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Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

June 1966

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

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IN NUCLEAR SCIENCE ABSTRACTS

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Reactor Technology
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AEC Research and
Development Report

ARGONNE NATIONAL LABORATORY
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Argonne, Illinois 60439

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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

June 1966

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FOREWORD

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

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

The last six reports issued
in this series are:

December 1965	ANL-7132
January 1966	ANL-7152
February 1966	ANL-7176
March 1966	ANL-7193
April 1966	ANL-7204
May 1966	ANL-7219

REACTOR DEVELOPMENT PROGRAM

Highlights of Project Activities for June 1966

EBR-II

Operation of Run No. 18, begun on May 25, was concluded successfully on June 25.

Results of examination of a Mark-IA fuel subassembly which had achieved a maximum burnup of 1.2 a/o in the reactor showed that the sodium bond level (which is an indicator of fuel swelling) was approaching the pre-determined allowable limit. Accordingly, further reactor operation has been held up while additional selected subassemblies are inspected and the performance of fuel further evaluated.

Thirteen subassemblies were fabricated in the Fuel Cycle Facility and ten subassemblies were transferred to the reactor in a continuing routine operation.

ZPPR

The general construction package for the facility was submitted to contractors for bids on May 26, with a "due date" of July 7. The hold order on procurement of equipment was canceled on May 31.

ZPR-3

A series of experiments has been initiated with plutonium assemblies having well-degraded spectra to provide data for evaluating calculational methods and current compilations of nuclear cross sections.

Plutonium Recycle Experiment (EBWR)

The average uniform temperature coefficient of reactivity between 79°F and 153°F for the full loading when poisoned with approximately 4.4 g/gal H_3BO_3 was determined as $(3.9 \pm 0.7) \times 10^{-5} \Delta k/^\circ\text{F}$.

AARR

The drawings and text of the Title I report has been submitted by the Architect-Engineer for review.

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I. LIQUID-METAL-COOLED REACTORS

A. EBR-II

1. Operations

Reactor Run No. 18 was completed after 875 MWdt. EBR-II has now been operated for a total of 8490 MWdt. Following Run No. 18 and after allowing sufficient cooling time, two fuel subassemblies, Mark-IA subassemblies Nos. C-169 and C-185, having burnups of 1.2 a/o and 1.1 a/o, respectively, were transferred to the Fuel Cycle Facility for surveillance measurements.

Reactor Run No. 19, scheduled for 490 MWdt, was started on June 27. Operation beyond 1.3 a/o burnup was contingent on the results of the surveillance measurements on the above removed subassemblies.

On June 28, when the results of the above measurements became known, the reactor was shut down. Fuel expansion in C-169, as determined by change in sodium bond level, had approached the preset limit. This was unanticipated because Mark-IA fuel with its larger expansion volume, was expected to yield a higher burnup. Shutdown will continue until the reason for the unexpected growth can be determined and the basis for further operation above 1.2 a/o burnup evaluated. 80 MWdt were produced in the reactor Run No. 19.

Fuel changes for Run 19 were accomplished by replacement of ten enriched core-type subassemblies, including one control subassembly by a similar Mark-IA type and the replacement of two enriched inner blanket subassemblies by two of depleted uranium type. This latter change was made to compensate for the reactivity increase caused by the presence of additional Mark-IA subassemblies in the reactor and the unavailability of experimental irradiation subassemblies which had previously been programmed for insertion.

Nine randomly selected outer blanket subassemblies were lifted and replaced in their respective grid positions as part of the continuing program of blanket surveillance. No evidence of sticking or binding was observed.

The results of two tests with the secondary sodium pump to determine the rate of temperature rise in the windings after loss of cooling water due to loss of normal electrical power indicate that the pump may be operated at low voltage, reverse head, as would be necessary for at least 30 min after loss of cooling water, without overheating the pump. Further testing is contemplated to confirm this value.

During a routine overspeed trip check of the turbine-generator on June 24, a noise was heard in the vicinity of the turbine end of the generator, and this was followed by an abnormal vibration of the machine. During a

test of this type, it is standard practice to check the trip speed three times. The noise occurred during the third check following the first two uneventful and satisfactory checks. Measurement of vibration amplitude on the adjacent generator bearing indicated deflection of 3 mils in the vertical direction. Previously measured amplitude had been negligible. The turbine-generator was shut down and will not be operated until inspection by the manufacturer's representative has been accomplished and necessary corrections made.

2. Maintenance and Modification.

In the shutdown period between Runs 18 and 19, maintenance tasks and minor modifications were carried out.

A satisfactory leak-rate test was performed on the reactor building air inlet valves; the Datex (IBM) card system for the fuel-handling system was modified to accept additional card-input data; the pump end of the turbine-driven feedwater pump was disassembled, inspected, and minor repairs accomplished. No cause could be found for its previously experienced marginal performance.

Cables which had been damaged by radiation were removed from a low level nuclear instrument thimble and replaced.

3. Reactor Improvements

a. Oscillator System Mark II. The temporary control console and drive unit wiring (see Progress Report for May 1966, ANL-7230, p. 2) were checked out. Final preparations for the test runs in sodium at elevated temperatures were completed.

The first 350°F sodium runs were started with the dummy oscillator rod and the actual drive unit operating at speeds up to 300 rpm. Torque readings taken after each run confirmed satisfactory performance of the drive. Tests at speeds up to 600 rpm are being conducted.

b. Monitor for Spent-fuel Subassembly. Normal fuel-handling procedures involve several transfers of identically shaped core subassemblies between the reactor and the storage rack during the same shutdown period. A Spent Fuel Subassembly Monitor has been designed to prevent reloading a previously irradiated fuel element into the core. This reinsertion would expose the subassembly to higher than normal burnup, which could damage the subassembly or, possibly, the reactor.

The Spent Fuel Subassembly Monitor consists of a special high-temperature gamma ionization chamber and necessary readout instrumentation to differentiate between the gamma-radiation levels of a spent subassembly and a refabricated core subassembly. The gamma chamber will be located in a thimble extending through the "X" nozzle into the primary

tank. The chamber is thus located at the periphery of the transfer arm circle. A mechanism within the thimble allows the chamber to be raised or lowered 52 in. at an elevation corresponding to the core section of the subassembly when it is supported by the transfer arm. Calculations indicate that the radiation (including background) after 14 days of storage varies by a factor of 10^4 between a refabricated core subassembly and a 1%-burnup spent core subassembly. For every core subassembly, a slightly modified fuel-handling sequence will incorporate a transfer-arm stop at the Spent Fuel Subassembly Monitor location just prior to its transfer to the gripper mechanism for insertion into the reactor. This procedure will guarantee identification of subassemblies, thus aiding the normal, careful accounting procedures.

The preliminary design study has been completed, the conceptual design drawings made, the purchase specifications completed, and the work of providing detailed drawings has been started.

Construction of the prototype and the initial tests are expected to be completed by January 1967.

4. Reactor Physics

Calculations were made to determine the effect of stainless steel axial end blankets in place of depleted uranium on reactor operation. The analysis indicated that the power distribution in the core would be unaffected by the stainless steel, but for a constant core power there would be a loss of 1.5% power due to loss of fissions in the blanket. There would also be a calculated loss of $0.001 \Delta k/k$ over the entire core with the stainless steel blanket. The conclusion was reached that this changeover could be accomplished without any significant effect on reactor power operation. However, further calculations are being made to determine the effect of the change in the neutron and gamma flux on the grid plate.

The first control rod of the Mark-IA type was inserted into the core in position 5-C-3 (control rod No. 12). The measured worth of the rod was about 170 lh. This is in agreement with the increased enrichment of the Mark-IA fuel and shortened fuel length.

5. Fuel Cycle Facility

Measurements of fuel elements from two additional (see Progress Report for May 1966, ANL-7219, p. 6) Chicago-manufactured Mark-IA irradiated subassemblies (C-185, 1.13 a/o, and C-169, 1.22 a/o maximum burnup) showed higher sodium bond levels than were measured in Mark-I fuel of comparable burnup values. Burnup values are plotted as a function of fuel volume change, and are shown in Fig. 1 together with corresponding curve obtained from Mark-I fuel.

Fig. 1. Increase of Fuel-pin Volume vs Maximum Burnup for Mark-I and -IA Fuel

The pressure and volume of the void space in the top of these fuel cans will be measured at ambient temperature. These pins will be de-canned, and length and diameter measurements of the fuel pins will be obtained. Additional Mark-IA fuel subassemblies at the 1.2 a/o burnup level will be removed from the reactor for measurements to confirm these data.

Arcing between the top turn of the induction coil and the steel furnace frame occurred in both melt-refining furnaces. One furnace was replaced with a spare furnace unit; the other has been removed from the cell for decontamination and rebuilding. Examination of the furnaces, after the arcing, showed an apparent loss of insulating (electrical) properties of the induction-coil supports. This could be a result of deposition of condensed sodium or fission products that were not contained in the fume trap that fits over the top of the crucible during the melt refining runs. One furnace (A) had operated for 30 runs, and the other (B) for 50 runs, prior to failure; therefore, the number of runs does not appear to be the

contributing factor. However, a correlation of the amount of sodium-coated fuel melted in each furnace (220 kg in A and 260 kg in B) indicates that condensing sodium may be the cause of the failure. The coil supports for the furnace being rebuilt are being redesigned to increase the arc path length.

A summary of production is shown in Table I.

TABLE I. Production Summary for June 1966

Subassemblies received:			19
Subassemblies dismantled:			14
Subassemblies fabricated:			13
Subassemblies transferred to reactor:			10
Pins decanned:			1358
Melt refining:			
Avg pour yield	9 irradiated	7 recycle	
	92.6%	93.1%	
Injection casting runs:			15
Pin Processing:			
Accepts			1461
Rejects			253
Pins welded:			1504
Leak testing:			
Accepts			1270
Rejects			20
Bond testing (complete runs)			
Accepts			1107
Rejects			137
Surveillance:	C-180, C-201, C-185, C-169		

B. General Fast Reactor Physics

1. ZPR-3

A series of plutonium assemblies with well-degraded spectra has been designed for ZPR-3 to follow the program of measurements on Assembly 47, the SEFOR mockup. The program objective is to provide a set of integral data which can be used for the evaluation of calculational methods and nuclear cross-section compilations employed in the design of large plutonium-fueled fast power reactors. Thereby both the reactor designer and the nuclear physicist can be accommodated, and it will still be possible to construct assemblies simple in both composition and geometry.

Each assembly will consist of a cylindrical core zone surrounded by a depleted uranium blanket. The core compositions are based upon simple combinations of plutonium, depleted uranium, sodium, and graphite plates. The assemblies are divided into two sets with a constant ratio of U^{238} to Pu for each set. Within each set an interesting range of flux and adjoint spectra are predicted, with the flux fractions below 10 keV being typical of those predicted for very large fast power reactors.

The experimental program is designed to provide important information on critical enrichment and Doppler and sodium temperature coefficients of reactivity, which are of interest in power reactor design. The

neutron spectrum will be studied directly by the use of spectral indices. Such data will provide checks on the nuclear data for a range of materials which will be used in future power reactors.

The first of these assemblies, Assembly 48, is being investigated in the ZPR-3. The design composition of this assembly is given in Table II. Minor variations in the composition due to variations in the plutonium cladding, stainless steel content, and the increased fissile content of the ten safety control rods will be made.

TABLE II. Homogeneous Compositions for
Assembly 48
($\times 10^{24}$ atoms/cm³)

Isotope	Core ^(a)	Reflector ^(b)
Pu ²³⁹	0.001644	0.000
Pu ²⁴⁰	0.000106	0.000
Pu ²⁴¹	0.000010	0.000
Pu ²⁴²	0.000000	0.000
U ²³⁵	0.000016	0.000084
U ²³⁸	0.007406	0.039976
C	0.020765	0.000
Na	0.006230	0.000
Fe	0.009899	0.004453
Cr	0.002658	0.001196
Ni	0.001308	0.000588
Mo	0.000206	0.000
Al	0.000110	0.000

^aThe core length is 30.06 in.

^bThe reflector thickness is a nominal 30.0 cm, both axially and radially.

In the approach to critical, at approximately 2/3 of the estimated critical mass, the proton-recoil neutron spectrometer was loaded into the assembly. A series of experiments with both hydrogen- and methane-filled counters was performed. The gamma background of the core was found to be sufficiently low so as to not interfere with the measurements. These data are now being analyzed; the preliminary results indicate that the measurements should define the flux in the low-keV range of neutron energy.

Upon completion of the spectrum measurements, the approach to critical was concluded. Evaluations of control-rod worth, worth of edge fuel material, and comparisons of control-rod calibration and period-measuring techniques are now being done. This is to be followed by

evaluation of control-rod worth compared to normal drawer composition and additional neutron-spectrum measurements.

2. ZPR-6

Loading of the full-scale version of a 2600-liter carbide core (Assembly No. 5) was completed. The composition is given in Table III.

TABLE III. Atomic Densities in Assembly No. 5

Material	Atomic Density ($\times 10^{-24}$ atoms/cm ³) in Core	Atomic Density ($\times 10^{-24}$ atoms/cm ³) in Reflector
U ²³⁵	0.00153*	0.000081
U ²³⁸	0.01056	0.03993
Na	0.00920	0.000
C	0.01293	0.000
Fe	0.0904	0.004322
Ni	0.00113	0.00054
Cr	0.002386	0.00114

*Based on the assumption that all plates will be 1/16 in. thick.

3. ZPR-9

Central worth measurements were made with Assembly No. 11 of ZPR-9, a smaller zoned version of Assembly No. 4Z of ZPR-6 (see Progress Report for May 1965, ANL-7046, pp. 17-18). The reactivity worth of a sample was determined by measuring reactor periods with and without the sample. The reactor had about 15 lh excess reactivity without any sample. The results, together with those from Assembly 4Z, are shown in Table IV.

If the real and adjoint spectra at the center of Assembly No. 11 were identical to the spectra at the center of Assembly No. 4Z, the ratio of the worth of a sample in Assembly No. 11 to the worth in Assembly No. 4Z would be the same for all samples. The calculated spectral matching between Assembly No. 11 and Assembly No. 4Z was such that the largest deviation in any one energy group was about 3%. In order to insure adequate spectral matching, a good criterion would be the constancy of the ratios of the calculated worths of different samples in the two systems. To ascertain that the fluctuation in this ratio is due to differences in the real and adjoint spectra in the two assemblies, this ratio was calculated with cross-section set 224, the results being given in the last column of Table IV. The calculated results also show a considerable deviation in the ratios, particularly for beryllium, carbon, and sodium.

