, and weight-change data indicated that 99.8% of the neptunium was removed during a 5.6-hr fluorination.

Additional experiments will be performed to determine better conditions for removing neptunium from the sodium fluoride-neptunium fluoride complex.

d. Development of Decladding Procedures (D. Ramaswami)

Last Reported: ANL-7371, pp. 93, 94 (Aug 1967).

The first set of experiments to evaluate chlorine as an alternative to hydrogen chloride for removing Zircaloy cladding from UO_2 has been completed. An objective of these runs was to determine whether there would be a uranium loss during chlorination. In separate runs, sintered uranium dioxide pellets and chlorine were reacted at 200, 300, 400, 450, 500, 550, and 600°C. The pellets, 0.5 in. in diameter by 0.5 in. high and weighing ~16 g, were exposed to a chlorine flow of 250 ml/min for 3 hr.

In the experiments up to 500°C (see Progress Report for August 1967, ANL-7371, p. 94), surface scaling was observed in the range 400-500°C. In recent runs at 550 and 600°C, the reaction of UO_2 pellets with chlorine resulted in the formation of vapor and liquid products. It is concluded that if the reaction temperature is not permitted to exceed 500°C, the only product of the UO_2 -chlorine reaction will be solid and will not be detrimental to the fluid-bed decladding of Zircaloy-clad UO_2 fuel.

e. Engineering Development of Fluid-bed Fluorination Procedures (J. T. Holmes)

Last Reported: ANL-7371, pp. 95, 96 (Aug 1967).

(i) Process-development Studies for Uranium Dioxide Fuels.

Engineering-scale studies are being performed in the 3-in.-dia fluid-bed reactor facility to determine the effects of important process variables on the fluorination of U_3O_8 by bromine pentafluoride. The general procedure for a run consists of (1) prefluorination of the alumina bed, (2) oxidation of fragmented UO_2 pellets to U_3O_8 fines, and (3) fluorination of the fines with BrF_5 to produce volatile UF_6 .

The effects of six independent variables have been studied in a series of nine engineering-scale experiments: temperature (300 or 225°C), BrF_5 partial pressure (~0.16 or ~0.29 atm), fluidizing-gas velocity (0.53 or 0.89 ft/sec), UO_2 charge (2.2 or 4.4 kg), alumina mesh size (-100 or 48-100), and the presence or absence of (nonradioactive) fission-product

compounds. The UF_6 production rate was increased at (1) the higher temperature, (2) the larger UO_2 charge, (3) the larger BrF_5 partial pressure, and (4) the greater fluidizing-gas velocity. The BrF_5 utilization efficiency was improved (1) at the higher temperature and (2) with the larger UO_2 charge. The use of either 48 to 100 mesh or -100 mesh alumina and the presence of fission-product compounds did not significantly affect the results. Although no run was made combining all the "best" conditions (300°C, 0.29 atm, 0.89 ft/sec, and 4.4 kg UO_2), it is estimated that these conditions would give an average UF_6 production rate of 164 lb $\text{UF}_6/(\text{hr})(\text{ft}^2)$ and an average BrF_5 utilization efficiency of 60% during the time between the start of the BrF_5 step and fluorination of 75% of the uranium in the reactor. These estimated levels are considered highly acceptable for process application.

f. Engineering-scale Development for UO_2 - PuO_2 Fuel
(N. M. Levitz)

Last Reported: ANL-7382, pp. 116-119 (Sept 1967).

(i) Engineering-scale Alpha Facility. Data have been obtained on the uranium concentration in the alumina bed during the BrF_5 equipment test run, run UBr-16 (see Progress Report for August 1967, ANL-7371, p. 94), and are reported here to complete the reporting of data for that run. About 99.8% of the uranium charged was removed during the BrF_5 step and >99.9% by the end of the fluorine step. The BrF_5 equipment appears to be functioning adequately on the basis of this test.

Two of three campaign-type fluorination experiments have been performed to further demonstrate that production, transport, and recovery of PuF_6 on an engineering scale are practical. Each campaign consists of three successive fluorination experiments, followed by a cleanup-fluorination experiment to recover PuF_4 deposited in the lines and equipment. In each fluorination, 200 g of plutonium (as -325 mesh PuF_4) is charged. A single bed consisting of 6500 g of 48-100 mesh prefluorinated alumina was used in a campaign.

In a fluorination run, the PuF_4 charge was added, and the bed was heated to the desired initial temperature (300°C in Campaign 1, 375°C in Campaign 2, and 450°C in Campaign 3). A flow of 90 v/o fluorine was then started, and the temperature was programmed to increase to 550°C in 5 hr (Campaigns 1 and 2) or 3 hr (Campaign 3). The PuF_6 formed was collected in cold traps. At the end of each fluorination, the PuF_6 was transferred in an inert-gas stream to chemical traps, each of which contained 2600 g of NaF. The objectives of these runs were to observe the effect of initial temperature level on the plutonium concentration in the final alumina bed and to evaluate cold-trap efficiency, PuF_6 production rate (i.e., PuF_4 fluorination rate), yield, and material balance.

On the basis of grab samples alone, temperature did not affect plutonium retention in the first two campaigns. At the end of the first campaign (see Progress Report for September 1967, ANL-7382, p. 117), the final dumped bed contained 0.01 w/o plutonium, representing removal of 99.9% of the plutonium; the final dumped bed from the second campaign contained 0.029 w/o plutonium, representing removal of 99.7% of the plutonium. In Campaign 1 the plutonium content of final grab samples agreed with the dumped-bed samples, but in Campaign 2 the final grab-sample analyses were lower than for the dumped-bed samples, which indicated a small holdup of plutonium-containing material in the column. The total column inventory appears to be better assessed from analyses of samples from the dumped bed.

Fluorination rate data were obtained in two ways: (1) from weight-change data, using a scale on which the cold trap is positioned, and (2) by measuring the neutron activity in the cold trap with a neutron count rate meter. The data of both campaigns showed similar trends with time--a period of increasing amount of plutonium in the cold trap during the first 3 hr, and virtually no collection of PuF_6 during the last 2 hr. On this basis, the fluorination run periods were shortened in Campaign 3. Calculation of material balances awaits further analyses.

Sampling and analysis of NaF and alumina samples have been tested analytically to obtain information on the sampling and analytical errors that are important in evaluating the results obtained in material-balance calculations. One ~800-g batch of NaF pellets containing ~1.9 w/o plutonium and ~1.12 w/o uranium, one ~800-g batch of NaF pellets containing ~0.06 w/o plutonium and ~0.047 w/o uranium, and one 6000-g batch of alumina containing ~0.013 w/o plutonium and 0.0038 w/o uranium were used. Ten-gram samples were obtained from each batch by processes involving riffing and grinding. The NaF was ground in a disc mill, riffled to obtain a 10-g sample, then ground to fine powder and analyzed; the alumina was only riffled to give a 10-g sample before it was ground and analyzed. Five samples from each batch were each analyzed for uranium and plutonium, each sample being analyzed three times. The results led to the following conclusions:

(1) The standard deviations of the plutonium analyses (0.5, 1.8, and 1.8%) compare favorably with the standard deviation (2%) expected for the current method of analysis.

(2) A sampling plus analytical error of 4% was detected when NaF samples were analyzed for plutonium. The effects of sampling errors could be reduced by analyzing two samples from a NaF batch rather than one sample.

(3) For the alumina samples, no sampling error can be detected and the present riffing procedure should continue to be used.

2. Compact Pyrochemical Processes

a. Process Chemistry of Molten-salt Systems (I. Johnson)

Last Reported: ANL-7382, p. 119 (Sept 1967).

In the current pyrochemical process for fast-breeder-reactor fuels, liquid-magnesium alloys are contacted with MgCl_2 -rich molten salts. Because magnesium metal is very slightly soluble in molten MgCl_2 , it was of interest to determine the dependence of the solubility of magnesium in MgCl_2 on the magnesium concentration in a liquid alloy. Magnesium metal dissolved in the salt phase used for a salt-transport separation, for example, might lead to an irreversible transfer of magnesium metal between the donor and acceptor alloys. This could affect the rate of plutonium transfer and the separation achieved.

Previous experimental studies of the solubility of magnesium in MgCl_2 have indicated that when magnesium dissolves in molten MgCl_2 , a dimeric species (Mg_2^{++}) is formed (see Progress Report for February 1967, ANL-7308, p. 37). Since these solutions are highly colored, spectroscopic studies may provide more complete information on the nature of the species. Solutions of magnesium in MgCl_2 can be contained in optical absorption cells made from crystalline MgO . Initial experiments have indicated a very strong absorption band beginning at about 6000 Å and becoming too intense to measure by 4750 Å. An attempt will be made to prepare more dilute solutions so that the absorption band can be resolved.

b. Mass Transfer in Salt-Metal Systems (R. D. Pierce)

A major advantage of pyrochemical processes is that high mass-transfer rates are obtained in liquid metal-molten salt systems. The objective of this task is to investigate mass-transfer rates in liquid salt-metal systems of process interest. These investigations fall into two general categories: (a) studies of the mechanisms of mass transfer in liquid metals and salts, and (b) measurements of physical properties of liquid metals and salts that affect the mass-transfer rates of solutes. This work is being carried out as a graduate-student thesis.