TABLE IV. Central Worth Measurements in ZPR-9 Assembly No. 11

Sample and Weight (g)	Weight of Can (g)	Gross Worth (1H)(a)	Net Worth (1h)	Net Worth in Assembly 4Z (1h)	Worth in Assembly 11 Worth in Assembly 4Z	
					Measured	Calculated
Be (114.308)	Bare	+0.671 ± 0.057	0.671 ± 0.057	+1.107 ± 0.010	+0.606	+0.090
C (103.0)	Bare	-0.220 ± 0.057	-0.220 ± 0.057	+0.197 ± 0.010	-1.116	+9.36
Ni (546.0)	Bare	-1.973 ± 0.057	-1.973 ± 0.057	-1.148 ± 0.010	+1.718	+1.90
V (183.67)	55.30 (SS)	-0.498 ± 0.057	-0.334 ± 0.060	-0.133 ± 0.010	2.511	+2.00
Li (28.30)	57.03 (SS)	-1.378 ± 0.023	-1.208 ± 0.029	-0.595 ± 0.010	2.030	-
Zr (406.0)	Bare	-1.136 ± 0.023	-1.136 ± 0.023	-0.650 ± 0.005	1.748	1.93
Mo (599.0)	Bare	-5.293 ± 0.023	-5.243 ± 0.023	-3.415 ± 0.010	1.550	1.89
Al (166.90)	Bare	-0.518 ± 0.023	-0.518 ± 0.023	-0.297 ± 0.013	1.744	2.08
B ¹⁰ (29.29)	55.73 (SS)	-35.676 ± 0.030	-35.520 ± 0.035	-23.339 ± 0.200	1.522	1.88
B ₄ C (59.62)	64.70 (SS)	-9.876 ± 0.028	-9.684 ± 0.034	-6.410 ± 0.025	1.511	-
Polyethylene (58.98)	Bare	+28.652 ± 0.028	28.652 ± 0.028	+19.696 ± 0.010	1.454	-
Nb (481.2)	30.10 (SS)	-5.900 ± 0.048	-5.811 ± 0.051	-3.791 ± 0.010	1.533	1.88
W (1108.0)	-	-8.928 ± 0.030	-8.928 ± 0.030	-5.742 ± 0.010	1.555	1.90
Ta (924.70)	27.54 (SS)	-18.30 ± 0.10	-18.22 ± 0.10	-11.907 ± 0.200	1.530	1.88
U ²³⁸ (1153.80)	Bare	-5.908 ± 0.010	-5.908 ± 0.010	-3.857 ± 0.010	1.532	1.88
U ²³⁵ (27.719)	SS can (47.30)	+1.946 ± 0.025	1.833 ± 0.028	+1.246 ± 0.030	1.471	1.94
U ²³⁵ (64.183)	SS can (47.30)	+4.827 ± 0.050	+4.714 ± 0.053	+2.890 ± 0.030	1.631	-
Na (51.38)	56.42 (SS)	-0.282 ± 0.015	-0.114 ± 0.035	-0.049 ± 0.005	2.326	2.41
SS 304 (499.6)	Bare	1.401 ± 0.040	-1.401 ± 0.040	-0.849 ± 0.010	1.650	-
Pu (117.157)	Al (7.093)	+13.714 ± 0.050	+13.795 ± 0.055	+8.44 ± 0.20	1.634	1.93
U ²³³ (146.708)	SS (20.06)	-	-	-	1.809	-
(97.15% U ²³³)	Al (5.286)	20.987 ± 0.200	+21.067 ± 0.200	11.643	-	-
	SS (19.977)	-	-	-	-	-

^a1% Δk/k = 423 lh.

The new apparatus for Doppler measurement has been installed. This equipment is an improved and automated version of that used for the ZPR-6 measurements. It is expected to reduce substantially the reactor time required to make the measurements. Debugging of the apparatus is completed.

A series of measurements on 1/2-in.-dia Doppler samples is underway. The first measurements are designed to test the consistency of the measurements in the small-zoned Assembly No. 11A with the results in the ZPR-6 Assembly No. 4Z.

4. ZPPR

The package for the general construction of the ZPPR facility was submitted to contractors for bids on May 26. These bids are due on July 7. The "hold order" on procurement of equipment was cancelled on May 31.

The following is the present status of equipment procurement:

a. Reactor Bed and Tables. All materials have been received by the manufacturer, Giddings and Lewis, from outside suppliers. The ball screw has been ground and is being heat-treated. Sixty percent of the machining of transmission components has been completed. Rough machining of the casting has been delayed until early July.

b. Matrix Drawers. The vendor shipped two batches of drawers to ANL in early June. The first batch contained 100 drawers, of which 19 were rejected. The second batch of 300 was inspected, and 10 were found

unacceptable. The rejected drawers have been sent to the manufacturer for repairs or replacement. The major cause of rejection was presence of cracks on the weld area on the front of the drawer, which apparently resulted from the final stretch sizing operation. Two ANL representatives visited the manufacturer, Mechanical Products, Seattle, Washington, to review the fabrication techniques and quality control. The vendor was instructed to notify ANL when 600 drawers had been fabricated and inspected by the vendor so that an ANL representative could travel to the plant to inspect the drawers prior to shipment.

c. Nuclear Instrumentation. Fabrication of the nuclear instrumentation was restarted on June 22. All purchased components have been received. The assembly of the units is 53% complete. The estimated overall completion of the nuclear instrumentation is 73%.

An extensive analysis of the special ZPPR fail-safe trip circuit was made. Each active individual component was either opened or shorted, and the circuit condition was monitored. The circuit was tested with respect to high (65°C) and low (0°C) temperature, and high and low supply voltage. This trip circuit checks out to be fail-safe with respect to single component failure.

d. Area Gamma Monitors. The gamma monitors are the same type as used in EBR-II. All purchased components have been received, and assembly is 75% complete.

e. Other Reactor Components. Bid packages for the following items were sent to potential suppliers (dates of submission and date due are shown for each item):

- a. Source drives: June 1-July 15.
- b. Rod drive mounting frame: June 15-July 15.
- c. Knees: June 1-July 15.
- d. Loading platform assembly: June 16-July 15.
- e. Rod drive mounting plate: June 7-July 15.
- f. Drawer springs: June 8-July 15.
- g. Matrix tie-down assemblies: June 8-July 15.
- h. Fuel rod drawer and ball joint assemblies: June 8-July 15.
- i. Poison safety rod drive assemblies: June 16-July 15.
- j. Reactor shields: June 20-July 15.
- k. Tube bundling: June 21-July 13.
- l. Reactor cooling plenum: June 17-July 15.
- m. Stack monitor: June 17-July 15.
- n. Beta-gamma air particulate monitors: June 17-July 15.

These bids will integrate with the construction bid to formulate the latest cost estimate.

C. General Fast Reactor Fuel Development

1. Metal Fuels

The development of metallic uranium-plutonium-based alloys as fuels for power breeder reactors has continued. The high-melting (1150°C) U-Pu-Zr and U-Pu-Ti alloys are suitable as high-burnup fuels and show good compatibility with stainless steel and vanadium alloys. The determination of the thermal conductivities of these alloys is completed; the mechanism of compatibility and the phase diagrams are being studied.

a. Thermal Conductivity. The thermal conductivities of six alloys tested are listed in Table V. Increasing the amounts of titanium or zirconium lowers the conductivity for the same content of plutonium.

TABLE V. Thermal Conductivities ($\text{W/cm}^{\circ}\text{C}$) of U-Pu-Zr and U-Pu-Ti Alloys

Casting No.	D-40		D-53		D-38		D-44		D-54		D-50	
Composition: ^a	Pu	Ti	Pu	Ti	Pu	Ti	Pu	Zr	Pu	Zr	Pu	Zr
a/o	10.0	13.3	15.1	23.9	11.4	29.8	14.7	15.0	12.7	21.9	15.5	25.3
w/o	11.4	3.0	18.6	5.9	15.0	7.8	16.2	6.2	14.7	9.7	18.4	11.5
Temp ($^{\circ}\text{C}$)												
100	0.105		0.095		0.087		0.109		0.100		0.092	
200	0.139		0.125		0.117		0.142		0.132		0.117	
300	0.171		0.155		0.142		0.177		0.164		0.144	
400	0.201		0.185		0.163		0.210		0.196		0.171	
500	0.228		0.216		0.184		0.241		0.225		0.197	
600	0.254		0.245		0.201		0.264		0.253		0.219	
700	0.278		0.275		0.216		0.280		0.278		0.234	
800	0.302		0.306		≈ 0.229		0.293		0.301		0.248	
900	≈ 0.326		≈ 0.335		-		≈ 0.301		≈ 0.323		≈ 0.260	

^aBalance uranium.

The effect of plutonium is not obvious. The (U,Pu) alpha and UZr_2 phases have about twice the conductivity of the (U,Pu) zeta and U_2Ti phases. The addition of plutonium to these alloys will decrease the amount of alpha phase, increase the amount of zeta phase, and should lower the thermal conductivity. The two reference alloys (U-15 w/o Pu-10 w/o Zr and U-15 w/o Pu-6.5 w/o Ti) have about the same thermal conductivity.

The carbon content of all of the alloys was generally around 300 ppm and the oxygen content about 150 ppm. The D36 and D38 castings, however, contained 2400 and 1400 ppm of oxygen, but the effect on thermal conductivity has not been determined as yet.

b. Compatibility. The good compatibility of Type 304 stainless steel with certain U-Pu-Zr alloys at 700 to 800°C (see Progress Report for July 1965, ANL-7082, p. 20) appears from recent findings to be due in large part to the diffusion of oxygen from the steel into the U-base alloys. Upon diffusion of oxygen from the commercial steels, which generally contained more than 200 ppm of oxygen, Zr-rich phases stabilized by oxygen are

formed in interdiffusion layers. The phases, all of which have melting temperatures above 800°C, are tentatively identified as (1) a solution of oxygen in alpha zirconium, (2) a Zr-rich gamma-uranium solution containing plutonium, termed γ_2 , and (3) a phase that is based on the ternary gamma phase that occurs in the Zr-Ni-O system.¹ During annealing for six months at 800°C the formation of these phases in diffusion layers in a Type 304 stainless steel/U-18 w/o Pu-14 w/o Zr alloy couple is seen from results of microprobe analyses to restrict completely the diffusion of iron, nickel, and chromium into the uranium-base alloy. Uranium and plutonium, however, penetrate the phases and diffuse into the steel to form phases that are highly enriched in iron and nickel but not chromium. The total distance across which interdiffusion occurred both in fuel and clad during annealing for six months is at most only 35 μ .

The concentration of nickel in Fe-Cr-Ni alloys containing 18 w/o Cr and less than 23 ppm of oxygen has been determined to influence markedly compatibility with the U-18 w/o Pu-14 w/o Zr alloy at 750°C. With increasing nickel content, the thickness of diffusion layers formed during annealing for seven days decreases; from 170 μ at 5% nickel to 82 and 15 μ at 12 and 20% nickel, respectively. With Type 304 stainless steel containing 8.5% nickel, a diffusion layer only 4 μ in thickness is formed under the same test conditions. The apparent better compatibility of the Type 304 stainless steel is attributed to its higher oxygen content, 420 ppm.

An electron microprobe analysis has been made of a diffusion couple consisting of an U-15 a/o Pu-26 a/o Zr alloy and a modified Type 316 (niobium stabilized) steel containing 11.5 a/o Ni (see Progress Report for March 1966, ANL-7193, p. 21). The couple had been annealed for seven days at 750°C. The analysis indicated extensive diffusion of nickel, uranium, and plutonium. Nickel diffuses 230 μ into the uranium-base alloy to form Zr-rich phases with uranium. Uranium and plutonium diffuse into the steel over a distance of 20 μ . The total width of the interdiffusion zone, 250 μ , contrasts with 5 μ for Type 304 stainless steel under the same test conditions. The enhanced diffusion observed for the Type 316 stainless steel is attributed to its lower oxygen content, 43 ppm, as contrasted to 420 ppm oxygen in Type 304 steel. At the lower oxygen level, formation of oxygen-stabilized Zr-rich phases does not occur.

c. Thorium-Uranium-Plutonium Alloys. The experimental work on the phase diagram of the Th-U-Pu system has been concluded. The system was chosen because of its potential of providing high-temperature fuel alloys for sodium-cooled fast reactors. The investigation made it possible to select alloys that fill the requirements of high-temperature stability. The two most promising alloys, Th-10 w/o U-10 w/o Pu and Th-20 w/o U-10 w/o Pu, are being irradiated in CP-5 and to date have reached burnups of 0.5% at 630°C and 0.34% at 600°C, respectively.

¹Nevitt, M. V., and Downey, J. W., Trans. AIME 221, 1014-1017 (Oct 1961).

A schematic diagram of the liquidus surface is shown in Fig. 2. The liquidus isotherms in the thorium corner of the system are based on observations that were made during the preparation of various alloys in this composition range. The miscibility gap of the binary Th-U system is

Fig. 2. Schematic for the Liquidus Surface in the Th-U-Pu System (temperatures in degrees centigrade)

probably closed in the ternary system at low plutonium concentrations as shown in Fig. 2. A peritectic should extend across the system from the Th-U side to the Th-Pu side if the α -Th phase forms from the β -Th phase by peritectic reaction. A eutectic valley extends from the Th-U side to the plutonium corner of the Th-Pu system where a four-phase reaction, α -Th + L $\rightarrow$ Cp + ϵ , near 610°C is probable; the compound Cp was recently re-determined by Portnoff and Calais to be Pu_7Th_3 .²

Differential thermal analysis (DTA) showed the solidus temperatures of the

Th-10 w/o U-10 w/o Pu and Th-20 w/o U-10 w/o Pu alloys to be about 917 and about 955°C, respectively, little different from the earlier estimates of 925 and 940°C, respectively. Since the solidus temperature is determined by the melting of only a small amount of (U,Pu) phase, the observed effects are small, and the solidus data should be considered accurate only to $\pm 10^\circ\text{C}$. A larger heat effect occurs in both alloys at 990°C, a temperature above which a larger portion of the alloys will be liquid.