An investigation has been initiated to study the transport of solutes between a single fused-salt phase and a single liquid-metal phase. This investigation should provide basic information related to the salt-metal extraction operations utilized in pyrochemical processing. A tilt-pour furnace and supporting equipment have been designed and constructed to study the distribution and diffusivity of cerium in $\text{Cd-Mg/MgCl}_2\text{-NaCl-KCl}$ systems.

Computer programs describing diffusivity measurements using the capillary bath method have been written and run successfully. Determination of the solute concentration profile in a capillary, perhaps through the use of an electron probe analyzer, would be desirable. Comparison of the results from a profile determination with the values calculated from the computer program could be used to test the assumption that the solute is transported by molecular diffusion alone.

A Mg_3Ce (65.8 w/o Ce) alloy that has been prepared in the tilt-pour furnace will be used to add cerium to the system. Salt-metal distribution measurements have begun. These experiments will study the effects of temperature, and magnesium and cerium concentrations in the alloy.

c. Laboratory Process Development (I. Johnson)

Last Reported: ANL-7371, pp. 97, 98 (Aug 1967).

A pyrochemical process is being investigated for ceramic (e.g., oxide or carbide) fast-breeder-reactor fuels clad with stainless steel. The first step in the pyrochemical flowsheet is the decladding of the stainless steel-clad ceramic reactor fuels. One decladding method that appears to be promising is the dissolution in molten zinc of the cladding and the stainless steel structural material of the fuel subassembly. Because reclaiming the solvent zinc for reuse might be desirable, simple methods of separating iron, chromium, and nickel from zinc are being investigated.

A previous experiment (see Progress Report for May 1967, ANL-7342, p. 81) established that aluminum can be used to form the intermetallic compounds, Fe_2Al_5 , CrAl_7 , and NiAl_3 , which are insoluble in zinc. These intermetallics float on the zinc, and can be separated from it by bottom-pouring the zinc away from the intermetallics. The levels of iron, chromium, and nickel were decreased significantly by the addition of aluminum, but the residual aluminum concentration in the zinc was somewhat high, 0.9 w/o. This concentration of aluminum could cause reduction of PuO_2 in a subsequent decladding step with a consequent loss of plutonium to the zinc solvent. The iron, chromium, and nickel removals achieved in the first experiment indicated that by controlling the quantity of aluminum added to the zinc solution, acceptable removals of stainless steel constituents, as well as a zinc product with a lower aluminum content, could be achieved.

An experiment was therefore performed in which 44 g of aluminum, the stoichiometric amount, was added to a zinc solution containing 37 g of iron (6.4 w/o iron) to form Fe_2Al_5 . (Chromium and nickel were omitted in this experiment because they are present in relatively small quantities in stainless steel, and also because the previous experiment had shown that these elements are readily removed by aluminum addition.)

After all the aluminum had been added to the Zn-Fe solution at 800°C, the temperature was lowered to 500°C and a filtered sample of the zinc phase was taken. Analysis of this sample showed that only 0.048 w/o iron and 0.18 w/o aluminum were present. These results indicate that by adding the proper amount of aluminum, iron can be removed effectively from zinc through the formation of an aluminum intermetallic compound, without leaving an undesirably large amount of aluminum in the zinc.

d. Pilot-plant Operations (R. D. Pierce)

Last Reported: ANL-7382, pp. 119-122 (Sept 1967).

A glovebox facility is being constructed to contain integrated process equipment for conducting pyrochemical engineering studies on the processing of mixed uranium and plutonium fast breeder reactor fuels. The processing equipment to be installed in this facility will be designed to handle unirradiated fuel on a 1- to 2-kg uranium scale. The glovebox will be provided with an argon atmosphere to prevent atmospheric contamination of the processing reagents.

The glovebox shell has been installed in the laboratory, and piping for the argon purification system has been completed. Electrical wiring for the box and the purification system are being installed.

e. Materials Testing and Evaluations (R. D. Pierce)

Last Reported: ANL-7382, p. 122 (Sept 1967).

A Zn-5 w/o Mg alloy is being used as an acceptor alloy in the plutonium salt-transport process. Alloys containing high concentrations of zinc can be contained in tungsten, tungsten alloys, and certain ceramics such as alumina. However, these materials are difficult to fabricate, and more easily fabricated materials are being sought for this usage.

Two ductile rhenium alloys, W-25 w/o Re and Mo-50 w/o Re, have been tested as agitator blades in a system consisting of pure zinc and a MgCl_2 -30 m/o NaCl-20 m/o KCl salt mixture at 800°C. In this method of testing, four samples of the material to be exposed are placed on an agitator shaft so that one blade is exposed to the metal phase, one to the metal-salt interface, one to the salt phase, and one to the vapor phase of the metal-salt system in which the material is being tested. The W-Re alloy was tested for 192 hr, and the Mo-Re alloy was tested for 50 hr. Metallographic examination after exposure of the alloys showed no detectable corrosion (limit of detection = 0.01 mm) of the W-Re alloy, and only ~0.02 mm corrosion of the Mo-Re alloy, primarily by surface dissolution. These results indicate that rhenium alloys could be useful for containing high-zinc systems;

the alloys would probably be used only as thin vessel liners or for special applications, such as valves, because of the high cost of rhenium. These alloys will be tested further when more of the materials become available.

Another potential container material, vitreous carbon, has been tested for corrosion resistance to both Zn-Mg-U-halide salt systems and Cu-Mg-U-halide salt systems. Vitreous carbon is a fragile, nonporous, glass-like form of carbon. It has a low density compared with that of graphite and is much less chemically active because of its lack of porosity. Two different sizes of crucibles were obtained for these studies. The smaller crucibles (1.6-in. OD by 1.6 in. high by 0.10-in.-thick wall) were not wet by the metal phases of the systems tested, and displayed only limited wetting by the salt phases. The larger crucibles (3.9-in. OD by 7.1 in. high by 0.10-in.-thick wall) were chosen for more extensive tests of this material. Analyses of failures experienced with the larger crucibles in these experiments indicate that thermal shock was the major factor. An attempt was made to reduce the thermal shock by placing a crucible within a tight-fitting graphite secondary container. This procedure did not prevent the crucible from failing during an experiment.

Although vitreous carbon has desirable chemical properties for a container material, it is not recommended for use in these systems because of its low resistance to physical and thermal shock. No further testing of this material is planned.

3. General Chemistry and Chemical Engineering

a. Identification of Mechanism(s) of Carbon Transport in Sodium Systems (F. A. Cafasso)

Last Reported: ANL-7342, pp. 68, 69 (May 1967).

The Progress Report for May 1967, ANL-7342, p. 68, indicated that a filterable, carbon-bearing species migrated from a ^{14}C -labeled ferritic steel through liquid sodium at 650°C to Type 304 stainless steel. This finding suggested that a particulate species was involved in the transport of carbon through sodium. However, the sodium had been sampled at the end of the experiment after having been cooled, solidified, and remelted, not during the experiment while at temperature. Thus, the filterable carbon might have been in solution during the migration. To check this possibility, similar transport experiments will be performed, but the sodium samples, both filtered and unfiltered, will be taken at temperature. Identification of the ^{14}C -bearing species will also be attempted; the nature of the species will be deduced from analyses of the labeled products in the off-gases, solutions, and residues resulting from aqueous dissolution of the sodium samples.

The following steps have been taken in preparation for these experiments:

(1) The Fe- ^{14}C alloy to be used as the carbon source has been prepared. Autoradiographs indicate a uniform distribution of ^{14}C throughout the surface of sheets of this alloy. Crucibles are being fabricated from this material.

(2) A facility for loading the ^{14}C -labeled crucibles with reactor-grade sodium within a helium-atmosphere glovebox has been assembled. The sodium is dispensed from a heated, Type 304 stainless steel vessel attached to the top of the glovebox. This vessel is large enough (9 liters) so that all the transport experiments currently planned can be performed with the same batch of sodium.

(3) Several vacuum-furnace tubes equipped with special devices for sampling liquid sodium up to $\sim 650^\circ\text{C}$ have been assembled.

(4) Analytical techniques appropriate for the detection of the expected low levels of ^{14}C -bearing species (< 0.17 ppm carbon in sodium) are being examined.

b. Total Vapor Pressure and Oxygen Potentials in the Ternary System U-Pu-O (P. E. Blackburn)

Last Reported: ANL-7249, pp. 41-43 (Aug 1966).