The solid-state reaction scheme for the composition and temperature range of interest is shown in Fig. 3. Reaction temperatures were determined by DTA. There are three four-phase reactions, two of Class III and one of Class II, as follows:

- (a) Class III: $\gamma_{\text{U}} + \alpha_{\text{Th}} + \beta_{\text{U}} \rightarrow \eta$ at 714°C;
- (b) Class III: $\alpha_{\text{Th}} + \beta_{\text{U}} + \eta \rightarrow \zeta$ at 610°C;
- (c) Class II: $\alpha_{\text{Th}} + \beta_{\text{U}} \rightarrow \alpha_{\text{U}} + \zeta$ at 595°C.

It is characteristic of the Th-U-Pu system that the η phase is formed by a ternary peritectic reaction different from the U-Pu-Ti system,

²Portnoff, A. Y., and Calais, D., La solubilité du plutonium dans le thorium α , J. Less-Common Metals 9, 74-76 (1965).

Fig. 3. Solid-state Reactions in the Intermediate Temperature Range for the Th-U-Pu System

for which η is formed by a Class II reaction, and also from the U-Pu-Zr system, for which η enters the ternary system from the binary U-Pu side. On the other hand ζ is formed in all of these systems, as well as in the U-Pu-Mo system, by a peritectic Class III reaction inside the ternary system.

DTA and dilatometry showed that the Th-20 w/o U-10 w/o Pu alloy undergoes two transformations. At 738°C the (U,Pu) phase transforms reversibly: $\gamma \rightleftharpoons \beta$, and at 590°C: $\beta \rightleftharpoons \alpha$. Weak DTA arrests at 681 and 690°C indicate that the η phase temporarily enters the structure as the alloy passes through a three-phase field. The same effects, though weaker still, are observed in the Th-10 w/o U-10 w/o Pu alloy, and confirm that both alloys are essentially two-phase alloys whose structure consists of a (U,Pu) phase with hardly any thorium in solid solution

and a thorium matrix phase with possibly 5 to 8% plutonium and less than 0.5% uranium. Only traces of the undesirable ζ phase should be present.

d. Phases and Transformations in U-Pu-Ti System. Differential thermal analysis (DTA) and electron microprobe analysis of U-Pu-Ti ternary alloys confirmed that plutonium may replace uranium in the binary U_2Ti compound and that the compound exists in the ternary system as $(U,Pu)_2Ti$. This corroborates recent work at Los Alamos.³ The electron microprobe analysis was performed with several alloys annealed in the $(U,Pu)_2Ti + \gamma$ two-phase region.

The equilibrium transformation temperature of 898°C as determined by Knapton⁴ for the transformation of the bcc gamma solid solution to U_2Ti in the binary system was confirmed by DTA, as shown in Fig. 4. Transformation temperatures were determined at various rates from 0.5 to 25°C/min in specimens of two different sizes, a 0.1-g micro specimen and a macro specimen of about 16 g. As shown in Fig. 4 the equilibrium transformation temperature is close to the peak temperature indicated by DTA on slow heating.

³Quarterly Status Report Advanced Reactor Technology for the Period Ending January 31, 1965, LA-3244.

⁴Knapton, A. G., The System U-Ti, J. Inst. Metals 83, 497-504 (1954/55).

Fig. 4. Formation of U_2Ti from Gamma-phase Peak Temperatures by Differential Thermal Analysis

The addition of plutonium to an alloy containing U-33.3 a/o Ti stabilizes the γ phase to lower temperatures. In alloys containing 16.7 and 23.3 a/o plutonium, the gamma phase is stabilized to 817 and 782°C, re-

spectively. A lower temperature transformation occurs at 604°C, which indicates that the solubility of plutonium in the $(U,Pu)_2Ti$ compound is less than 23.3 a/o. This result differs from the solubility of plutonium in the compound as determined at Los Alamos.

Fig. 5. Section through U-Pu-Ti Ternary System at 33.3 a/o Ti

A tentative isoplethal section at 33.3 a/o Ti is shown in Fig. 5. Transformation temperatures corresponding to the start of the decomposition of the gamma phase are shown as open circles. The completion of the transformation to $(U,Pu)_2Ti$ is experimentally difficult to determine and is shown as a dashed line. In the U-23.3 a/o Pu-33.3 a/o Ti alloy two separate transformations were found, at about 600 and 585°C, as shown in Fig. 5.

An analysis of the solid-state reactions in the uranium corner of the U-Pu-Ti system is given in Fig. 6. This analysis accounts for the six binary solid-state transformations in the U-Pu and U-Ti binary systems, and is supported by dilatometric and DTA data for ternary alloys. Three major reactions at 655, 650, and 575°C were found to occur in alloys of interest as fast reactor fuels. The highest-temperature isothermal reaction, a Class II reaction (following the notation of Rhines⁵), involves the

⁵Rhines, F. N., Phase Diagrams in Metallurgy, McGraw-Hill Book Co., Inc., New York (1956).

transformation $\beta + \gamma \rightarrow (U,Pu)_2Ti + \eta$. The ξ phase forms peritectoidally by a Class III reaction at 650°C. The lowest-temperature isothermal reaction occurs at 575°C, that is, $(U,Pu)_2Ti + \beta \rightarrow \alpha_U + \xi$. After the reaction at 575°C occurs, the three-phase region $(U,Pu)_2Ti + \alpha_U + \xi$ is stable to room temperature. The two Class II reactions at 600 and 585°C are based on a limited amount of data and are subject to revision as additional alloys in this region are investigated.

Fig. 6. Solid-state Reactions in Uranium Corner of U-Pu-Ti System

e. Mechanical Properties of U-Pu Alloys. Data on the mechanical properties of injection-cast uranium and uranium-plutonium alloys were obtained to help understand the properties of the ternary alloys. Alloys with 0, 5, 10, 15, and 20 a/o plutonium were fabricated and their elastic moduli measured. In Fig. 7 is shown how the elastic modulus varies at room temperature for this series of alloys. The modulus drops off rapidly from about $23 \times 10^3 \text{ kg/mm}^2$ for unalloyed uranium to about $13 \times 10^3 \text{ kg/mm}^2$ for a U-10 a/o Pu alloy, and changes very little from that point to 20 a/o plutonium.

The elastic modulus was expected to show a discontinuity for the transition from α to $\alpha + \xi$ U-Pu at 15 a/o plutonium. The composition

Fig. 7. Variation of Elastic Modulus with Plutonium in Uranium

U-20 a/o Pu has a ζ matrix. No such discontinuity was observed in going from the α matrix to the ζ matrix; rather a large change in modulus occurred by solution of plutonium in the α -phase region.

2. Jacket Development

a. Vanadium-base Alloys. A total of 66 m of 0.558-cm-OD x 0.041-cm wall V-15 w/o Ti-7.5 w/o Cr tubing was received from the Superior Tube Company, who drew the tubing to finished size from tube-reduced blanks furnished by ANL (see Progress Report for February 1966, ANL-7176, p. 27). The actual yield was about 25% higher than the estimated yield.

Eddy-current inspection of the tubing revealed only one short (2.5 cm long) defect in the total shipment. The tubes appeared excellent to visual examination and were within dimensional specifications. The vendor processed the tubing primarily by mandrel drawing and reported that they experienced no difficulty with the alloy. (This was the first commercial redraw experience with the V-15 w/o Ti-7.5 w/o Cr alloy.)

b. Creep of V-Ti-Cr Alloys. The effect of annealing temperature on the creep of V-15 w/o Ti-7.5 w/o Cr at 700°C and 30 kg mm⁻² stress is quite appreciable. The rupture life for a 900°C anneal is 24.5 hr compared to 270 hr for a 1250°C anneal.

This alloy has a short second stage of creep. An alloy with a lower titanium content and a higher chromium content (V-5 w/o Ti-15 w/o Cr) annealed at 1250°C shows a much longer second stage of creep. An internally pressurized (with helium) Nb-1 w/o Zr tube specimen tested at 700°C and 30 kg mm⁻² stress fractured in 231 hr. Under these temperature and stress conditions, an internally pressurized Type 304 stainless steel tube ruptured spontaneously.

3. Ceramic Fuels

a. Particulate Oxide-fuel Test Specimens. A fuel of interest is UO₂-PuO₂ in the form of a vibratorily compacted matrix of angular particles. During the reporting period, fabrication of 24 rods loaded with UO₂-20 w/o PuO₂, which was processed at Battelle Northwest by the Nu-Pak technique, was completed. The rods were jacketed in Type 304 stainless steel, Hastelloy-X, or V-20 w/o Ti, and have either a rod or spring fuel retainer. All rods supplied were gamma radiographed after loading and before encapsulating. Table VI sums the pertinent characteristics of the completed rods.

TABLE VI. Particulate Oxide Fuel Test Specimens

Specimen Number	Fuel Weight (g)	Fuel Volume (cc)	Packing Fraction ^(a) (%)	Theoretical Density (%)	Jacket Material	Retainer Form	Column Quality ^(b)
SOV-1	85.05	9.71	81.9	79.9	304 SS	Rod	B
SOV-2	85.05	9.72	81.8	79.8	304 SS	Rod	B
SOV-3	88.55	9.72	85.2	83.1	304 SS	Rod	B
SOV-4	87.35	9.70	84.2	82.1	304 SS	Rod	B
SOV-5	88.65	9.70	85.5	83.4	304 SS	Rod	B
SOV-6	88.61	9.71	85.4	83.3	304 SS	Rod	B
SOV-7	90.70	9.72	86.8	85.1	304 SS	Rod	B
SOV-8	90.55	9.77	86.2	84.5	304 SS	Spring	B
SOV-9	91.00	9.72	87.1	85.4	304 SS	Rod	A
SOV-10	84.74	9.73	81.4	79.4	304 SS	Spring	A
SOV-11	85.36	9.74	81.9	79.9	304 SS	Spring	B
HOV-1	92.20	10.51	81.7	80.0	Hastelloy-X	Spring	B
HOV-2	92.17	10.52	81.6	79.9	Hastelloy-X	Rod	B
HOV-3	92.20	10.52	81.6	79.9	Hastelloy-X	Spring	C
HOV-4	92.21	10.51	81.7	80.0	Hastelloy-X	Rod	C
HOV-15	91.75	10.48	81.8	79.8	Hastelloy-X	Rod	A
HOV-6	92.18	10.50	82.2	80.1	Hastelloy-X	Rod	B
HOV-8	97.81	10.50	86.7	85.0	Hastelloy-X	Spring	B
HOV-9	97.59	10.51	86.4	84.7	Hastelloy-X	Rod	B
HOV-10	97.70	10.50	86.5	84.8	Hastelloy-X	Rod	B
HOV-11	92.05	10.52	81.8	79.8	Hastelloy-X	Rod	B
TVOV-1	82.33	9.80	76.8	74.4	V-20 w/o Ti	Spring	C
TVOV-2	84.19	9.78	78.5	76.1	V-20 w/o Ti	Spring	B
TVOV-3	85.77	9.78	80.0	78.8	V-20 w/o Ti	Spring	A

^aPacking Fraction = (bulk volume - void volume)/(bulk volume).

^bA: completely uniform; B: of intermediate uniformity; C: acceptably uniform.

In the past feed preparation for vibratory compaction processes was carried out by laboriously crushing materials by hand. This procedure was found to be time-consuming, inefficient, and tedious. Investigations into commercially available, laboratory-type jaw crushers revealed them to be large and cumbersome from the standpoint of glovebox operation. In addition, control over jaw spacing is crude, and hourly capacities upward of 10 to 20 times our normal criticality limit are typical. During this reporting period a jaw crusher was built to conform more nearly to our requirements. About 1 kg of Nu-Pak UO_2 - PuO_2 was fed through the crusher as a trial. An attempt was made to attain as high a yield as possible in the size fractions 10/18, 20/30, and 35/50 mesh. The yields in each fraction were as follows: 10/18 mesh - 59.0%, 20/30 mesh - 14.1%, 35/50 mesh - 12.2%, and <50 mesh - 14.6%. These results exceeded our expectations and will be checked.

b. Compatibility with Carbides

(i) Hyperstoichiometric (U,Pu) Carbide. Compatibility couples containing Hastelloy-X and Type 304 stainless steel versus hyperstoichiometric (U,Pu) carbides, after heat treatment at 700°C for 42 days, have been examined metallographically. The carbide contained 5.25 w/o equivalent carbon. A reaction zone having an average width of 4 μ was found in the

Hastelloy-X specimen. This zone is identical in appearance to that found in a specimen heat treated at 800°C for 42 days (average width = 8 μ) which was found to contain the major elements of Hastelloy-X as well as U and Pu. In the Type 304 stainless steel specimen, a band having an average width of 4 μ was found, as compared to the 24 μ average width found at 800°C for 42 days. This band is distinctly seen metallographically after etching, but electron microprobe analysis shows only a gradual carbon concentration gradient extending beyond the differently etched band. For this reason, it appears that, although the band seen metallographically is indicative of a certain carbon content being reached in the stainless steel, the width of the band, per se, is of uncertain value as far as the physical properties of the steel are concerned.

(ii) Hyperstoichiometric (U,Pu) Carbides with Metal Additions.

The effects of certain metal additions to hyperstoichiometric uranium-plutonium carbides are being studied as a possible means of improving compatibility with cladding materials, especially in the presence of liquid sodium. (U,Pu) carbides containing approximately 5.3 w/o equivalent carbon, to which chromium, molybdenum, and tungsten, in amounts of 3.0, 5.5 and 10.5 w/o, respectively, were added, have been examined metallographically and with an electron microprobe. The compositions containing molybdenum and tungsten (fired at 1900°C for 2 hr) were two-phase, the matrix being (U,Pu)C and virtually all the molybdenum and tungsten concentrated in the second phase. The composition containing Cr (fired at 1700°C for 3 hr) appears to be more complex, as the microprobe results show the "second phase" to be of various compositions. Separate compositions containing differing amounts of chromium were found in this phase. The analysis of the actual compositions of these phases has not been completed. No evidence of (U,Pu) sesquicarbide or dicarbide phases was found in any of the samples.