Data on oxygen and uranium activities have been obtained for the two-phase U- UO_2 system. Hydrogen was recirculated over U- UO_2 in the range 1970 to 2260°K until the $\text{H}_2/\text{H}_2\text{O}$ ratio of the gas mixture became reasonably constant. The water pressures, measured with a Beckmann hygrometer, were taken to be equilibrium pressures and were converted to partial-molar free energies of oxygen, $\Delta\bar{G}(\text{O}_2)$, using the JANAF tables for H_2O .*

The partial molar free energy of uranium, $\Delta\bar{G}(\text{U})$, was obtained from the calculated $\Delta\bar{G}(\text{O}_2)$ values and literature values** for the free energy of formation of UO_2 [$\Delta\text{Gf}^\circ(\text{UO}_2)$]. The equation for this calculation,

$$\Delta\bar{G}(\text{U}) = -\Delta\text{Gf}^\circ(\text{UO}_2) - \Delta\bar{G}(\text{O}_2) + xRT, \quad (1)$$

was derived with the aid of the Duhem-Margules equation for the single-phase urania region. The term x expresses the composition of urania at the hypostoichiometric phase boundary, as represented in the formula UO_{2-x} .

*JANAF Tables, Dow Chemical Co., Midland, Michigan (March 31, 1961).

**Schick, H. L., et al., Thermodynamics of Certain Refractory Compounds, Fourth Quarterly Progress Report, AVCO RAD-SR-63-105 (June 15, 1963).

The mean value of $\Delta\bar{G}(U)$, computed from 11 experiments, was $+0.5 \pm 3.3$ kcal, which corresponds to a uranium activity of $0.9^{+0.1}_{-0.4}$. This value is in accord with that of Drowart and co-workers,* whose measurements of uranium pressures over uranium and over U- UO_2 indicate a uranium activity >0.6 . A much lower activity, approximately 0.1, has been proposed by Thorn and co-workers.** They suggest that a large decrease in uranium activity may occur when uranium is saturated with oxygen, and that this accounts for the much lower uranium vapor pressures they observe over U- UO_2 as compared with those over pure uranium.

This study fills the last gap in the activity measurements for the U-O system. Earlier, oxygen partial pressures over hypostoichiometric urania, UO_{2-x} , were determined in this laboratory.† Other studies for the range UO_2 - U_3O_8 have been summarized by Hagemark.†† Experimentally determined oxygen pressures for the range U_3O_8 - UO_3 are given by Katz and Rabinowitch.‡

c. Vapor-species Partial Pressures in the Ternary System U-Pu-O (P. E. Blackburn, J. E. Battles)

Last Reported: ANL-7382, pp. 103, 104 (Sept 1967).

Mass-spectrometric effusion studies on the evaporative behavior of the U-Pu-O system have been initiated. The binary Pu-O system is being investigated first since no mass-spectrometric work has been done to establish the relationships between the Pu, PuO, and PuO₂ gaseous species and the condensed plutonia phase.

Plutonium dioxide contained in tungsten or rhenium Knudsen cells was vaporized at 1700 to 2100°C into a Bendix TOF mass spectrometer. Reduction of the PuO₂ by both metals was observed by the presence of tungsten and rhenium oxide species in the vapor. The tungsten presented a more reducing environment than the rhenium under similar experimental conditions, as indicated by the greater intensity and longer duration of the tungsten oxide than rhenium oxide vapor species. Examination of the residues showed that considerable melting may have occurred. Also, a bright, metallic-appearing, uniform deposit formed on the inner surface of the lids of both the tungsten and rhenium cells.

*Pattoret, A., Smoes, S., and Drowart, J., Mass Spectrometry on the Uranium-Oxygen and Uranium-Carbon Systems, Thermodynamics, Vol. I, p. 377, IAEA, Vienna (1966).

**Ackermann, R. J., Rauh, E. G., and Thorn, R. J., J. Chem. Phys. 42, 2630 (1965).

†Tetenbaum, M., and Hunt, P. D., The Total Vapor Pressure of Uranium-bearing Species and the Partial Pressure of Oxygen over Urania, ANL-7225, p. 137 (Nov 1966).

††Hagemark, K., and Broli, M., J. Inorg. Nucl. Chem. 28, 2837 (1966).

‡Katz, J. J., and Rabinowitch, E., The Chemistry of Uranium, National Nuclear Energy Series, Div. VII, Vol. 5, p. 256, Dover Publications, Inc., New York (1951).

After the interaction between the PuO_2 and rhenium cell had apparently ceased (i.e., ion currents were constant with time at temperature, and rhenium oxide species were no longer observable), the vapor phase consisted of PuO , PuO_2 , and Pu ; roughly in the ratio 2.5/1/0.04, and an undetermined amount of oxygen. (The same species and relative concentrations were found with the tungsten cell, except that $\text{WO}_2(\text{g})$ was still observed after 20 hr at 2023°C .)

Analysis of the residue in the rhenium cell gave an O/Pu atom ratio of 1.86 ± 0.01 for the (apparently) congruently vaporizing material. If this composition is ascribed to the midrange temperature of the experiment (1900°C), then there is good agreement between this value and those obtained from effusion experiments with tungsten cells by Ackermann *et al.** (O/Pu = 1.83) and Ohse and Ciani** (O/Pu = 1.85). Furthermore, the effective heat of vaporization, considering only the major vapor species PuO and PuO_2 , was 135.1 kcal/mole, which is comparable with the results obtained by Ackermann *et al.** with tungsten and rhenium cells (133.8 kcal/mole) and by Mulford and Lamar† with tungsten cells (133.9 kcal/mole).

The residues and deposits obtained in our experiments are being examined by X-ray diffraction, metallographic, and electron-microprobe methods. If rhenium, as well as tungsten, appears to be an unsuitable container material for studying the high-temperature equilibria in the Pu-O system, other materials (e.g., iridium) will be tested.

d. Preparation of Nuclear Materials (P. A. Nelson)

The main objectives of this task are (1) to develop methods for preparing fast-reactor oxide fuels from the hexafluoride products of the fluoride volatility process, and (2) to develop improved methods of converting such oxides or mixed oxides from any source to fast-reactor carbide fuel. At a later date, methods of preparing advanced reactor fuels such as $(\text{U,Pu})(\text{C,N})$ and $(\text{U,Pu})(\text{C,S})$ from the oxides may be undertaken if these materials are needed.

Currently, a fluidized-bed process is being developed to convert UO_2 to UC. This process is also expected to be applicable for the preparation of $(\text{U,Pu})\text{C}$. The preparation of UC and $(\text{U,Pu})\text{C}$ from oxide feed materials is attractive because oxides are less expensive than the metallic feed material required for the methane-metal fluidized-bed processes also under development at ANL.

*Ackermann, R. J., Faircloth, R. L., and Rand, M. H., *J. Phys. Chem.* **70**, 3698 (1966).

**Ohse, R. W., and Ciani, V., presented at Third International Symposium on High Temperature Technology, Asilomar, California (Sept. 17-20, 1967).

†Mulford, R. N. R., and Lamar, L. E., in *Plutonium 1960*, p. 411, E. Grisen, W. B. H. Lord, and R. D. Fowler (eds.), Cleaver-Hume Press, Ltd., London (1961).

A factor that must be considered in the conversion of mixed uranium-plutonium oxides to carbides is the possibility of plutonium volatilization. In the standard carbothermal reduction technique using static beds, in which $(U,Pu)O_2$ and carbon are reacted under vacuum at 1500 to 2000°C, plutonium volatilization might be a serious problem. This problem is especially likely for large batch sizes where temperatures near the top of this range are required to obtain efficient removal of the carbon monoxide reaction product. However, plutonium volatility might not be a problem in a fluidized bed because (1) the required temperature may be lower than in the static-bed conversion as a result of more efficient carbon monoxide removal, and (2) the plutonium that vaporizes would be expected to condense on elutriated fines in the cooler upper regions of the reactor and be returned to the bed with the fines.

A fluidized-bed reactor for conversion of UO_2 to UC was constructed for operation at temperatures up to 2000°C and pressures of full vacuum to 10 atm. The associated instrumentation and a system for purifying and recycling the off-gas from the reactor are currently being installed.

Methods of adding carbon to the fluidized bed are being investigated. One attractive method is the use of a methane-hydrogen fluidizing gas. Thermodynamic calculations indicate that at high pressures (10 atm) and high hydrogen concentrations (~98 to 99%), carbon deposition can be avoided under conditions that would support a reasonably high rate of reaction.

Another method of carbon addition being studied is that of addition of graphite to the UO_2 feed material. To determine the feasibility of this method of carbon addition, 500 g of UO_2 -carbon particles were prepared. Graphite, UO_2 , and polyvinyl alcohol were ground together in a ball mill for about 20 hr. After ball-milling, the mixture was dampened with distilled water, and then small agglomerated particles were produced by forcing the material through a 20-mesh sieve. The particles thus formed had a log-normal size distribution with a volume median diameter of 640 μ and a geometric standard deviation of 1.33. The particle density was 4.77 g/cc.