4. Irradiation of Materials for Fast Reactors

a. Examination of Irradiated EBR-II Fuel Cladding. Additional tube-burst tests (see Progress Report for April 1966, ANL-7204, pp. 19-20) have been made at 900 and 1000°C on irradiated Type 304L stainless steel tubing from EBR-II fuel rods. Five specimens from the jacket of fuel rod C139-E37 and five from fuel rod C139-E47 have also been tested, respectively, at 900 and 1000°C.

At 900°C the maximum tangential rupture strength as calculated from the bursting pressure was $15,200_{-500}^{+900}$ psi for the five specimens along the length of the jacket. Similarly, at 1000°C the strength was $10,000_{-600}^{+300}$ psi. Compared to unirradiated specimens, the maximum tangential rupture strength for the irradiated specimens was smaller for tests at 800, 900, and 1000°C, as shown in Table VII.

TABLE VII. Effects of Irradiation on the High-temperature Tangential Rupture Strength of Type 304 Stainless Steel Tubing

Test Temperature (°C)	Avg Max Tangential Strength (psi)	
	Unirradiated	Irradiated
800	28,500	23,100
900	20,100	15,200
1000	12,400	10,000

In tests made at 700°C and below the irradiated specimens were stronger than the unirradiated.

Uniform elongation as determined from diameter measurements after burst testing decreased from 2.6 to 5.1% at 900°C to 1.5 to 4.0% at 1000°C. Comparison with previously reported data indicates that the uni-

form elongation is a minimum at a test temperature of 400 to 500°C and that there is a relative maximum at about 800°C.

b. Uranium-Plutonium Alloys. Twenty full-length (14 in.) metal fuel rods are undergoing irradiation in EBR-II. Eighteen are U-Pu-Zr alloys jacketed in either V-20 w/o Ti, Type 304 stainless steel, Type 316 stainless steel, Hastelloy-X, or Hastelloy-X-280 tubings. These rods have maximum jacket temperatures of 630°C and at present have reached a calculated maximum 3.4 a/o burnup. Two additional rods, fueled with U-Pu-Ti alloy and jacketed in V-20 w/o Ti alloy tubing, are being irradiated at maximum jacketing temperatures of 540°C and have attained a maximum burnup of 3.2 a/o. Additional details on the design of the fuel rods and the irradiation conditions are summarized in Table VIII.

TABLE VIII. Status of Metal Fuel Irradiations in Progress

Test Reactor	Capsule or S/A No.	Specimen Number	Design Parameters					Operating Conditions			
			Fuel Composition (w/o)	Effective Density (%)	Cladding Composition (w/o)	Cladding OD (in.)	Cladding Thickness (in.)	kW/cc ^(a)	Max Cladding Temp (°C)	Burnup to Date	
										a/o (U + Pu)	fiss/cc x 10 ^{-20(a)}
CP-5	CP-45	IN15	U-19 Pu-14 Zr	66	V-20 Ti	0.208	0.015	2.6	640	8.6	18.6
EBR-II	XG06	ND23	U-15 Pu-10 Zr	66	V-20 Ti	0.209	0.016	1.7	540	3.2	7.6
EBR-II	XG05	ND24	U-15 Pu-10 Zr	66	V-20 Ti	0.209	0.016	1.7	535	2.9	7.0
EBR-II	XA07	ND28	U-15 Pu-9 Zr	75	304 SS	0.208	0.021	2.0	630	3.4	9.5
EBR-II	XA07	ND41	U-15 Pu-9 Zr	75	304 SS	0.208	0.021	2.0	625	3.3	9.2
EBR-II	XA07	ND32	U-15 Pu-9 Zr	75	316 SS	0.196	0.015	2.0	605	3.1	8.7
EBR-II	XA07	ND43	U-15 Pu-9 Zr	75	Hastelloy-X	0.196	0.015	2.0	615	3.2	9.0
EBR-II	XA07	ND25	U-14 Pu-12 Zr	76	304 SS	0.208	0.021	1.9	600	2.9	7.5
EBR-II	XA07	ND27	U-14 Pu-12 Zr	76	304 SS	0.208	0.021	1.9	605	3.0	7.8
EBR-II	XA07	ND26	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	1.8	590	2.8	7.3
EBR-II	XA07	ND29	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	1.8	595	2.9	7.5
EBR-II	XA07	ND30	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	1.9	615	3.2	8.3
EBR-II	XA07	ND31	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	1.9	610	3.1	8.0
EBR-II	XA07	ND33	U-14 Pu-12 Zr	76	Hastelloy-X	0.196	0.015	1.9	605	3.1	8.0
EBR-II	XA07	ND34	U-14 Pu-12 Zr	76	Hastelloy-X	0.196	0.015	1.9	610	3.1	8.0
EBR-II	XA07	ND35	U-14 Pu-12 Zr	76	Hastelloy-X	0.196	0.015	1.9	615	3.2	7.5
EBR-II	XA07	ND37	U-14 Pu-12 Zr	66	Hastelloy-X-280	0.208	0.015	1.8	610	3.2	8.0
EBR-II	XA07	ND39	U-14 Pu-12 Zr	66	Hastelloy-X-280	0.208	0.015	1.8	610	3.2	8.0
EBR-II	XA07	ND44	U-14 Pu-12 Zr	66	Hastelloy-X-280	0.208	0.015	1.8	600	3.1	7.7
CP-5	CP-44	IN14	U-15 Pu-10 Ti	69	V-20 Ti	0.203	0.015	2.4	560	7.5	16.3
EBR-II	XG05	NC17	U-15 Pu-10 Ti	63	V-20 Ti	0.209	0.016	1.6	540	3.0	6.6
EBR-II	XG06	NC23	U-15 Pu-10 Ti	63	V-20 Ti	0.209	0.016	1.6	540	3.2	7.0
CP-5	CP-41	4N10	U-10 Pu-10 Fz	77	V-20 Ti	0.196	0.016	1.9	530	7.1	20.8
CP-5	CP-41	5N11	U-10 Pu-10 Fz	79	V-20 Ti	0.193	0.016	2.0	530	7.1	21.3
CP-5	CP-41	6N12	U-10 Pu-10 Fz	84	V-20 Ti	0.189	0.016	2.1	530	7.1	22.7
CP-5	CP-41	1N7	U-15 Pu-10 Fz	77	V-20 Ti	0.196	0.016	1.9	530	7.1	20.8
CP-5	CP-41	2N8	U-15 Pu-10 Fz	79	V-20 Ti	0.193	0.016	2.0	530	7.1	21.3
CP-5	CP-41	3N9	U-15 Pu-10 Fz	84	V-20 Ti	0.189	0.016	2.1	530	7.1	22.7
CP-5	CP-43	1N13	U-15 Pu-10 Fz	73	V-20 Ti	0.199	0.015	2.3	540	6.6	18.4
CP-5	CP-50	1N16	Th-20 U	75	V-20 Ti	0.196	0.015	1.9	620	0.63	0.29
CP-5	CP-50	4N19	Th-20 U	75	V-20 Ti	0.196	0.015	1.9	620	0.63	0.29
CP-5	CP-50	2N17	Th-10 Pu-10 U	75	V-20 Ti	0.196	0.015	2.0	660	0.73	0.34
CP-5	CP-50	5N20	Th-10 Pu-10 U	75	V-20 Ti	0.196	0.015	2.0	660	0.73	0.34
CP-5	CP-50	3N18	Th-10 Pu-20 U	75	V-20 Ti	0.196	0.015	1.9	660	0.66	0.49
CP-5	CP-50	6N21	Th-10 Pu-20 U	75	V-20 Ti	0.196	0.015	1.9	620	0.66	0.49

^aBased on effective density.

Five instrumented, temperature-controlled capsules, which contain several different types of experimental alloy fuels, are in the CP-5 reactor.

U-Pu-Zr alloy is being irradiated in CP-5 in V-20 w/o Ti alloy tubing and has reached a calculated 8.6 a/o burnup at a maximum jacket temperature of 640°C. U-Pu-Ti alloy, also jacketed in V-20 w/o Ti alloy tubing, has attained a calculated 7.5 a/o burnup at maximum clad temperatures of 560°C.

U-Pu-Fs alloy specimens being irradiated in CP-5 are designed to study fuel-density parameters which have a direct bearing on maximum attainable burnup. These specimens have reached a maximum burnup of 7.1 a/o at maximum cladding temperatures of 530°C.

Six specimens of vibrationally compacted U-Pu-Fs metal powders were removed from the reactor after a calculated 3.8 a/o burnup. A neutron radiograph of the specimens while still in the capsule showed all claddings to be intact. A radiograph had been taken of these specimens at 1.3 a/o burnup; the measured fuel column length had decreased by an average of 3%. Four of the specimens had segmented into two or three separate sections, which is attributed to the ratcheting effect of the sodium bond which had been intermixed with the powders to enhance the thermal conductivity of the compact. The latest radiograph showed that the fuel lengths had decreased an average of 8% and separation between segments had decreased. The initial compacted theoretical density was only 57%. The postirradiation examination will include fission gas recovery and analysis. The resulting data will be compared with calculated results on the probability of fission gas escape by recoil.

Capsule CP-48 containing the first series of six U-Pu-Zr specimens jacketed in iron- and nickel-base alloy tubing was removed from the reactor after a calculated 1.7 a/o burnup. The postirradiation examination of three specimens jacketed in Type 304 stainless steel, Type 316 stainless steel, and Hastelloy-X tubing failed to show any significant changes in length, diameter, or density. The specimens had operated at a maximum cladding temperature, indicated by thermocouples, of 600°C. The jackets were punctured; the fission gas recovered is being analyzed. Selected areas of the specimens were sectioned for metallographic studies. The examination of the fuel-jacket interfaces did not reveal any significant reaction between the fuel and jacket materials.

Three identical specimens in the same capsule were operated at maximum indicated cladding temperatures of 660°C. These specimens showed formation of a liquid phase between the fuel and cladding, which ultimately resulted in cladding failure.

c. Ceramic-fuel Irradiations in EBR-II. The status of the ceramic-fuel rods under irradiation in EBR-II is summarized in Tables IX and X. No new capsules were introduced during the month.

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

Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions			
		Effective Density (%)	Cladding Composition	Cladding OD (in.)	Cladding Thickness (in.)	kW/cc ^(a)	Max Cladding Temp (°C)	Burnup to Date	
								a/o (U + Pu)	fiss/cc x 10 ^{-20(a)}
XG05	SMV-2	84	304 SS	0.297	0.020	2.1	645	3.0	8.4
XG05	HMV-5	80	Hastelloy-X	0.297	0.015	2.2	670	3.1	8.2
XG05	NMV-11	84	Nb-1 w/o Zr	0.281	0.012	2.1	645	3.2	8.8
XO08	NMP-2	82	Nb-1 w/o Zr	0.281	0.012	1.6	545	1.6	4.3
XO08	NMV-4	80	Nb-1 w/o Zr	0.281	0.012	2.4	635	2.3	6.1
XO08	NMV-7	80	Nb-1 w/o Zr	0.281	0.012	2.1	605	2.1	5.6
XO08	NMV-12	86	Nb-1 w/o Zr	0.281	0.012	2.4	635	2.3	6.5
XO08	HMV-1	80	Hastelloy-X	0.297	0.015	2.1	640	2.1	5.4
XO08	HMV-4	80	Hastelloy-X	0.297	0.015	2.3	670	2.3	6.0
XO08	HWMP-1	82	Hastelloy-X + W	0.297	0.020	1.6	555	1.6	4.2
XO08	HWMP-1	83	Hastelloy-X + W	0.297	0.020	2.4	685	2.3	6.3
XO09	SMV-1	82	316 SS	0.306	0.024	1.6	570	1.6	4.4
XO09	SMP-1	80	316 SS	0.306	0.024	2.1	640	1.2	3.3
XO09	VMV-1	86	Vanadium	0.301	0.022	2.5	640	1.9	5.3

^aBased on effective density.

TABLE X. Status of UO₂-20 w/o PuO₂ Fuel Irradiations in Progress in EBR-II

Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions			
		Effective Density (%)	Cladding Composition	Cladding OD (in.)	Cladding Thickness (in.)	kW/cc ^(a)	Max Cladding Temp (°C)	Burnup to Date	
								a/o (U + Pu)	fiss/cc x 10 ^{-20(a)}
XO09	SOV-5	82	304 SS	0.297	0.020	1.7	555	1.6	3.3
XO09	SOV-6	82	304 SS	0.297	0.020	1.7	565	1.7	3.6
XO11	SOV-3	83	304 SS	0.296	0.019	1.9	610	1.0	2.0
XO11	HOV-4	80	Hastelloy-X	0.295	0.014	1.8	600	1.0	1.9
XO11	TVOV-1	77	V-20 w/o Ti	0.297	0.020	1.7	575	1.0	1.8
XO11	SOV-7	85	304 SS	0.296	0.019	2.0	630	1.0	2.0
XO11	SOV-1	80	304 SS	0.296	0.019	1.7	590	1.0	1.9
XO11	HOV-10	86	Hastelloy-X	0.295	0.014	1.9	615	0.93	2.0
XO11	HOV-15	80	Hastelloy-X	0.295	0.014	1.7	590	0.90	1.8

^aBased on effective density.

Group C-4 is presently being encapsulated for irradiation. This group consists of physically mixed UC and PuC powders, one rod of vibrationally compacted solid solution (U,Pu)C powder, and one rod of pellets of UC-20 w/o PuC.

d. Oxide-Cermet Irradiations in EBR-II. The status of three stainless steel-PuO₂ cermet specimens manufactured by Battelle Northwest Laboratories and prepared for irradiation by Argonne National Laboratory is shown in Table XI.

TABLE XI. Status of Cermet Fuel (Clad in Type 304 Stainless Steel) Irradiations in Progress in EBR-II

Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions			
		Fuel Composition (w/o)	Effective Density (%)	Cladding OD (in.)	Cladding Thickness (in.)	kW/cc ^(a)	Max Cladding Temp (°C)	Burnup to Date	
								a/o (U + Pu)	fiss/cc x 10 ^{-20(a)}
XO11	5P-9	SS-40 PuO ₂	98	0.301	0.015	0.88	495	1.3	1.0
XO11	5P-12	SS-27 PuO ₂	99	0.294	0.015	0.59	450	1.3	0.66
XO11	5U-14	SS-27 UO ₂	98	0.298	0.013	0.45	435	1.0	0.53

^aBased on effective density.

e. Fast-neutron Irradiation of Jacket Materials. The status and identification of the capsules, being irradiated in EBR-II, are given in Table XII. All specimens are in an argon-helium environment.

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

S/A No.	Capsule Number	Design Parameters			Operating Conditions	
		Cladding Composition	Type of Specimen	No. of Specimens	Max Specimen Temp (°C)	Exposure to Date (n/cm ²)
XA07	AS-9	V-20 w/o Ti	Tensile	16	590	2.5×10^{22}
XA07	AS-10	Hastelloy-X	Tensile	16	590	2.3×10^{22}
XA07	AS-11	304 SS	Tensile	16	590	2.3×10^{22}
XA08	AS-1	V-20 w/o Ti	Tube-burst	12	540	1.8×10^{22}
XA08	AS-2	V-20 w/o Ti	Tube-burst	12	540	1.8×10^{22}
XA08	AS-3	Hastelloy-X	Tube-burst	12	540	1.9×10^{22}
XA08	AS-4	Hastelloy-X	Tube-burst	12	540	1.9×10^{22}
XA08	AS-5	304 SS	Tube-burst	12	540	1.9×10^{22}
XA08	AS-6	V-20 w/o Ti	Tensile	16	580	2.0×10^{22}
XA08	AS-7	Hastelloy-X	Tensile	16	580	1.8×10^{22}
XA08	AS-8	304 SS	Tensile	16	580	1.8×10^{22}
XA08	AS-12	V-20 w/o Ti	Tensile	16	580	1.8×10^{22}
XO09	AS-14	V-20 w/o Ti	Tensile	13	670	1.5×10^{22}
XO09	AS-15	V-20 w/o Ti	Tensile	13	670	1.5×10^{22}
XO09	AS-27	304 SS	Tensile	13	670	1.6×10^{22}
		Hastelloy-X				
XO10	AS-16	V-20 w/o Ti	Tensile	13	500	5.9×10^{21}
XO10	AS-17	V-20 w/o Ti	Tensile	13	500	6.4×10^{21}
XO10	AS-18	V-20 w/o Ti	Tensile	13	500	5.6×10^{21}
XO10	AS-19	V-20 w/o Ti	Tensile	13	500	5.9×10^{21}
XO10	AS-20	V-20 w/o Ti	Tensile	13	500	6.5×10^{21}
XO10	AS-21	V-20 w/o Ti	Tensile	13	500	7.7×10^{21}
XO10	AS-22	Hastelloy-X	Tensile	13	500	6.7×10^{21}
XO10	AS-23	304 SS	Tensile	13	500	7.7×10^{21}
XO10	AS-24	304 SS	Tensile	13	500	6.5×10^{21}
XO10	AS-25	304 SS	Tensile	13	500	6.2×10^{21}
XO10	AS-26	304 SS	Tensile	15	500	6.3×10^{21}

5. Fuel for Zero-power Reactors

a. U-Pu-based Alloys. Some consideration has been given to the possibility of rolling the fuel plates for ZPPR, to determine whether or not rolling would produce anisotropic expansion characteristics in U-25 w/o Pu-2.5 w/o Mo alloy. A casting of this alloy was hot rolled at 650°C to 50% reduction, then machined into dilatometer specimens parallel and transverse to the rolling direction. The material was extremely brittle and broke repeatedly when attempts were made to turn specimens on a lathe. Dilatometer specimens were finally cut on an Elox machine (electrical discharge cutting) and only a final very light cut taken on the lathe.

The expansion characteristics are virtually the same in both the rolling and transverse directions. The data are given in Table XIII along with those of the as-cast alloy.

TABLE XIII. Expansion Characteristics of
U-25 w/o Pu-2.5 w/o Mo Alloy

	As-rolled		As-cast
	Rolling Direction	Transverse Direction	
Mean coefficient of expansion, $\times 10^6/^{\circ}\text{C}$, 25 to 200°C	14.2	15.3	15.4
Mean coefficient of expansion, $\times 10^6/^{\circ}\text{C}$, 200°C to trans- formation temperature	25.5	24.8	26.2
Transformation range, °C	545-620	545-620	540-630
Expansion, Δl	0.41%	0.41%	0.46%
Mean coefficient of expansion, $\times 10^6/^{\circ}\text{C}$, Transformation to 780°C	23.7	24.2	25.3

Compatibility tests were run for U-25 w/o Pu-2.5 w/o Mo and U-28 w/o Pu-2.5 w/o Mo against Type 304L stainless steel, at 600, 650, and 700°C, for one week. In both alloys there was a slight reaction between the specimen and stainless steel at 600°C (120 μ penetration of stainless steel into fuel, 15 to 20 μ reaction layer of fuel in stainless steel). At 650°C the stainless steel penetration into the fuel was much more severe, and at 700°C alloying and melting occurred.

These tests were conducted with both the stainless steel and fuel interfaces lapped to a fine finish and held together in a picture-frame clamp under pressure. Another type of compatibility test which will more closely simulate actual reactor conditions of the fuel and jacket interface is being designed.

b. ZPPR Fuel

(i) Purpose of Program. A program is being developed to test a method for making ZPPR fuel elements. A pilot-lot manufacturing technique was fixed and is being used to produce ZPPR fuel elements. Draft 2 of Specification No. PF-1600, Specification for Uranium-Plutonium-Molybdenum Alloy ZPPR Fuel Elements, was completed and sent to interested vendors for comment.

The pilot-lot process consists of (1) producing a uranium-molybdenum alloy by vacuum induction melting and casting, (2) producing precision bars of uranium-molybdenum-plutonium alloy by vacuum induction melting and casting, (3) machining of core plates from the castings, and (4) enclosing the core plates in leaktight stainless steel jackets.

(ii) Castings. Premelting of the uranium and molybdenum is advantageous from two standpoints: (1) the molybdenum is more soluble in uranium than in plutonium, and (2) the danger of losing plutonium by vaporization at the higher temperatures necessary to dissolve the molybdenum is eliminated. Additional advantages could be realized in a production process because uranium-molybdenum alloys present a lesser problem as regards criticality and accountability, and need not be contained in a glovebox.

The uranium-molybdenum binary alloy was melted in a magnesia crucible and poured into graphite molds. The crucible and the molds are protected with an yttria wash. The charge was heated to a temperature over 1450°C and held for approximately 25 min while the molybdenum dissolved. The binary alloy poured near 1400°C.

The uranium-molybdenum binary alloy was then recharged along with additional uranium and with plutonium. Again magnesia crucibles and graphite molds, protected with yttria, were used. The alloy was heated to above 1250°C, held for 30 min, and poured at 1285°C. The castings are 9.5 in. long and have a cross section very close to that specified for the fuel cores.

(iii) Machining. Two machining techniques are available for cutting core plates from castings: (1) room-temperature machining with carbide tools, and (2) hot machining with high-speed steel tools. The former, which is preferred, requires the more expensive tools; however, it is faster, does not have temperature as a variable, and avoids the possibility of burning a glove.

One side of each casting is faced, to produce a reference side, on a small vertical mill with a carbide-tipped end-mill cutter. The castings are transferred to a heavy-duty mill and/or clamped on aluminum pads, and are parted with carbide-tipped slitting saws. The core slugs are cut approximately 0.030 in. oversize.

The two ends of the core slug are faced with a carbide-tipped end-mill cutter. The length of the core slug is measured and recorded, as is the squareness and parallelism of the end surfaces. The core slug is weighed and the amount of metal to be removed from the remaining side is calculated. This amount is removed by end milling. All edges are chamfered and the completed core plate is weighed.

(iv) Element Assembly: Element assembly consists of the following operations: (1) one end plug is welded into the jacket sleeve making a jacket can, (2) the core plate is cleaned and weighed, (3) the core plate and second end plug are placed in the jacket, and (4) the end plug is welded to the jacket sleeve.

The technique for welding the bottom end plug to the jacket is similar to that for closure welding. The same TIG welder, welding fixture, and chills are used; however, the first weld is not made under reduced pressure and no set force is applied to the end plug.

Core-plate cleaning is extremely important. Each core plate is scrubbed with a stainless steel brush and wiped with alcohol-moistened tissues to remove foreign and particulate matter. Particulate matter such as oxide or pieces of core alloy which remain on the plates will contaminate the jacket mouth when the core is placed in the jacket.

The jacket can, with its identification number etched on the side, is placed in the welding fixture. This fixture supports the material being welded and chills it. An aluminum funnel is placed in the jacket sleeve to protect the jacket mouth while the core is being loaded.

The closure welds are made by means of a tungsten-electrode, inert-gas-shielded, line welder at a pressure of one-third to one-half atmosphere. The welder is located in a helium-filled glovebox and the welding gas is a helium-argon mixture. Both the jacket sleeve and the end plug are chilled and a 40 pound compressive load is placed on the assembly during welding to insure contact between the core plate and end plugs.

(v) Jacket Hardware. The ZPPR jacket hardware was delivered during this reporting period. Each jacket consists of three pieces: one short length of rectangular tubing, termed a jacket sleeve, and two short lengths of rectangular wire, termed end plugs. All are AISI Type 304 stainless steel.

The sleeves are made from sheet stock. The sheet is sheared and rolled to form 9.5-in.-long tubes. The tubes are welded by the tungsten-electrode, inert-gas-shielded arc welding process using a controlled-arc line welder. A steel bar is placed inside the tube and the tube is die flattened to a cross section that is roughly rectangular. The corners are die-formed in a press brake and the tubes are annealed. Final size is achieved by expanding the tube within a die by means of wedges.

The 9.5-in.-long rectangular tubes are cut to the desired length on an abrasive cutoff wheel. The ends are finished by grinding.

The raw material for the end plugs is precision drawn square wire. The wire is cut with an abrasive saw, and the short lengths are "ganged" for finishing on a milling machine.

All hardware components are cleaned by vapor degreasing and detergent washing.

(vi) Element Inspection. Each element is surveyed, decontaminated if necessary, leak tested, weighed, and inspected. Weighing and physical inspection are performed in a conventional manner. Surveying is carried out in a helium glovebox with a gas-filled proportional counter. Leak testing is a duplex process which utilizes a helium mass spectrometer to find small leaks and a bubble test to find larger leaks.

6. Corrosion Studies

a. Refractory-metal Alloys for Service in Oxygen-contaminated Sodium. After 86 days of exposure to sodium (at 650°C; flowing at 6.1 m/sec; containing oxygen to the extent of 10-12 ppm), samples of V-20 w/o Ti are still in excellent condition. Total weight gain (dissolved oxygen - no external film) is 0.75 mg/cm². This value is still close to the empirical relationship (see Progress Report for May 1966, ANL-7219, p. 14, in which the exponent of t was incorrectly printed as 0.278)

$$\Delta W(\text{mg/cm}^2) = 0.28 t^{0.21} (\text{days}).$$

After 59 days exposure as above, V-15 w/o Ti-7.5 w/o Cr samples are also in excellent condition. Total weight gain is 0.45 mg/cm, only about 65% of the weight gain of V-20 w/o Ti at the same exposure.

Some alloys in sheet form were exposed to sodium in the static-refreshed system for one week. The results are shown in Table XIV. Oxygen content was 18 ppm at the end of the exposure.

TABLE XIV. Corrosion of Vanadium-base Alloys in Sodium at 650°C

Alloy (compositions in weight percent)	External Loss (μ per side)
V-5 Cr (50% cold rolled)	11
V-15 Ti-7.5 Cr-1 Y (50% cold rolled)	25
V-40 Ti-5 Cr (50% cold rolled)	18
V-20 Ti (annealed)	16

Levitation-melted castings of alloys of the V-Cr-Al ternary type were also exposed for seven days under similar conditions. The results are shown in Table XV.

TABLE XV. Experimental Castings Exposed to Sodium at 650°C

Alloy (compositions in weight percent)	Weight Loss (mg/cm ²)
V-25 Cr-10 Al	1.29
V-20 Cr-10 Al	2.40
V-20 Cr-15 Al	0.46
V-20 Ti (annealed sheet control)	9.70

It is apparent that the corrosion resistance increases as both the chromium or aluminum content is increased. These samples will be exposed for a longer period to see if the marked improvement over V-20 w/o Ti can be maintained.

b. Effect of Impure Sodium Environment on Creep Properties of V-20 w/o Ti. Stressed tensile specimens of V-20 w/o Ti were exposed to flowing (at 15 cm/sec) sodium at 650°C for seven days in a dynamic loop. Subsequent creep tests conducted in vacuum (pressure $< 10^{-6}$ mm Hg) at 650°C and 15 kg/mm² stress indicated that this sodium exposure resulted in degradation of the creep resistance.

The sodium used is thought to have contained about 20 ppm oxygen. This oxygen concentration is higher than would normally be expected in a fast power reactor. Similar tests will, therefore, be conducted in sodium of higher purity (about 10 ppm oxygen).

The exposed samples were examined in an effort to learn the cause of the degradation of the creep properties. Microhardness traverses were made of the hardened cases of two tensile specimens, R7 and R9. Similar measurements were also made on other V-20 w/o Ti coupons which had been exposed in 650°C sodium for seven days in refreshed static autoclaves; coupons C3, C15, and C24 were exposed to sodium of 45, 42, and 16 ppm oxygen, respectively. The results are given in Table XVI. The case microhardness profiles for specimens R7 and R9 are in the same range as those of coupons C3 and C15, and all are in a higher range than that of coupon C24 (at distances from the surface of about 25 μ or less). Thus, if case microhardness of V-20 w/o Ti alloy is related to the oxygen content in the test sodium, one may suspect that oxygen content of the sodium in the dynamic loop was higher than the reported 20 ppm. We may be able to check this conclusion at a later date when we have completed the testing of the additional tensile specimens in sodium containing 10 ppm oxygen.