A high fluidizing-gas velocity would be desirable with either method of carbon addition for efficient removal of the carbon monoxide reaction product. Therefore, fluidization experiments were conducted in a 2.75-in.-dia clear plastic column at room temperature to determine the maximum gas velocity both for high-density (9.97 g/cc, 91% of theoretical) UO_2 particles considered suitable for reaction with methane-hydrogen fluidizing gas and for the UO_2 -carbon particles described above. The high-density UO_2 particles had a volume median diameter of 183 μ with a geometric standard deviation of 1.165. The minimum fluidizing velocity for

these particles was 0.3 ft/sec (superficial), whereas the maximum fluidizing velocity (the velocity at which fines began to leave the bed) was 3.25 ft/sec at a bed expansion of about 280% of the initial height. On the other hand, the larger UO_2 -carbon particles that were prepared as described in the preceding paragraph began to fluidize at 2 ft/sec and expanded to 350% of the initial bed height at the maximum practical fluidizing velocity of 5 ft/sec.

Although the measurements were made with helium at room temperature, they are applicable for the contemplated operating conditions of hydrogen-gas flow at 1400 to 1800°C. The maximum fluidizing velocities obtained are expected to be sufficient for the rate of carbon monoxide removal required for full-scale (20 to 100 kg/batch) production of UC or (U,Pu)C in 10-hr runs.

As stated earlier, installation of the high-temperature fluidized-bed reactor for oxide conversion is nearing completion. Conversion of 0.5 to 1.0-kg batches of UO_2 to UC in this equipment should begin soon.

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Synthesis of Uranium Monoarsenide (UAs)

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Applications of Neutron Radiography

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Strong Absorption and Direct Effects in Compound Reactions

P. A. Moldauer

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The Influence of Axial Heat Diffusion in Liquid-Metal-Cooled Ducts
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Development of Fluoride Volatility Methods for Reprocessing Reactor
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*University of Illinois.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)--Research and Development

1. Fuel and Core Development

a. Core Structure Development (W. J. Kann)

Last Reported: ANL-7382, pp. 126-131 (Sept 1967).

A contract for stress analysis of core structures has been established with Franklin Institute Research Laboratories (FIRL). The requirements for the various tasks have been reviewed. FIRL supplied a schedule indicating that all work will be completed by the end of June 1968. Additional information required by FIRL is being forwarded as it becomes available.

Fabrication of the fixture and liner is 50% complete for the beam-tube liner seal test. Preliminary drawings of the vertical experimental facilities liner-seal-test fixture have been completed and are being reviewed. These fixtures will be used to test the quality and effectiveness of the seals between the liners and the permanent beryllium reflector.

2. Component Development

a. Beam Tubes (A. R. Jamrog)

Last Reported: ANL-7382, pp. 131-137 (Sept 1967).

The prototype beam-tube adapters were inspected, accepted, and received. The facility for testing the prototype beam tubes is being assembled.

An alternate connector for the through beam tube has been designed and fabricated. The new design eliminates the reduction of the inside diameter of the beam tube at the connector end, which the experimenters found unacceptable. To verify design characteristics, the connector assembly will be subjected to operating and load tests.

3. Instrumentation and Control (J. H. Tessier)

Last Reported: ANL-7371, pp. 103-107 (Aug 1967).

a. Investigation of Power-excursion Models. Survey calculations were made to compare analytical models for predicting the power-excursion behavior of the AARR. The models compared (an analog computer program and the CHIC-KIN* digital code developed at Westinghouse) were adjusted

*Redfield, J. A., CHIC-KIN--A FORTRAN Program for Intermediate and Fast Transients in a Water-Moderated Reactor, WAPD-TM-479 (Jan 1965).

for these studies. The essential differences between them were reduced to the treatment of transient thermal and hydrodynamics processes.

The analog model employed a coarse spatial representation of core heat-transfer conditions with no provision for including the effects of heat load upon coolant hydrodynamics. In contrast, the CHIC-KIN code incorporated a detailed thermal model and also had coupling between heat load and coolant flow through use of the momentum-integral model.* Thus, the analog program uses a constant flow model, whereas CHIC-KIN calculates the influence of heat load upon flow in a continuous manner. This difference in the models resulted in significant differences in calculated results.

Simulated reactor conditions used as a basis for these calculations were:

- (1) The reactor initially in steady state, with normal plant conditions of fuel pressure and coolant flow rate at powers of 100 and 10 MW.
- (2) Neutronic constants, safety system conditions, and fuel-plate oxide scale appropriate to beginning and end of core life.
- (3) Reactor scram initiated by a high flux-to-flow trip or a power-rate trip.
- (4) Power excursion produced by a positive step-function reactivity insertion.

Analog calculations were made to determine the magnitude of reactivity addition necessary to just cause melting of the core hot spots. The hot spots are considered to be small circular regions (5/64-in. diameter), located at the exit of hot channels, and in total represent no more than 5% of core fuel volume.

Threshold values of reactivity determined in this manner are shown in Table IV.A.1.

TABLE IV.A.1. Threshold Conditions for Hot-spot Melting--Analog Model

Case No.	Time in Core Cycle	Initial Reactor Power (MW)	Reactivity Addition ($\Delta k/k$)
1	Beginning of life	100	0.0132
2	Beginning of life	10	0.0215
3	End of life	100	0.0072
4	End of life	10	0.0140

*Meyer, J. E., Hydrodynamic Models for the Treatment of Reactor Thermal Transients, Nucl. Sci. Eng. 10, 269-277 (1961).

In parallel calculations, the reactivity values of Table IV.A.1 were used as input to the CHIC-KIN code using the same assumptions for initial reactor conditions. In each case, the calculations were performed for an average and a hot channel in the core. Table IV.A.2 summarizes the results.

TABLE IV.A.2. Results of CHIC-KIN Calculations--Reactivity Addition of Table IV.A.1

Case No.	Average-channel Melt	Hot-channel Melt
1	Questionable	Yes
2	Yes	Yes
3	No	Yes
4	Questionable	Yes

Where a positive indication of the occurrence of melting is indicated in Table IV.A.2, the result should be interpreted as involving extensive fuel melt in the affected channel as opposed to consideration of only the hot-spot zones. This is due to the occurrence of gross flow excursions and high pressures internal to the channel, produced by rapid density changes in the heated coolant. These pressures are considered sufficient to cause gross channel deformations leading to the likelihood of extensive meltdown. When melting is indicated as questionable in Table IV.A.2, the transient pressure levels are not sufficient to clearly indicate large channel deformation, and further study is needed to resolve this question. However, the CHIC-KIN calculations were designed to minimize transient pressures and flow perturbations by suppressing the program feature of producing steam voids, thereby greatly reducing fluid acceleration effects.

In summary, the foregoing results illustrate that the analog model provides an unduly optimistic prediction of the tolerance of the reactor for hypothetical, rapid-reactivity additions. Future calculations of the power-excursion behavior of the AARR should employ a model, such as CHIC-KIN, which treats the coupling of thermal and hydrodynamic phenomena.

b. Transient-stress Analysis. Preliminary to the evaluation of transient thermomechanical effects in the AARR core, the involute fuel plate under static loading conditions is being studied. Equations for a homogeneous plate were derived, and a corresponding computer program for constant transverse pressure was written.

Variables such as change of plate properties, temperature gradients, and transient loading in general have been neglected. Their

superposition will be considered after the soundness of simpler solutions has been ensured.

Solutions for all boundary conditions are obtained from explicit expressions for transverse and radial displacements, radial and shear loads, and bending moments in terms of six constants of integration. Evaluation of these unknowns involves the selection of the proper boundary conditions and the solution of six simultaneous algebraic equations.

The steady-state solution of plate deflections for pinned and fixed edges has been obtained. Another boundary condition, where the inside edge of the plate is fixed and the outside edge retains its initial slope after deformation, but is permitted to rotate along a circular arc, is also under consideration as best simulating the actual condition of the fuel plates in the core. There is some doubt regarding the preliminary numerical solution for this case.

In the area of transient thermal-stress analysis, the effect of mechanical-thermal coupling and inertia has been considered. Apparently, it will be adequate to consider the effect of inertia and neglect that of coupling; if inertia is found to be negligible, then mechanical-thermal coupling should also be small.

Solutions of thermal-stress problems including the above effects indicate that inertia effect becomes important only when massive bodies are involved. Neglecting mechanical-thermal coupling and inertia appears to be appropriate in considering the fuel plates. Thus, the transient thermal-stress analysis of the fuel plate can be decoupled from the transient thermal analysis.

Because computer programs for three-dimensional transient heat transfer are available, the output of such calculations will be used as input to the stress-analysis problem. To this end, a two-dimensional variation should be considered as the first step. The three-dimensional variation may be formulated later if it is appropriate.

4. Planning and Administration

a. Project Definition and Safety Analysis (M. Levenson)

Last Reported: ANL-7371, p. 100 (Aug 1967).

The first meeting with the AARR subcommittee of the AEC Advisory Committee on Reactor Safeguards (ACRS) was held on September 29, 1967; on October 6, 1967, a meeting was held with the full ACRS. The committee advised that it would draft a letter to the AEC indicating that the Preliminary Safety Analysis Report (PSAR) was acceptable and that construction

could proceed with the conditions that: (1) The ACRS wishes to be informed about certain items when their design or construction details become available, and (2) the seismic-design criteria for the facility should be agreed upon by ANL and the AEC Division of Reactor Licensing (DRL) before any concrete is poured. There had been some disagreement concerning what criteria would be acceptable. After the October ACRS meeting, ANL submitted new proposed seismic-design criteria for DRL approval and is awaiting comments or concurrence.