TABLE XVI. Results of Examination of V-20 w/o Ti Exposed to Sodium at 650°C for Seven Days

Property	Sample No. and Type					
	R7 Tensile	R9 Tensile	C24 Coupon	C15 Coupon	C13 Coupon	R11 Tensile-Unexposed
Oxygen Concentration in Sodium (ppm)	20 (?)	20 (?)	16	42	45	-
Case Microhardness, KHN _{50g}						
At 20 μ from edge	1505	1478	1187	1465	1598	231
At 25 μ from edge	1265	1293	1138	1012	1438	253
At 30 μ from edge	760	794	880	610	1109	240
At 35 μ from edge	377	391	400	306	670	244
At 40 μ from edge	296	279	287	297	325	-
At 50 μ from edge	275	277	244	292	300	249
At 100 μ from edge	268	263	238	247	242	249
Surface Chemistry						
Nitrogen at surface, ppm	444*	-	-	-	406**	243
Nitrogen in core, ppm	190/206*	-	-	-	213/214**	240/238

*Analysis of combined specimens R7 and R9.

**Specimens from another run under similar test conditions.

According to Mathews *et al.*,⁶ oxygen, carbon, and nitrogen are effective strengtheners of vanadium. Nitrogen is the most potent, being 1.5 times more effective than carbon and oxygen. Nitrogen concentrations in the case and core are also shown in Table XVI. The hardness (DPH in kg/mm²) of annealed vanadium is empirically related to the oxygen equivalent (O_e) by: $DPH = 40 + 600 O_e$, where $O_e = 1.30 (\% C) + 1.50 (\% N) + 1.0 (\% O)$. In our case, carbon pickup by the V-20 w/o Ti alloy is not expected to be very important since the test equipment is constructed of an effective carbon-getter material, 18-8 austenitic stainless steel. As to nitrogen pickup, there is no significant difference in surface nitrogen concentration between the tensile specimens and the specimens exposed in the refreshed static autoclaves. Thus, the assumption that the increased case microhardness of the V-20 w/o Ti tensile specimens and coupons C15 and C3 (as compared to C24) is related to the oxygen content in the test sodium seems reasonable.

It is interesting to note that the nitrogen concentration at the surface of the specimens exposed to sodium is about twice that in the core. This suggests either (1) nitrogen pickup from test sodium or (2) internal migration of nitrogen outwards to the surface. Consideration of the limited analytic data suggests that pickup from the sodium is the more important factor.

The surface concentration of nitrogen was obtained by immersing the specimens in concentrated hydrofluoric acid for 5-15 min and then analyzing the acid solution for nitrogen by the Kjeldahl method. The balance of the samples were subsequently dissolved in new acid solution and similarly

⁶Mathews, D. R., Keith, G. H., and Loria, E. A., Effect of Interstitial Impurities on the Mechanical Properties of Electro-refined Vanadium at Low Temperature, Bureau of Mines Report 6637 (1965).

TABLE XV. Experimental Castings Exposed to Sodium at 650°C

Alloy (compositions in weight percent)	Weight Loss (mg/cm ²)
V-25 Cr-10 Al	1.29
V-20 Cr-10 Al	2.40
V-20 Cr-15 Al	0.46
V-20 Ti (annealed sheet control)	9.70

It is apparent that the corrosion resistance increases as both the chromium or aluminum content is increased. These samples will be exposed for a longer period to see if the marked improvement over V-20 w/o Ti can be maintained.

b. Effect of Impure Sodium Environment on Creep Properties of V-20 w/o Ti. Stressed tensile specimens of V-20 w/o Ti were exposed to flowing (at 15 cm/sec) sodium at 650°C for seven days in a dynamic loop. Subsequent creep tests conducted in vacuum (pressure $< 10^{-6}$ mm Hg) at 650°C and 15 kg/mm² stress indicated that this sodium exposure resulted in degradation of the creep resistance.

The sodium used is thought to have contained about 20 ppm oxygen. This oxygen concentration is higher than would normally be expected in a fast power reactor. Similar tests will, therefore, be conducted in sodium of higher purity (about 10 ppm oxygen).

The exposed samples were examined in an effort to learn the cause of the degradation of the creep properties. Microhardness traverses were made of the hardened cases of two tensile specimens, R7 and R9. Similar measurements were also made on other V-20 w/o Ti coupons which had been exposed in 650°C sodium for seven days in refreshed static autoclaves; coupons C3, C15, and C24 were exposed to sodium of 45, 42, and 16 ppm oxygen, respectively. The results are given in Table XVI. The case microhardness profiles for specimens R7 and R9 are in the same range as those of coupons C3 and C15, and all are in a higher range than that of coupon C24 (at distances from the surface of about 25 μ or less). Thus, if case microhardness of V-20 w/o Ti alloy is related to the oxygen content in the test sodium, one may suspect that oxygen content of the sodium in the dynamic loop was higher than the reported 20 ppm. We may be able to check this conclusion at a later date when we have completed the testing of the additional tensile specimens in sodium containing 10 ppm oxygen.

TABLE XVI. Results of Examination of V-20 w/o Ti Exposed to Sodium at 650°C for Seven Days

Property	Sample No. and Type					
	R7 Tensile	R9 Tensile	C24 Coupon	C15 Coupon	C13 Coupon	R11 Tensile-Unexposed
Oxygen Concentration in Sodium (ppm)	20 (?)	20 (?)	16	42	45	-
Case Microhardness, KHN _{50g}						
At 20 μ from edge	1505	1478	1187	1465	1598	231
At 25 μ from edge	1265	1293	1138	1012	1438	253
At 30 μ from edge	760	794	880	610	1109	240
At 35 μ from edge	377	391	400	306	670	244
At 40 μ from edge	296	279	287	297	325	-
At 50 μ from edge	275	277	244	292	300	249
At 100 μ from edge	268	263	238	247	242	249
Surface Chemistry						
Nitrogen at surface, ppm	444*	-	-	-	406**	243
Nitrogen in core, ppm	190/206*	-	-	-	213/214**	240/238

*Analysis of combined specimens R7 and R9.

**Specimens from another run under similar test conditions.

According to Mathews *et al.*,⁶ oxygen, carbon, and nitrogen are effective strengtheners of vanadium. Nitrogen is the most potent, being 1.5 times more effective than carbon and oxygen. Nitrogen concentrations in the case and core are also shown in Table XVI. The hardness (DPH in kg/mm²) of annealed vanadium is empirically related to the oxygen equivalent (O_e) by: $DPH = 40 + 600 O_e$, where $O_e = 1.30 (\% C) + 1.50 (\% N) + 1.0 (\% O)$. In our case, carbon pickup by the V-20 w/o Ti alloy is not expected to be very important since the test equipment is constructed of an effective carbon-getter material, 18-8 austenitic stainless steel. As to nitrogen pickup, there is no significant difference in surface nitrogen concentration between the tensile specimens and the specimens exposed in the refreshed static autoclaves. Thus, the assumption that the increased case microhardness of the V-20 w/o Ti tensile specimens and coupons C15 and C3 (as compared to C24) is related to the oxygen content in the test sodium seems reasonable.

It is interesting to note that the nitrogen concentration at the surface of the specimens exposed to sodium is about twice that in the core. This suggests either (1) nitrogen pickup from test sodium or (2) internal migration of nitrogen outwards to the surface. Consideration of the limited analytic data suggests that pickup from the sodium is the more important factor.

The surface concentration of nitrogen was obtained by immersing the specimens in concentrated hydrofluoric acid for 5-15 min and then analyzing the acid solution for nitrogen by the Kjeldahl method. The balance of the samples were subsequently dissolved in new acid solution and similarly

⁶Mathews, D. R., Keith, G. H., and Loria, E. A., Effect of Interstitial Impurities on the Mechanical Properties of Electro-refined Vanadium at Low Temperature, Bureau of Mines Report 6637 (1965).

analyzed for nitrogen. The results are, therefore, qualitative, and micro-probe techniques would be useful to evaluate the nitrogen concentration gradient in a more precise manner.

D. General Fast Reactor Fuel Reprocessing Development

1. Skull Reclamation Process

After melt refining of fuel pins discharged from the EBR-II reactor, a residue or skull containing uranium (about 5 to 10% of the melt refining charge) remains in the crucible. A skull reclamation process is being developed to recover and to purify the uranium contained in the skull. Although the skull reclamation process for the EBR-II fuel does not recover plutonium, much of the chemistry and technology being accumulated in the course of this process development has potential utility for the processing of plutonium-uranium oxide and carbide fuels. Currently, the skull reclamation process is being tested with nonirradiated uranium in remotely operated plant-scale equipment (~4 kg uranium per batch) comprising a reduction furnace and a retorting furnace.

The new series of runs (total of eight) in the reduction furnace has been completed (see Progress Report for March 1966, ANL-7193, p. 34). In the new series, the noble metal extraction step and the uranium oxide reduction step were conducted under more vigorous reaction conditions (higher temperatures, longer contact times, and faster mixing) than in earlier runs (see Progress Report for December 1965, ANL-7132, pp. 23-24). Uranium recoveries of about 93% (equivalent to about 99% overall recovery for the melt refining and skull reclamation processes) were achieved, and average fission product removals (% of charge) were satisfactory (molybdenum, 69%; ruthenium, 83%; cerium, 97%; zirconium, 60%).

One of the objectives of these runs was to examine the need of having fluoride ion in the salt for achieving high reduction of uranium oxide. The runs clearly demonstrated the need for about 10 m/o fluoride ion in the salt. With it, uranium oxide reductions exceeding 99% were achieved.

2. Processes for Fast Reactor Fuels

Pyrochemical processes are being developed for processing fast reactor fuels of the ceramic type (e.g., oxide or carbide). A process which utilizes liquid metal-molten salt extractions and salt-transport separations⁷ is currently being investigated for the separation of fissile and fertile

⁷In the salt transport separations, the fissile and fertile materials are selectively transferred from one liquid metal solution (donor) to another (acceptor) by cycling a molten salt phase which acts as a carrier between the two metal solutions. Noble and refractory metal fission products remain in the donor alloy.

constituents of the fuel and the fission products. The conceptual process flowsheet has been described previously (see Progress Report for May 1966, ANL-7219, pp. 21-24).

In the salt-transport separations step magnesium-copper alloy is being considered for use as the donor solvent and magnesium chloride for use as the salt carrier between the donor alloy and the acceptor alloys. Zinc-magnesium alloys are under consideration for use as the acceptor solvents. As the salt-transport step nears completion, the concentration of the uranium in the donor alloy decreases. To determine whether the uranium distribution coefficient between the molten transport salt and the liquid donor alloy is affected by the decreasing uranium concentration in the donor alloy, the uranium distribution coefficient (concentration in salt/concentration in alloy) between molten magnesium chloride and liquid Cu-6 w/o Mg alloy was measured at 750, 800, and 850°C. The composition of the Cu-6 w/o Mg alloy was fixed at the liquidus in the copper-magnesium system.⁸ At each temperature, the uranium concentration in the copper-magnesium alloy was varied from about 0.1 w/o to the saturated value (2.1 w/o at 750°C to 7.6 w/o at 850°C). The distribution coefficients were found to be independent of the uranium concentration in the metal phase and to have the following values:

Temp. (°C)	Distribution Coefficient (K_d)
750	0.22
800	0.29
850	0.42

Engineering investigations of the salt transport separations step were also continued. An additional salt-transport experiment was completed in which the partitioning of uranium from various refractory and noble metal fission product elements was investigated. To prepare the feed for the salt transport step, unirradiated UO_2 was reduced from suspension in molten 47.5 m/o $MgCl_2$ -47.5 m/o $CaCl_2$ -5 m/o CaF_2 at 830°C by liquid Cu-15 w/o Mg alloy containing added ruthenium, molybdenum,⁹ zirconium, and palladium metals. The salt containing the MgO reaction product was removed from the system via a transfer line. In the salt-transport step, the uranium in the molten Cu-5.5 w/o Mg donor alloy was transferred incrementally to a molten Mg-35 w/o Zn acceptor alloy by cycling liquid $MgCl_2$ between the two alloys. This was accomplished by

⁸To maintain a fixed copper-magnesium composition, a "metallurgical buffering" technique is employed. The technique makes use of the relationship between the liquidus temperature and composition in the copper-magnesium phase diagram. By starting with a two-phase alloy consisting of copper-magnesium solid solution and the liquid phase as set by the liquidus curve of the phase diagram at the operating temperature, a constant magnesium concentration in the liquid copper-magnesium alloy can be maintained. Subsequent magnesium transport to the copper-magnesium alloy merely results in more liquid phase of the same composition.

⁹Not appreciably soluble in the donor alloy.

having the molten metal alloys in adjacent vessels and then alternately pressuring a charge of MgCl_2 through a transfer line from one alloy to the other. In each vessel, the salt and metal phases were mixed to promote chemical equilibration, and then the phases were allowed to separate by settling. Analytical results showed that nearly 99% of the uranium had been transported to the magnesium-zinc acceptor alloy. The precipitated uranium product was then washed with Mg-30 w/o Zn alloy, after which the wash solution was removed. The washed uranium product was dissolved in a Zn-13 w/o Mg solution which was then retorted to recover the uranium. Analyses of the uranium-zinc-magnesium product solution showed the following overall removals of fission product elements: zirconium, 98.5%; palladium and molybdenum, 99.9%; ruthenium, 99.99%. The effectiveness of the wash step was indicated when analyses of the supernatant magnesium-zinc (acceptor alloy), the wash solution, and the uranium product solution showed that a single wash removed more than 90% of the palladium, molybdenum, ruthenium, and copper which had transferred with the uranium product. However, the zirconium that was transferred (~1.5%) with the uranium coprecipitated with the uranium and was not removed by the wash.

3. Pyrochemical Reduction of NpO_2

Low-temperature pyrochemical processes have been previously used to prepare uranium, plutonium, and thorium metal by the reduction of their oxides. A recent laboratory experiment shows that this method can be extended to the reduction of NpO_2 . About 2.3 g of neptunium oxide, suspended in molten 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl , were reduced with liquid Zn-10 w/o Mg alloy for 2 hr at 800°C and at a stirring speed of 800 rpm. About 99.8% of the neptunium oxide was reduced. The zinc-magnesium-neptunium alloy was then retorted to remove the zinc and magnesium, after which the neptunium was consolidated by arc melting.

E. Sodium Technology

1. Core Component Test Loop (CCTL)

This loop was formerly named the Dynamic Component Test Loop (see Progress Report for March 1966, ANL-7193, p. 40) and prior to that the Large FARET Loop. The new name reflects the recent interest of Battelle-Northwest (BNW) in using the facility to test mocked-up sections of the core configuration of the FFTF.