5. Physics Experiments and Analysis (D. H. Shaftman)

Last Reported: ANL-7382, pp. 137-141 (Sept 1967).

Multigroup diffusion-theory calculations have been made to study reactor-physics effects of a possible increase in the diameter of cooling holes in the permanent beryllium reflector of the AARR. The change being considered, namely, an increase from 1/8- to 3/16-in. diameter, is intended to provide a greater margin of safety against transient voiding of coolant in the beryllium (e.g., in the event of an electrical outage which resulted in a reduction of coolant flow) and to ease the problem of fabricating the permanent beryllium reflector.

Most of the water volume in the various azimuthal sectors of the permanent beryllium reflector is directly associated with facilities of the experiments. Thus, an increase of as much as 50% in coolant-hole diameter would mean a 125% increase in coolant-hole volume but less than a 20% increase in the local average water content in each macroscopic sector of the permanent beryllium reflector. In some zones of the permanent beryllium reflector, the increase in water volume is only ~5% of the present design water volume.

For such an increase in coolant-hole diameter, there would be an insignificant reduction (~0.04%) in the available excess reactivity. The calculated thermal-neutron flux at the tip of the blind beam tubes is reduced by less than 1%. Correspondingly, the users' fluxes would probably be reduced very slightly. The thermal-neutron flux in the center of each through tube is reduced by less than 5%--possibly by only ~3%. Two cases have been analyzed for the perturbation of the thermal-neutron flux in the principal sector of the beryllium reflector containing vertical irradiation facilities. In one case, the target volume was assumed to be filled with aluminum. In the second case, additional neutron poisoning, corresponding to actual target material, was included. In each case, an increase in coolant-hole diameter in the permanent beryllium, from 1/8 to 3/16 in., caused small reductions in the calculated thermal-neutron flux; maximum reduction was ~4%.

V: NUCLEAR SAFETY

A: Other Reactor Kinetics

1. Accident Analysis and Safety Evaluation

a. Accident Mechanics (G. J. Fischer)

Last Reported: ANL-7342, p. 112 (May 1967).

As described in our last report on this subject, the formation of a one-dimensional reactor-dynamics calculational model was completed by coupling heat transfer plus fuel expansion, two-phase coolant flow, and point kinetics modules. These modules are called as needed by a main (or control and decision) module. The modules of this formulation are fully compatible with the ARC system,* but they have also been linked into a computer program independent of ARC. The version of the computer code currently in use was written in FORTRAN IV for use on both the CDC-3600 and the IBM-360 Model 50/75 computers.

Since the last progress report on this work was written, several improvements have been made in the method of calculating the reactivity feedback effects. The Doppler feedback reactivity is based on the volume average fuel temperature over a fuel pin segment and is weighted using the square of the axial core power distribution. The Doppler feedback reactivity is also adjusted for changes in the local sodium density (or void as the case may be) by interpolating between Doppler reactivity coefficients for a non-voided and a fully voided core. The probable noncoherence of boiling initiation across the radius of the core can now be represented as comprised of four different groups of fuel subassemblies which start boiling at different times. Specifying three input parameters permits the successive triggering of the reactivity due to boiling in four core regions to be computed. For a fuel-management procedure that replaces a fraction of the fuel at any radius at each refueling, varying power density due to fissile fuel burnup appropriate to the internal breeding ratio of the core can become a source of non-coherence of voiding. This accident model can also be described by this program addition. Extensions of the capability of the code are under development.

The capability of the code in its present form can be demonstrated by a sample accident calculation for a representative 1000-MWe fast reactor. The reference core for the calculations presented here is a 4900-liter $\text{UO}_2\text{-PuO}_2$ system having a fertile-to-fissile ratio of 7.2. The core height and diameter are 100 and 250 cm, respectively, and the upper and lower axial blankets are 30 cm thick. The core composition and

* Toppel, B. J., The Argonne Reactor Computation System, ARC, ANL-7332 (to be published).

the physical parameters are summarized in Tables V.A.1 and V.A.2, respectively. A perturbation calculation using the MACH-1 code was used to obtain space-dependent reactivity coefficients. The value of reactivity versus region of core voided (across the entire core radius) thus obtained (Table V.A.3) is used, through a table look-up and interpolation routine, in calculating the sodium-reactivity feedback.

TABLE V.A.1. Composition of Reference Core

Volume Fractions		Isotopic Distribution	
Sodium	0.50	^{239}Pu	0.70
Fuel	0.34	^{240}Pu	0.25
Stainless Steel	0.16	^{241}Pu	0.05

TABLE V.A.2. Parameters for Reference Oxide Reactor

Core length, L	100 cm
Blanket thickness, R	30 cm
Fuel-rod diameter	0.650 cm
Fuel density. Fuel was gas bonded. Thickness of gas bond was 0.010 cm at full operating power, before start of transient.	10.8 g/cc
Coolant volume fraction	0.50
Heat-generation factor in blanket	0.10
Coolant inlet temp	500°C
Coolant outlet temp at $t = 0$	625°C
Coolant mass velocity at $t = 0$	700 g/cm ² sec (8.42 m/sec)
Peak fuel heat generation at $t = 0$	300 cal/cm ³ sec (148 kW/kg)
Channel exit plenum pressure, P_{ex}	3.0 atm

TABLE V.A.3. Sodium-reactivity Coefficients from MACH-1 Calculation

Channel Segment		$\delta k_{\text{Na}}(Z)$
Core	$Z = 0$ (midplane) to 10 cm	$+3.49 \times 10^{-3}$
	10-20 cm	$+2.99 \times 10^{-3}$
	20-30 cm	$+2.07 \times 10^{-3}$
	30-40 cm	$+9.22 \times 10^{-4}$
	40-50 cm	-2.21×10^{-4}
Blanket	50-60 cm	-5.55×10^{-4}
	60-70 cm	-3.02×10^{-4}
	70-80 cm	-1.75×10^{-4}
Reactivity for complete voiding of core = \$5.14		
$\beta_{\text{eff}} = 3.2 \times 10^{-3}$		
$\ell = 3.9 \times 10^{-7}$ sec		

Some of the results of a \$1/sec reactivity transient for the oxide core described above are summarized in Figs. V.A.1-5. The figures are produced directly by a graph-drawing routine incorporated in the code. The time at which sensible boiling is initiated is t_b .

For purposes of comparison, another calculation was run using exactly the same parameters as for the transient summarized above, except the \$1/sec input reactivity ramp was cut off after 1 sec. In this problem, the coolant did not reach the boiling point and the transient eventually leveled out at a power about 3.3 times the initial steady-state power. At this new steady state, the maximum fuel centerline temperature was 4800°C and the coolant outlet temperature was 891°C.

Fig. V.A.1. Temperature Profiles of Coolant and Cladding at Various Times

Fig. V.A.2. Vapor-void-fraction Profiles at Various Times

Fig. V.A.3. Fuel-centerline Temperatures at Various Times

Fig. V.A.4. Reactor Power Level vs. Time

Fig. V.A.5. Feedback Reactivities vs. Time

2. Coolant Dynamics

a. Critical Flow (H. K. Fauske)

Last Reported: ANL-7382, pp. 148-149 (Sept 1967).

(i) Sodium Tests. The first phase of the experiments using sodium as working fluid has been completed. Measurements included stagnation temperatures from 1200 to 1600°F, critical pressures from 1 to 6 psia, and maximum expulsion rates up to 1000 lb/sec-ft². These are believed to be the first extensive data on flashing flow of sodium.

To facilitate a direct comparison between sodium and water, an identical subatmospheric water loop was built and the test section used for sodium was employed. Direct comparison between the two sets of data suggests that correlations developed for steam-water can be extrapolated to metallic fluids (sodium).*

The test section to be used in the second phase of the sodium experiment has been completed (ID = 1/8 in. and length-to-diameter ratio ≈ 60). With this test section, detailed axial-temperature as well as pressure gradient will be measured to determine the degree of nonequilibrium existing in flashing liquid-metal systems.

(ii) Sonic Velocity. Preliminary velocity-of-sound data have been obtained in air-water mixtures for void fractions between 1 and 15% and pressures ranging from 25 to 45 psia. The data obtained show good agreement with other studies.**

Construction is continuing on the high-pressure facility for measuring the velocity of sound.

b. Coolant Dynamics (R. M. Singer)

Last Reported: ANL-7382, pp. 149-150 (Sept 1967).

(i) Superheat. The sodium superheat experiment is proceeding with a lower sodium head and higher heating rate.

Data obtained thus far show that system pressure, heat flux, and the pressure-temperature history significantly affect incipient boiling superheats in sodium.†

*Fauske, H. K., Quinn, D. J., and Jeans, W. C., Sodium Flashing Experiment, to be presented at the ANS Winter Meeting, November 5-9, 1967, Chicago, Illinois.

**Karplus, H. B., The Velocity of Sound in a Liquid Containing Gas Bubbles, Armour Research Foundation Report, COO-248 (June 1958).

†Holtz, R. E., and Singer, R. M., On the Superheating of Sodium at Low Heat Fluxes, ANL-7383 (to be published).

A series of experiments was conducted in which the system was pressurized with purified argon, then the pressure reduced to some low value before applying the heat flux to illustrate the effect of pressure history on superheats. Reasonable agreement between theory and experiment was achieved. The effect of pressure history may account for some of the disagreement among data reported by various experimenters.

c. Convective Instability (R. P. Heinisch)

Last Reported: ANL-7382, p. 150 (Sept 1967).

The test section has been assembled, and 28 data runs have been obtained. Different positions (x and y) are being studied along with the parameter of heat flux. The thermocouples welded to the wall read within 1.5°F of those butted against the same wall at an equal horizontal position. However, this error is not tolerable when measuring a 5-15°F temperature difference between the wall and the fluid.

The data were run on the IBM-360 computer and are being reduced immediately.

Although theory and data have not been compared, the trends of the theory have been observed in the raw and the reduced data.

d. Electron-bombardment-heater (EBH) Test Facility
(R. D. Carlson)

Last Reported: ANL-7371, p. 120 (Aug 1967).

After the test vessel was outgassed, the first EBH test section was operated 20 hr at low power with no indication of mechanical difficulty.

e. Core Component Dynamics (M. W. Wambsganss)

Last Reported: ANL-7382, pp. 150-152 (Sept 1967).

Some preliminary simple models were studied in the investigation of the plastic deformation of cylindrical shells under impulsive loads. The usual procedures of limit analysis were used in conjunction with a simplified yield condition for a rigid, perfectly plastic material to determine characteristic deformation lengths, dependent only on shell geometry, for the statically and dynamically applied ring loads. The regions of permanent deformation for the analogous band-load problems depended also on the length of the band of load application and, for the dynamic case, on the maximum load for some combinations of parameters. The prediction of maximum displacements and energy dissipation for the dynamic-load cases requires the numerical integration of sets of nonlinear differential equations.

3. Fuel Meltdown Studies with TREAT

a. Meltdown Studies with Metallic Fuels (C. E. Dickerman)

Last Reported: ANL-7371, pp. 108-110 (Aug 1967).

TREAT Transient 1050 was a transient experiment, involving fuel failure and the movement of fuel and coolant, conducted using a small cluster of seven EBR-II fuel pins, with the loop pump shut off. Transient instrument data have been reported.* Although the details of the instrumentation readings were generally consistent, one large discrepancy emerged from study of the data: Integration of the flowmeter curve showed a total expulsion of liquid metal that would have resulted in a void extending somewhat in excess of 40 cm below the bottom of the test section. (Hence the void would extend about 35-40 cm below the flowmeter.) Inspection of the loop showed that the loop had been breached in the thin-walled pump-tube section, and some sodium had leaked into the sealed secondary containment around the loop. Accordingly, the loop was sectioned to check the condition of the loop body below the test section. Solidified fuel alloy was found, extending to approximately 45 cm from the test-section inlet. The inlet pressure transducer port was filled with cast fuel. Extensive alloying had taken place between the fuel and the steel pump tube. Although the shape of the opening in the tube suggested failure under tension, eutectic formation with the fuel could have weakened the wall.

Thus a significant portion of the flowmeter output from Transient 1050 was probably due to movement of molten uranium alloy. The transient gives a direct experimental demonstration of extensive secondary postfailure fuel movements and the possibility of trapping sodium in "pockets" of molten fuel. Because of the extensive voiding, the 16-atm pressure spike recorded by the pressure transducer may have not been an accurate recording of the actual pressure spike; also, the point of origin of the spike probably was below the test section.

b. Development of Experimental Methods (C. E. Dickerman)

A complete shipping package has been designed for transporting the Mark-II integral TREAT sodium loop** to and from the TREAT reactor in Idaho. The packaging will also accommodate the new Mark-II TREAT transparent capsule.† Shipping components include:

* Reactor Development Program Progress Report for February 1967, ANL-7308, p. 87.

** Reactor Development Program Progress Report for March 1967, ANL-7317, p. 106 (1967).

† Reactor Development Program Progress Report for July 1967, ANL-7357, p. 130 (1967).

(1) A 2-R container, which is a 12-in.-OD cylindrical container with bolted flexitallic gasketed flanged closure, into which the experimental loop is sealed. This item fits inside:

(2) A shipping container, which is a 22-in.-OD cylindrical container, also with a flexitallic gasketed flange. This container provides the sealed, structural protection for the 2-R assembly. It exists in two versions:

(a) A "low-activity" design, suitable for transporting samples that have only the activity induced by TREAT exposure. Structurally, this version is essentially identical with the:

(b) "Shielded" design, which is to be used for transporting previously irradiated samples. The "low-activity" container is designed to be modified into the shielded version by insertion of a 3-in.-thick depleted uranium annular shield. During shipment, the assembled package is held on:

(3) A shipping skid.

All components have been designed to meet regulations on shipping and hypothetical accident conditions.

c. Production of Test Elements for Fuel Meltdown Studies with TREAT

Last Reported: ANL-7349, pp. 124-125 (June 1967).

Nineteen elements similar to the EBR-II TREAT elements reported in the Progress Report for June 1967 (ANL-7349) were made for fuel-meltdown studies in TREAT. Each element contained an 11-in.-long column of solid-solution, uranium-plutonium oxide pellets enclosed in a jacket of AISI Type 304L stainless steel. The pellet diameters were 0.245 to 0.247-in. OD, and the jacket tubes had a nominal ID of 0.250 in. with a 0.020-in.-thick wall. Depleted uranium dioxide pellets, used as thermal insulators, and tantalum discs, used as test markers, were loaded at each end of the fuel column. The fuel columns are maintained against the bottom end cap by coil springs that maintain a void space atop the fuel column of approximately 5 in.

The jacket tubing, which was procured from Central Shops, is from a lot that was tested and approved for use in EBR-II. The tubing was cut to a length of 17.250 in. and inspected for inside dimensions with an air gauge. A lower connector was TIG-welded into the bottom after the appropriate welding parameters were established by metallography. The

top 3/6 in. of each tube was enlarged 0.010 in. on the diameter to accommodate a loading funnel used to prevent contamination of the jacket tube during loading.

The pellets, which are $(U_{0.8}Pu_{0.2})O_2$ bodies from coprecipitated oxides, were weighed and inspected for diameter and length before loading. The pellets were loaded into the jacket tubes, by hand, through a plastic membrane. This technique completely eliminated rejections due to weld contamination.

The loaded jackets were sealed by TIG-welding an upper connector into the top of the jacket tube. Metallography was used to establish proper welding parameters. Each completed element was checked for contamination, tested for leaks, and inspected for dimensions before being accepted.

4. Materials Behavior, Equation of State, and Energy Transfer

a. Equation-of-state Studies (C. E. Dickerman)

Last Reported: ANL-7382, pp.142-147 (Sept 1967).

The thermal expansion of UC at high temperatures has been measured* to provide data for accident analyses and for use in analysis of future transient safety experiments on carbide fuel in the TREAT reactor; the method has been described previously.** Thermal-expansion coefficients were first measured for the TaC crucible material.†

Results of measurements of the thermal-expansion coefficient of sintered UC to 2400°C are shown in Fig. V.A.6. UC was observed in the

Fig. V.A.6
Thermal-expansion Coefficient of Solid UC

*Richards, H. K., "Physical Properties of Uranium Monocarbide at the Melting Temperature and in the Liquid State," Reactor Physics Division Annual Report, July 1, 1966 to June 30, 1967, ANL-7310 (to be published).

**Reactor Development Program Progress Report for November 1966, ANL-7279, p. 92, and for May 1967, ANL-7342, p. 115.

†Reactor Development Program Progress Report for August 1967, ANL-7371, p. 110.

solid state even at 2500°C; hence, from observations made during these experiments, the UC used for the measurements apparently had a melting point near 2550°C. This value is close to that of 2590°C, as reported by Chiotti,* and different from the 2450°C usually accepted. The values in Fig. V.A.6 are the result of several careful determinations.

The change in UC volume at the melting point was also measured. The height of UC in the crucible was measured at 2600°C and again at 2400 or 2410°C. The ratio of this difference and the height at room temperature gave, in three experiments, the following results for linear expansion: (1) 4.47%, (2) 3.8%, and (3) 3.975%.

Making these measurements at the exact melting point would be desirable, but the transition is not quite uniform. Over the range of 200°C used for the measurement, there also will be expansion in both the solid and liquid states. This variation is, however, less than 10% of the variation at the melting point, estimated from the expansion coefficient in the solid state, and was neglected as a first approximation. Inspection of the material afterwards revealed that the UC had many vertical cracks, indicating that it contracted essentially in all directions, when it solidified. It was therefore concluded that the observed values represented linear contraction or expansion rather than a volume change.