Meetings have been held at ANL with BNW personnel to formulate the scope of the proposed effort. A general agreement concerning the extent of the effort was reached. Methods of implementing the program and scheduling remain to be discussed.

2. Component and Materials Evaluation Loop (CAMEL)

The loop has been fully insulated and the thermocouples connected to their respective junction boxes. Final connections are being made to the digital data-acquisition system. The circuits in the data-acquisition system have been checked for proper operation. One defect was found and corrected.

The programmable X-Y plotter was received from the Hewlett Packard Corporation. This device has now been installed and also checked for operation.

II. GENERAL REACTOR TECHNOLOGY

A. Theoretical Reactor Physics

1. The ARC System

a. The MC² Code. The MC² code has undergone modifications and improvements in preparation for its inclusion in the ARC System. Some of these changes, indicated below, are intended to facilitate the interaction of the MC² library with the evaluated nuclear data file (ENDF/B). The development of the ENDF/B is being coordinated by the Brookhaven National Laboratory.

In the unresolved resonance treatment, the code now allows the number of degrees of freedom in the fission-width distribution to depend upon the particular resonant sequence involved. Also, first-order corrections due to overlap of uncorrelated sequences are now included in the calculation.

In the fine-group averaging treatment, the user may now select the weighting spectrum as either $1/E$, constant, or E . Also, in the interpolation procedure involved in the fine-group averaging, the code now permits up to 100 changes in the interpolation rule over the tabulated range. Thus, the first N points may be designated as linear on a $\ln \sigma$ versus $\ln E$ basis, the next N' points as linear on a σ versus $\ln E$ basis, etc.

A card-punching option has been implemented such that the user may obtain broad group-averaged macroscopic cross sections for the mixture and individual microscopic cross sections for each material. A small fundamental-mode code, called PØNE, has been written which will accept these cards. PØNE is useful for studying the effect of composition changes on spectra and reactivity relative to the reference composition problem from MC². This two-step process: (i) punching cards and (ii) use of PØNE, will not be necessary when the ARC System is operational. The ARC System is intended to eliminate just this sort of manual intervention in sequential problems.

Further improvements to MC² will include options to allow either a consistent P1 or B1 flux calculation to be performed. Also, the Legendre expansions for elastic scattering will be extended from the present 5 terms to the order of 20 terms consistent with recent work involving fitting angular distribution data for elastic scattering. The Legendre coefficient data will also be extended up to energies of the order of 10 MeV, and the code will include such data for all materials in the library. The user will, however, still have the option of selecting which materials are to receive the accurate Legendre expansion treatment.

b. Operational Testing. Formal testing of the operating system for ARC has begun. Various standard and nonstandard paths have been used to simulate examples of the types of tasks to be accomplished with the ARC System. For this purpose, pseudo-computational modules and a corresponding set of pseudo-interface quantities have been defined.

The initial testing of the ARC System is being performed at the System 360/Model 50 Computer at the IBM offices in Chicago, pending delivery of the Argonne machine at the end of July 1966.

B. High-temperature Materials Studies

I. Ceramics

a. (Th-U-Pu) Phosphides. Studies of the reactions of UAs (see Progress Report for May 1966, ANL-7219, p. 36) and of its solid solutions with UP have been made.

UAs powder heated in vacuum gave a very sharp X-ray powder pattern. The compound exhibited the NaCl-type structure and had a lattice constant of 5.779 ± 0.001 Å. A pellet of UAs heated in vacuum for 4 hr at 1800°C yielded a slightly diffuse powder pattern with a lattice constant of 5.777 Å. The small reduction of unit-cell size may be a result of selective loss of arsenic, resulting in a substoichiometric composition.

A melting point of $2450 \pm 30^\circ\text{C}$ was obtained for UAs. It is not known whether this represented the value for stoichiometric material; however, volatility losses were thought to be low.

Mixtures of UAs and UP were blended, pelletized, and fired in vacuum at 1800°C . X-ray analysis showed complete solid solubility of the two components. Preliminary lattice constants for a number of compositions are as follows:

Composition (% UP)	Lattice Constant (Å)
0	5.777
25	5.723
37.5	5.699
50	5.675
62.5	5.651
75	5.629
87.5	5.606

A plot of lattice constant versus composition yielded a smooth curve that showed a negative deviation from linearity (Vegard's law). Systems

exhibiting negative deviation are far less common than those showing positive deviation. This appears to be the first case observed with actinide NaCl-type compounds.

b. Thermal Stability of Plutonium Ceramics

(i) Gd₂O₃ Evaporation. Testing of the glovebox thermogravimetric analysis apparatus was concluded, as were Knudsen cell experiments on the evaporation of Gd₂O₃. The glovebox is now being leak-checked in preparation for use with plutonium.

It was reported (see Progress Report for April 1966, ANL-7204, p. 47) that a constant weight gain was observed with an empty Knudsen cell heated at high temperatures in vacuum. This weight gain was quite reproducible and weight-gain-rate measurements were made in the temperature range from 2450 to 2625°K in a vacuum of 2×10^{-6} Torr. Linear rates were observed for times as long as 18 hr.

The most plausible explanation for the weight gain is that tungsten is transferred from the tungsten-mesh heating element to the crucible by vapor transport. If it is assumed that the tungsten pressure in the system is that of tungsten at the temperature of the heating element, the expression for the crucible weight-gain rate is $R_0 = R_d - R_e$, where R_0 is the observed weight-gain rate, R_d is the rate of tungsten deposition from the element at temperature T_d , and R_e is the rate of evaporation of tungsten from the crucible at temperature T_e .

In order to calculate a weight-gain rate to compare with the observed rate, the temperatures of the cell and element were measured for numerous settings of furnace power. A plot of temperature versus set point showed a difference of about 60 to 70°K between element and cell temperatures in the range from 2373 to 2673°K. The temperatures obtained, combined with tungsten-vapor-pressure data, were used to calculate theoretical weight-gain of rates crucibles.¹⁰ The theoretical rate curve is shown as a solid line in Fig. 8. The scatter in the experimental data is mainly due to the fact that the rates were calculated from very small total weight changes (about one mg). Nevertheless, the good agreement between measured and calculated rates supports the idea of tungsten deposition.

Knudsen cell runs on the evaporation of Gd₂O₃ were made in the temperature range from 2413 to 2592°K. Two different cells were used with orifice diameters of 0.900 and 1.710 mm, respectively. Linear weight-loss rates were found in all of the runs. Calculations are being made on the data and the results will be presented in a future report.

¹⁰Dushman, S., Scientific Foundations of Vacuum Technique, 2nd Ed., John Wiley and Sons, Inc., New York (1962).

Fig. 8. Plot of Logs of Calculated and Experimental Weight-gain Rates vs. $1/T_e$

appears to change its characteristics during the time of the experiment. It is also expected that the higher furnace temperatures combined with the larger heat pulses will promote diffusion of the graphite into the specimen and change its physical properties.

Past experience with nuclear ceramic fuels has shown that tungsten and its carbide form a tightly adhering coating without undue reaction with the substrate. For this reason a method was developed for depositing a firmly adherent layer of tungsten carbide, tungsten metal, or a mixture of the two on specimens of various characteristics.

The apparatus is essentially a vertically mounted quartz tube with a furnace (capable of operation at 1000°C) located on the upper section. The lower section is wrapped with a low-temperature heater, and this part of the assembly can also be cooled in liquid nitrogen.

A means for evacuation as well as maintaining a flow of inert gas from the lower to the upper chamber and subsequent passage through a dry ice condenser into a fume-hood exhaust are provided. A yoke-shaped specimen holder is provided that can be adjusted vertically to travel from the upper high-temperature region to the low-temperature zone at the bottom of the reaction chamber.

c. Development of Thermal-diffusivity Rig for Ceramic Materials.

Thermal diffusivities and heat capacities of nuclear ceramics are being measured in order to determine heat transfer properties. This technique is now being extended to higher temperatures with the aid of a laser installation (see Progress Report for March 1966, ANL-7193, p. 65).

The front surface of a disc is heated by laser pulses above the furnace temperature and the subsequent heat rise of the back surface recorded on an oscilloscope. During the early stages a xenon flashtube was used to produce the heat pulse. Under these conditions a thin layer of graphite sprayed on the front surface adequately satisfied the boundary conditions of the experiment, i.e., high, uniform, and reproducible energy absorption and total opacity. The laser beam, because of its higher energy concentration, affects the durability of the graphite layer and

A run consists of the following sequence of events: first, a measured amount of tungsten carbonyl is placed in a test tube which is located in the bottom of the apparatus. Next, the disc to be plated is placed in the yoke, which is then positioned in the center of the furnace. The vessel is then sealed, purged, and evacuated. While the carbonyl is being cooled in liquid nitrogen to prevent premature dissociation, the specimen is heated in a stream of dry, high-purity gas to a temperature between 600 and 1000°C. The bottom is then warmed to approximately 200°C, whereupon the plating action begins. The specimen is lowered gradually until its temperature has dropped to between 250 and 400°C. The ratio of tungsten to tungsten carbide as well as the thickness of the deposit is controlled by the specimen temperature, flow rate of the gas, and duration of the plating process. A plating time of one hour will generally produce a layer of 0.5-mil thickness.

Satisfactory coatings have been plated on high-density spinel and arc-cast uranium carbide. The coatings appear to be resistant to handling, of uniform high emittance, and possess extreme opacity even in thin layers. The apparatus is ready for adaptation to glovebox use on plutonium-bearing ceramics.

2. Corrosion Studies

a. Dissolution Kinetics in Liquid-metal Systems. The feasibility of using internal-friction measurements for determining the interstitial impurity content of liquid metal-refractory metal systems (by measuring the impurity level in the refractory metal in equilibrium with the liquid metal) is being studied. The most attractive feature of this technique is the potential for accurate measurements at very low levels (i.e., a few ppm) of impurities.

Literature on internal friction indicates that the interstitial impurity content of body-centered cubic refractory metals is related to the height and breadth of internal-friction peaks.

Preliminary experimental results in tantalum at a frequency of 0.28 cps indicate internal-friction peaks at 135, 320, and 331°C. These temperatures agree quite well with values obtained from the literature which attribute the peaks to oxygen, nitrogen, and carbon, respectively. Results on a specimen which had been outgassed for 1.5 hr at a temperature of 2400°C and in a vacuum of 10^{-7} Torr indicated reduction in the peak heights (by more than a factor of ten) attributed to oxygen and nitrogen as compared with an "as received" tantalum sample. A significant oxygen peak was still detectable in the outgassed specimen, indicating some friction due to the small amount of oxygen remaining in the sample.

Investigation of the method is continuing to determine if it is the most suitable for the desired purpose.

3. Irradiation Testing

A summary of the irradiations of carbides and sulfides in MTR is shown in Table XVII. All capsules are instrumented. The cladding is Nb-1 w/o Zr.

TABLE XVII. Status of Ceramic Fuel Irradiations in Progress in MTR

Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions			
		Fuel Composition (w/o)	Effective Density (%)	Cladding OD (in.)	Cladding Thickness (in.)	kW/cc ^(a)	Max Clad Temp (°C)	Burnup to Date	
								a/o (U + Pu)	fiss/cc x 10 ^{-20(a)}
56-11	MV-2	UC-20 PuC	79	0.281	0.012	1.2	470	5.4	13.9
56-8	MV-3	UC-20 PuC	81	0.281	0.012	1.2	715	5.9	15.6
56-8	MV-5	UC-20 PuC	80	0.281	0.012	1.2	705	5.6	14.6
56-11	MV-6	UC-20 PuC	80	0.281	0.012	1.2	480	5.9	15.4
56-13	Z-4	UC-20 PuC	79	0.174	0.015	1.2	665	0.8	2.1
56-13	Z-5	UC-20 PuC	79	0.174	0.015	1.2	585	0.8	2.1
56-13	Z-7	UC-20 PuC	79	0.174	0.015	1.2	570	0.8	2.1
56-13	C-45	PuC	84	0.174	0.009	1.4	700	0.8	2.2
56-8	S-7	US	80	0.281	0.012	1.0	535	4.2	8.1
56-8	S-8	US	89	0.281	0.012	1.0	725	6.2	13.4
56-8	S-9	US	76	0.281	0.012	1.0	750	6.2	11.4
56-8	S-10	US	91	0.281	0.012	1.0	690	6.2	13.6
56-11	S-15	US	82	0.281	0.012	1.0	380	3.3	6.5
56-11	S-16	US	90	0.281	0.012	1.0	510	5.2	11.3
56-11	S-17	US	88	0.281	0.012	1.0	500	3.9	8.3
56-11	S-18	US	77	0.281	0.012	1.0	610	5.7	10.6

^aBased on effective density.

The postirradiation examination of the specimens in capsule ANL-56-9 (see Progress Report for April 1966, ANL-7204, pp. 48-49) is continuing. Burnup of specimen MV-4 was 4.2 a/o based on a technetium analysis. This is 20% higher than the estimated burnup of 3.5 a/o reported in ANL-7204. Metallographic examination at 1000X of the PuC fraction in specimen MV-4 did not resolve the structural features present in sufficient detail to enable identification.

C. Other Reactor Fuels and Materials Development

1. Nondestructive Testing

a. Determination of Elastic Moduli of High-temperature Materials by Ultrasonics. In the hope that the trailing pulses will be more easily discerned after the dimensions are changed, the rod of V-15 w/o Ti-7.5 w/o Cr (see Progress Report for May 1966, ANL-7219, p. 47) is being remachined.

In conjunction with the transducer development program, several pieces of sintered stainless steel and of sintered nickel were examined to determine their ultrasound transmission velocities for longitudinal waves. The lower-density nickel samples were very hard to measure.

b. Ultrasonic Instrument and Transducer Development. Evaluation of four transducer probes (see Progress Report for May 1966, ANL-7219, pp. 47-48) for reduction of unwanted echoes was completed. Following is a summary of the results:

1. ANL-25--2.22 cm long, 70.6% dense (-325 mesh), sintered nickel backing with an acoustic impedance (Z) of about $27 \times 10^6 \text{ kg/m}^2\text{-sec}$. The increased backing length (formerly 1.27 cm long) did not significantly reduce reflections from the back face of the backing member, and is of no important benefit.