By including the estimated thermal expansion over the 200°C difference between the two measurements near the melting point, it may be reasonable to decrease the observed linear variation by the average of the measured values given above or 4.08%. If corrected for the estimated thermal expansion over 200°C, the value of 3.75% appears reasonable. The volume change would result in about 12.2% before correction and about 11.2% after correction for the thermal expansion.

Only one experiment was conducted on liquid UC expansion. The results are rather uncertain. The volume expansion between 2650 and 2740°C appears to be about $1.10 \times 10^{-4}/^{\circ}\text{C}$; therefore $\alpha(\text{Length}) \approx 36 \times 10^{-6}/^{\circ}\text{C}$. This would be more than twice the value of the solid expansion coefficient at 2400°C.

b. Violently Sprayed Sodium-Air Reaction (L. Leibowitz, R. O. Ivins)

Last Reported: ANL-7302, pp. 93-95 (Jan 1967).

Safety analyses for sodium-cooled reactors require knowledge of the temperatures and pressures generated during sodium combustion in

*Chiotti, P., Experimental Refractory Bodies of High-melting Nitrides, Carbides, and Uranium Dioxide, J. Amer. Cer. Soc. 35, 123 (1952).

air. The reaction of sodium sprays in air is being studied. Pressure-generation rates and peak pressures and temperatures are being determined as functions of the initial conditions.

The apparatus used in this work, and preliminary tests of the injector system, have been previously described (see Progress Report for January 1967, ANL-7302, p. 93). Briefly, the apparatus consists of a pneumatically operated piston spray injector that will discharge approximately 11 g of molten sodium into a reaction chamber of known volume.

A series of preliminary experiments was conducted in an effort to characterize the particle-size distribution of the sodium spray. The spray injector head was inverted and mounted on top of a Pyrex glass pipe, 3 ft high and 6 in. in diameter. A Dewar of liquid nitrogen was placed at the outlet of the pipe. The sodium was sprayed into the Dewar through a nonreactive nitrogen atmosphere in the pipe, and sieve analyses were performed on the quenched spray. Results of these analyses did not indicate a predominant particle size; about 60 w/o of the particles had diameters that varied rather uniformly over the range of 0 to 840 μ . Visual observation of the particles larger than 840 μ revealed that they were agglomerates of smaller particles.

Three runs were made to observe the behavior of the spray photographically. Molten sodium at 400°C was injected (using a driving pressure of 1000 psig on the piston) into a sealed 3-ft-high Pyrex glass pipe containing dry air; high-speed motion pictures were taken of the burning phenomena. These films show that vigorous combustion occurs and that the intensity increases when the spray impinges on a baffle at the top of the pipe. This increase is probably due to particle breakup and dispersion upon impact. However, on disassembly of the apparatus, some unreacted sodium adhered to the baffle and the upper 1 in. of the pipe wall.

Currently, experiments are being performed using a steel pipe (3 ft high and 6 in. in diameter), equipped with viewports, as a spray chamber. Temperatures are recorded at points 4, 18, and 32 in. above the injector, and at the center of the top baffle. Two pressure transducers are mounted on the top of the chamber; one is of the strain-gauge type, and the other is a quartz crystal transducer with a high-speed response. A third pressure transducer of the strain-gauge type is located on the side of the chamber, 18 in. above the injector. Samples of the chamber atmosphere are taken before, immediately after, and 10 min after sodium ejection and are analyzed for oxygen content. In this set of experiments, the driving pressure on the piston will be varied, thereby changing the spray velocity and particle-size distribution. In addition, the oxygen content of the atmosphere in the reaction chamber will be varied from that of air (20%) to that of depleted air (5%).

Temperature and pressure measurements from preliminary experiments using this steel pipe apparatus confirm the observation noted above that reaction of sodium particles ceases after they adhere to the top of the chamber.

An attempt is also being made to predict mathematically the pressure rise rate in a closed volume due to the sodium-air reaction. The method is based on a modification of a model proposed by Mouradian and Baker.* If it is assumed that the sodium burning rate is controlled by the diffusion of oxygen to the combustion zone, the usual heat transfer and diffusion correlations can be used to determine steady-state reaction rates for single spherical particles of sodium at any given set of conditions. A quasi-steady-state approach and an averaging technique can then be used in correlating the reaction rates of the individual particles with the burning rate of the spray and the pressure rise rate of the enclosed volume. This method will be discussed in detail when it is more fully developed.

c. Segregation in Ceramic Fuels (M. G. Chasanov)

Last Reported: ANL-7317, pp. 110-112 (March 1967).

Migration and segregation of fissile and fertile material within a fast reactor fuel during its operational lifetime could have significant effects on the operational safety of the fuel. The migration of plutonium and fission products in a thermal gradient is being investigated for mixed oxide fuels, and the program will be extended to carbide fuels.

Development of the thermal-gradient furnace for use in the migration studies has been previously described, and several preliminary experiments using $\text{UO}_2\text{-CeO}_2$ pellets reported (see Progress Reports for September 1966, ANL-7255, p. 87, and March 1967, ANL-7317, p. 110). Experiments have now been initiated on $\text{UO}_2\text{-PuO}_2$ pellets. These experiments are being performed on right-circular cylindrical pellets $1\frac{1}{2}$ in. high by $1\frac{1}{2}$ in. in diameter which were prepared from coprecipitated $\text{UO}_2\text{-PuO}_2$ by Nuclear Materials and Equipment Corp.; the pellets are all nominally $(\text{U}_{0.8}\text{Pu}_{0.2})\text{O}_{2.00}$. Photomicrographs indicate the material to be a single phase with relatively fine grain size.

Two 100-hr-duration experiments were carried out in the thermal-gradient apparatus by heating these materials in an atmosphere of several hundred Torr of argon. In experiment P-1, the sample was contained in an inverted metal crucible fabricated from 20-mil tungsten; this served as an effective barrier to loss of material by vaporization. However, because of heat conduction down the metal walls, somewhat lower

* Mouradian, E. M., and Baker, L., Nuc. Sci. Eng. 15, 388 (1963).

gradients were obtained than in experiments without the crucible. Experiment P-2 was performed without a tungsten crucible around the pellet. After each experiment, the samples were analyzed for plutonium and uranium using an electron-microprobe analyzer. The results of these analyses, given below, should be regarded as preliminary and subject to revision.

Sample P-1 ($T_{\text{top}} = 1980^{\circ}\text{C}$, $T_{\text{bottom}} = 1280^{\circ}\text{C}$), on completion of the experiment, was characterized by a porous region centered about $1500\ \mu$ from the top surface; a vertical crack ran down the center of the pellet. A microprobe scan was made parallel to the vertical axis of the cylinder and about $600\ \mu$ from the centerline. No marked redistribution of plutonium was indicated; however, there was a small increase (about 1 w/o) in plutonium concentration in the vicinity of the porous region. Horizontal traverses across the sample with the microprobe showed no marked variations of plutonium concentration except in the porous region, where the concentration varied about ± 2 w/o from the initial composition. However, these concentration changes are relatively minor, and apparently no significant plutonium migration occurred in this sample.

Sample P-2 ($T_{\text{top}} = 2070^{\circ}\text{C}$, $T_{\text{bottom}} = 975^{\circ}\text{C}$), without the tungsten crucible, did not display the porosity observed in P-1. There was a weight loss of about 0.45%, coupled with growth of some dendrites at the top of the sample. A horizontal crack traversed the lower tenth of the sample, and considerable cracking occurred in the central portion of the pellet; the cracking was associated with areas where materials had been broken out during polishing of the mounted specimen. Microprobe analyses indicated that no substantial plutonium migration occurred in P-2. A vertical traverse by the microprobe at about $1500\ \mu$ from the centerline showed somewhat more variation in plutonium concentration than was observed in sample P-1, with a possible trend toward slightly higher plutonium concentrations at the hotter end of the sample. The somewhat greater variation in plutonium concentration may be due to analytical uncertainties, or possibly is a result of changes that may have taken place in the O/M ratio for this sample.

Two longer-term (500-hr) gradient experiments have been completed, and the samples are currently being examined.

5. TREAT Operations (J. F. Boland)

Last Reported: ANL-7382, p. 152 (Sept 1967).

a. Reactor Operations. The purpose and scope of this work are (1) to provide staff and operating support to carry out experimentation and neutron radiography, (2) to provide technical assistance to organizations preparing experiments for irradiation in the reactor, (3) to design and analyze a limited number of experiments of interest to the reactor safety program, and (4) to provide

reactor loading changes, reactor maintenance, and planning and scheduling related to operation, maintenance, and experimentation.

Two General Electric capsules, C4I and C4J, containing urania-plutonia fuel were subjected to one nuclear transient each.

One vibratory-compacted, UO_2 -powder fuel rod received one nuclear transient in a metal-water photographic capsule. Post-transient examination showed that the rod was close to failure as indicated by cladding deformation along the length of the rod and the cracking of the oxide layer formed on the cladding exterior.

Two items were neutron-radiographed: (1) a GE capsule C4J and (2) an EBR-II standard upper shield, containing an experimental melt-wire assembly.

Replacement of high-tensile carbon-steel studs with A-286 studs in the large sodium-loop flanges was begun.

b. Development of Automatic Power-level Control System. This task covers the design, procurement, and installation of an automatic power-level control system for the TREAT reactor. Such a sophisticated control system is required to permit (1) performance of quasi-steady-state, loss-of-flow experiments, and (2) transient shaping designed to simulate normal fast-reactor fuel-temperature profiles prior to a transient which simulates a reactor over-power transient. Program approval for this task was received during October, and engineering will be started in early November.

B. Effluent Control -- R&D -- Gaseous Effluent Studies -- Plutonium Volatility Safety

1. Plutonium Hexafluoride Safety (R. W. Kessie)

Last Reported: ANL-7308, pp. 93-95 (Feb 1967).

The safety aspects of handling large quantities of PuF_6 in fluoride-volatility fuel-reprocessing plants of the future have been considered. The amount of exposure of the public was calculated for a maximum credible accident involving PuF_6 release. The following assumptions were made:

(1) The maximum quantity of PuF_6 available for release at any one time would be 10 kg.

(2) The released PuF_6 would pass through at least one operating scrubber and then through two intact high-efficiency filters.

(3) After passing through the filters, the material would be discharged from a 100-meter-tall stack.

A ventilation air-cleanup system would need to be designed to ensure that a scrubber and two filters would be intact after the postulated accident. Hydrolysis of PuF_6 with moisture in the gas phase, followed by filtration through two high-efficiency filters, would allow 10^{-9} to 10^{-6} of the released plutonium to be discharged from the second filter. The maximum exposure at the ground, following a maximum credible accident and released from a 100-meter stack, was calculated for (1) fuel containing only the ^{239}Pu isotope and (2) fuel with plutonium isotopic compositions resulting from high burnup in fast or thermal reactors. For the former case, exposure would be less than 0.7% of the annual average allowed by Federal regulations;* for the latter case, exposure would be less than 10% of the allowable annual average.

Within the assumptions stated above, the cleanup technique involving hydrolysis and filtration, which has been tested with up to 6 g of PuF_6 , will allow a full-scale fluoride-volatility fuel reprocessing plant to be designed that will be safe with respect to a maximum credible accident involving PuF_6 release.

2. Effluent Gas Treatment (D. Vissers)

Last Reported: ANL-7382, p. 153 (Sept 1967).

a. Chemistry of Tellurium Fluorides. The volatile fluoride species of fission-product tellurium (tellurium hexafluoride) represents a troublesome component in the off-gases of the fluoride-volatility processing of spent nuclear fuels. It presents a serious off-gas decontamination problem because of the relatively high concentration of radioactive tellurium in the spent fuel, the chemically inert character of TeF_6 , and the low maximum permissible concentration ($1 \times 10^{-8} \text{ Ci/m}^3$) for continuous occupational exposure to fission-product tellurium.

Laboratory-scale gas-flow sorption studies (see Progress Report for July 1967, ANL-7357, pp. 144-145) have shown that activated alumina is an excellent sorbent for TeF_6 in fluorine-containing gas systems. To obtain a better understanding of the TeF_6 sorption on activated alumina, a series of static sorption studies were performed. Sorption isotherms were obtained at 5, 26, 40, and 60°C as a function of TeF_6 pressure, from which the heat of sorption for TeF_6 was estimated. The heat of sorption was approximately 5 kcal/mole. This compared to a heat of sublimation

* "Standards for Protection Against Radiation," Title 10, Code of Federal Regulations, Part 20 (10 CFR 20) (Aug. 9, 1966).

for TeF_6 of approximately 6.5 kcal/mole.* The results of these studies indicate that TeF_6 is physically sorbed on the surface of the activated alumina. Displacement of the TeF_6 from the activated alumina by fluorine, previously noted (see Progress Report for September 1967, ANL-7382, p. 153), is therefore not unexpected.

The gas-flow sorption studies with tellurium hexafluoride have been completed. These studies indicate that activated alumina in the form of a packed-bed sorbent will probably meet the removal requirements for tellurium hexafluoride in the presence of fluorine for the fluid-bed fluoride-volatility processing of fast reactor fuels. No further work is planned in this area.

C. Engineered Safety Features

1. Safety Features Technology--Containment

a. Two-dimensional Codes for the Analysis of Containment Structures (S. H. Fistedis)

Last Reported: ANL-7382, p. 154 (Sept 1967).

The difference equations have been formulated. The space derivatives are being evaluated by the midpoint method, which is used because (1) it requires fewer arithmetic steps than other methods, and (2) it eliminates the acceleration reversal which can occur when the mesh is sufficiently distorted and when mesh points become inverted.

The momentum equations at mesh point (I, J) in finite-difference form at time t^n are

$$\begin{aligned} \ddot{R}_{I,J}^n = \frac{-1}{8A\rho} \left\{ \left(P_{I+\frac{1}{2},J+\frac{1}{2}}^n - P_{I-\frac{1}{2},J-\frac{1}{2}}^n \right) \left(Z_{I,J+1}^n - Z_{I,J-1}^n + Z_{I-1,J}^n - Z_{I+1,J}^n \right) \right. \\ \left. - \left(P_{I-\frac{1}{2},J+\frac{1}{2}}^n - P_{I+\frac{1}{2},J-\frac{1}{2}}^n \right) \left(Z_{I,J+1}^n - Z_{I,J-1}^n + Z_{I+1,J}^n - Z_{I-1,J}^n \right) \right\} \end{aligned}$$

and

$$\begin{aligned} \ddot{Z}_{I,J}^n = \frac{1}{8A\rho} \left\{ \left(P_{I+\frac{1}{2},J+\frac{1}{2}}^n - P_{I-\frac{1}{2},J-\frac{1}{2}}^n \right) \left(R_{I,J+1}^n - R_{I,J-1}^n + R_{I-1,J}^n - R_{I+1,J}^n \right) \right. \\ \left. - \left(P_{I-\frac{1}{2},J+\frac{1}{2}}^n - P_{I+\frac{1}{2},J-\frac{1}{2}}^n \right) \left(R_{I,J+1}^n - R_{I,J-1}^n + R_{I+1,J}^n - R_{I-1,J}^n \right) \right\}, \end{aligned}$$

* Vissers, D. R., and Steindler, M. J., Laboratory Investigations in Support of Fluid-bed Fluoride Volatility Process. Part X. A Literature Survey on the Properties of Tellurium, Its Oxygen and Fluorine Compounds, ANL-7142, p. 20 (Feb 1966).

where

$$P^n = p^n + q^{n-\frac{1}{2}},$$

$$A = \frac{1}{8} \left\{ \left(Z_{I,J+1}^n - Z_{I,J-1}^n \right) \left(R_{I+1,J}^n - R_{I-1,J}^n \right) - \left(R_{I,J+1}^n - R_{I,J-1}^n \right) \left(Z_{I+1,J}^n - Z_{I-1,J}^n \right) \right\},$$

ρ is the density, and q is the von Neumann-Richtmyer artificial viscosity. The form of q is under study.

The pressure p and internal energy E of each mesh zone are obtained from the equation of state,

$$p = p(E, V_s),$$

and from the energy equation,

$$\frac{\partial E}{\partial t} = -P \frac{\partial V_s}{\partial t},$$

where $V_s = \rho^{-1}$ = specific volume. The equation of state of all materials considered in this problem is assumed to have the form $p = p(E, V_s)$.

The difference equations for these equations are

$$P_{I+\frac{1}{2}, J+\frac{1}{2}}^n = p_{I+\frac{1}{2}, J+\frac{1}{2}}^n + q_{I+\frac{1}{2}, J+\frac{1}{2}}^{n-\frac{1}{2}};$$

$$p_{I+\frac{1}{2}, J+\frac{1}{2}}^n = p \left(E_{I+\frac{1}{2}, J+\frac{1}{2}}^n, V_{sI+\frac{1}{2}, J+\frac{1}{2}}^n \right);$$

$$E_{I+\frac{1}{2}, J+\frac{1}{2}}^n = E_{I+\frac{1}{2}, J+\frac{1}{2}}^{n-1} - \frac{P_{I+\frac{1}{2}, J+\frac{1}{2}}^n + P_{I+\frac{1}{2}, J+\frac{1}{2}}^{n-1}}{2} \left(V_{sI+\frac{1}{2}, J+\frac{1}{2}}^n - V_{sI+\frac{1}{2}, J+\frac{1}{2}}^{n-1} \right).$$

These equations cannot be solved simultaneously for two unknowns p^n and E^n . In the actual computation, iterative techniques will be used. The time interval Δt at each cycle is controlled by the White stability number. Since the computer storage space and calculating time are the two major concerns in this problem, the programming techniques are being studied.

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