2. ANL-24--2.22 cm long, 67.3% dense (-500 mesh), sintered nickel backing with an acoustic impedance of about $21 \times 10^6 \text{ kg/m}^2\text{-sec}$. No recognizable reflections were noted from the back face of backing member, but initial pulse length was as long as that of ANL-25, owing to the higher reflection coefficient between the piezoelectric element and the 67.3% dense nickel backing. The higher attenuation due to the lower density did not solve the problem.

3. ANL-23--1.27 cm long, tungsten-loaded epoxy (10 parts by weight tungsten to one part by weight epoxy) backing with an acoustic impedance of about $11 \times 10^6 \text{ kg/m}^2\text{-sec}$. Initial pulse length was shorter than that of ANL-24 or ANL-25. No reflections were obtained from the back face of backing member. Improved attenuation by this material was beneficial despite the acoustical mismatch.

4. ANL-22--1.27 cm long, Fiberglass-loaded epoxy backing with an acoustic impedance of about $6 \times 10^6 \text{ kg/m}^2\text{-sec}$. This probe had a shorter initial pulse length than the other three. No reflections were observed from the back face of the backing member.

These four ultrasonic transducer probes contained 1.27-cm-dia, 5-Mc, lead zirconate-titanate piezoelectric elements. Except for the backing material, the probes were identical.

Another sintered metal sample, 79.9% dense (-325 mesh) sintered nickel, was received from the manufacturer. This sample is 2.22 cm long and 1.27 cm in diameter. It has an acoustic impedance of about $35 \times 10^6 \text{ kg/m}^2\text{-sec}$. Since sintered nickel at this density has low absorption to ultrasound, a 30° cone was machined at one end of the cylinder.¹¹ This cone scatters the sound and more effectively attenuates it. The sample was utilized as a backing member for ANL-26, along with use of a 1.27-cm-dia, 5-Mc, lead zirconate-titanate element. Scattered reflections from the backing member were observed at high-gain settings. It appears that a backing member of this type would be effective at frequencies of 5 Mc and up.

A comparison of the longitudinal velocities in -500 and -325 mesh nickel samples revealed no significant difference. Therefore, to achieve the desired acoustic impedance of $34 \times 10^6 \text{ kg/m}^2\text{-sec}$ with either particle size, one would employ about the same density.

¹¹Howry, D. H., et al., U. S. Patent #2,972,068, February 14, 1961.

Electronic circuitry for the pulse Schlieren system has been completed. With this system, pulses can be observed from 10 to 400 μsec after being generated by an ultrasonic transducer probe.

c. Development of a Neutron-image Intensification System. A revised gamma control circuit for the television system was tested with results that indicated the circuit would be useful for enhancing contrast in the system. A more complete evaluation is planned.

An attempt to observe a Laue neutron diffraction pattern from a MgO single crystal was made with the neutron-image intensifier. A collimated beam, approximately 3 mm in diameter and having an intensity of $1.1 \times 10^6 \text{ n/cm}^2\text{-sec}$, from the Juggernaut reactor was used. The MgO crystal was positioned between the collimator output and the neutron-image intensifier. The output phosphor for the image intensifier was viewed both directly and with an optical system (about 3X magnification) loaned for the test by the Picker X-Ray Corporation.

No Laue spots were directly observed with the intensifier. The presence of the Laue spots was confirmed by a film image of the neutron beam and surrounding area. The estimated maximum intensity of the diffraction spots from the film results was about $10^2 \text{ n/cm}^2\text{-sec}$. Neutron intensities of this order can be observed directly with the neutron intensifier if a reasonable area is involved.

Although the results of this experiment were negative, some interesting observations can be made. First, the physical arrangement for observing the diffraction patterns was reasonably convenient. The optical system provided binocular viewing and was vertically oriented so that the viewer could stand at the side of the equipment and look down at the neutron image presented. While viewing, it would be relatively simple to rotate the mounted crystal and thereby orient the crystal in a short time.

Secondly, one can conclude that a higher collimated neutron beam intensity at the crystal would provide a useful result with the arrangement described above. At CP-5, one could easily anticipate an improvement of neutron intensity of 10 times; AARR would provide improvement by an additional order of magnitude. In the tests at Juggernaut reactor, the intense diffraction spots ($\approx 3 \text{ mm}$ in diameter) had an approximate neutron intensity of 1.4 neutrons/spot for one visual accumulation period (0.2 sec). Such a low intensity would, of course, mean a high degree of uncertainty about the detection of the spot. Experimental tests with this intensifier tube indicate that for visual detection of a 3-mm-dia neutron irradiated area, the neutron intensity would have to be approximately $5 \times 10^3 \text{ n/cm}^2\text{-sec}$ or higher.

D. Engineering Development

1. Boiling Liquid-metal Technology

a. Niobium-1% Zirconium Loop. This facility is designed to investigate the heat transfer and two-phase flow characteristics of boiling sodium to a maximum temperature of 2100°F and a pressure of approximately eight atmospheres. Among the variables to be investigated are boiling heat flux and temperature difference up to the critical flux occurrence, boiling and adiabatic two-phase pressure losses, vapor volume fraction, and boiling stability parameters.

In preparation for loop operation, the vacuum chamber was pumped down to 10^{-8} Torr. The mass spectrometer indicated a large amount of water vapor and hydrogen as residual gas. After baking at 400°F, the system behavior did not follow previous operating experience. Further inspection revealed that the seals and bearing on the roughing pump had failed after approximately 1000 hr of operation. Repairs are underway and the possibility of using more reliable components is being investigated.

The remaining Chromel/Alumel thermocouple assemblies are to be delivered late in June. The contract for W/W-Re thermocouples has been cancelled with a rebid in progress.

The sodium in this loop has been circulated in the hot-trap system for approximately 700 hr. The whole system will begin operation as soon as repairs to the vacuum system are complete.

b. Heater Experiments

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

The present test setup utilizes a 0.030-in.-dia thoriated-tungsten cathode and a Schedule 160, 3/4-in.-nominal diameter pipe (anode). The pipe is surrounded by liquid sodium.

Approximately 225 hr of testing with the thoriated-tungsten cathode have been achieved. Heat fluxes exceeding 300,000 Btu/hr-ft² have been supplied to the anode during both steady-state and transient testing, with the peak sodium temperature rising to 1400°F.

Investigation by an outside vendor to produce necessary information for the final design of a transient electron-bombardment heater is nearly complete.

2. General Heat Transfer

a. Heat Transfer in Double-pipe Heat Exchangers

(i) Counter-current Turbulent Liquid-metal Flow. The special case of equal heat capacity flow rates ($H = 1$) of the two heat-exchanging fluids received additional analytical study during this past month. Previous results for this case had been obtained by use of Laplace transform techniques and plug-flow representations of the fluids. There were serious theoretical questions, however, related to the validity of Laplace transform methods when applied to the counterflow problem in general; e.g., the poles of the Laplace transform solutions are in general unbounded over the entire real axis of the complex plane so that the usual sufficiency conditions for the validity of the inversion integral do not apply. Recently, results equivalent to those obtained by Laplace transform techniques were obtained from the basic analytical solution for $H \neq 1$ by allowing H to approach unity continuously from both $H > 1$ and $H < 1$.

b. Heat Transfer in Liquid-metal-cooled Reactor Channels

(i) Nonuniform Heat Flux. As a preliminary to investigation of the effect of axial heat conduction in heated channels with nonuniform heat-flux distributions, a study of several uniform-flux cases not previously considered by others has been initiated. These cases, although highly idealized, are chosen to introduce possible effects of the types of channel inlet and outlet configurations that actually exist in either reactor channels or laboratory experiments. The first case being studied, which applies more to laboratory experiments than to reactor channels, considers very long unheated and insulated inlet and outlet ducts attached to a heated channel, all three sections having the same cross-sectional area and shape. A second case is intended as an idealized representation of inlet and outlet headers by considering liquid-metal flow from a semi-infinite space into the heated channel inlet and with a similar configuration at the outlet.

Results for the first case are shown in Fig. 9. Three different Nusselt numbers for a parallel plane channel uniformly heated on one side are plotted versus position along the channel length measured either from the inlet or outlet. Two of the Nusselt numbers, Nu and Nu^* , differ in the choice of bulk temperature. The first, Nu , is based on the actual bulk temperature as predicted by the analysis. The actual bulk temperature, however, is affected by axial conduction and as a result would be difficult to determine in experiments or for design predictions of surface temperatures in reactor coolant channels. The Nusselt number Nu^* is based on a fictitious bulk temperature computed by the customary

simple heat balance (which neglects axial conduction) and is the one usually determined in laboratory experiments and used for design predictions. The Nusselt number Nu_0 refers to the case of negligible axial heat conduction.

Fig. 9. Nusselt Number vs. Distance along Channel. Effect of axial conduction in a parallel plane channel heated uniformly on one side with long insulated inlet and outlet duct.

The results shown are for a Peclet number of 10, which is about the lowest possible for the turbulent flow of liquid metals and for which the effects of axial conduction are expected to be largest. The results indicate that, except for the channel outlet, Nu^* is less than Nu_0 , which means that axial heat conduction increases surface temperatures. The increase, however, is not very large. For example, a separate computation indicates that for a total rate of heat input sufficient to raise the average coolant temperature by 300°F , surface temperatures predicted by use of heat transfer coefficients which do not account for axial conduction, will be about 6°F too low.

E. Chemical Separations

1. Fluidization and Volatility Separations Processes

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

(i) Fluorination of UO_2 - PuO_2 -fission Product Pellets. Development studies are being performed in a 2-in.-dia fluid-bed reactor to determine optimum conditions for fluorinating UO_2 - PuO_2 pellets containing fission products. Recent experiments in these studies have involved the reuse of the alumina bed for the processing of three batches of UO_2 - PuO_2 -fission product pellets. The initial charge to the reactor for the first run consisted of 750 g of pellets (to form a fluidized-packed bed 2.2 in. deep) and 1200 g of alumina. Because the fluidized bed was sampled after fluorination of the pellets, the weight of the bed decreased in successive runs; the recycled alumina bed for the second run was 1115 g, and for the third run, 970 g. The pellet-to-alumina weight ratio was maintained at 0.6 for each run. In all three runs cesium fluoride was added to the alumina bed in proportion to the pellet charge (0.9 g CsF /kg pellets).

The fluorination sequence for the recovery of uranium and plutonium as volatile hexafluorides was carried out as follows: oxidation of the pellets for 4 hr at 450°C with 20 v/o oxygen, fluorination for 2 hr at 350°C with 5 to 15 v/o fluorine in nitrogen, followed by recycle-fluorination using 90 v/o fluorine in nitrogen for 3 hr at 350°C , 6 hr while the bed temperature was increased from 350 to 550°C at $5^\circ\text{C}/9$ min, and 1 hr at 550°C . In the processing of the third batch of pellets, the recycle-fluorination period at 550°C was extended to 4 hr.

The final bed, after fluorination of the third batch of pellets, contained 0.009 w/o uranium and 0.009 w/o plutonium, which corresponds to the removal of more than 99.9% of the uranium and 99% of the plutonium in the three pellet charges. The fluoride content of the final alumina bed was 4.42 w/o. These results again indicate that reuse of the alumina bed material for the fluorination of several batches of fuel is a feasible method for achieving high overall uranium and plutonium removals.

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

(i) Engineering-scale Alpha Facility. The primary objectives of experiments performed in the engineering-scale alpha facility are to investigate the possible difficulties in handling PuF_6 on an engineering scale and to determine the feasibility of possible fluoride volatility flowsheets. Initial experiments involve the processing of UO_2 - PuO_2 fuel containing synthetic fission products; later experiments may be made with low levels of

fission product activity. Several runs have been performed in recent months with plutonium-containing oxide fuel material. These runs involved the fluorination of $\text{UO}_2\text{-PuO}_2$ pellets to form a mixture of UF_6 and PuF_6 , and the separation of plutonium from the $\text{UF}_6\text{-PuF}_6$ mixture by thermal decomposition of PuF_6 to solid PuF_4 in a 2-in.-dia fluid-bed reactor.

Operating conditions for the thermal decomposer during the latest run were: bed temperature, 300°C ; hexafluoride feed rate, 20 g/min; superficial gas velocity, 0.15 ft/sec; gas residence time, 10 sec; and bed charge, 2 kg of minus 100 mesh alumina coated with PuF_4 in previous runs. Chemical analyses of samples of the fluid-bed material and of the UF_6 off-gas stream from the thermal decomposer indicated that essentially complete separation of plutonium from uranium was achieved. The average plutonium concentration of the off-gas stream was 1.4×10^{-4} w/o based on the hexafluoride content of the off-gas. Based on analysis of the fluid-bed material, a plutonium separation efficiency of 99.9% was achieved. It appears that the use of a fluid bed is a suitable technique for separating plutonium from $\text{UF}_6\text{-PuF}_6$ mixtures by thermal decomposition.

(ii) Process Development Studies for Uranium Dioxide Fuels.

An engineering-scale fluid-bed facility is being modified to handle bromine pentafluoride and bromine trifluoride for process studies with UO_2 fuel. The facility basically consists of: (1) a reagent feed system, (2) a 3-in.-dia fluid-bed reactor constructed of nickel, (3) equipment for continuous analysis of off-gases from the reactor, and (4) a system for the disposal of off-gases, which includes a sodium fluoride trap for collection of UF_6 and a scrub tower for disposal of other gaseous components.

The processing sequence which will be studied with UO_2 fuels involves two steps: (1) oxidation of the UO_2 pellets to U_3O_8 fines with 10 to 50 v/o oxygen in nitrogen at 400 to 500°C , and (2) fluorination of the U_3O_8 fines to UF_6 at 200 to 400°C , using BrF_5 diluted with nitrogen. Alternatively, the U_3O_8 fines may be converted to a mixture of UF_4 and UO_2F_2 by reaction with hydrogen fluoride prior to fluorination with BrF_5 .

The primary objectives of this process development program are to obtain UF_6 production-rate data at various fluorination conditions by continuously measuring the concentration of UF_6 in the off-gas stream, and to identify and measure the concentrations of the remaining constituents in the off-gas stream. Initial tests will be made using BrF_5 as the fluorinating agent; later experiments may investigate the use of BrF_3 .

2. General Chemistry and Chemical Engineering

a. Preparation of Ceramic Fuels for Fast Reactors. The experimental program for the preparation of UC and (U, Pu)C by a fluidized-bed

technique (see Progress Report for February 1966, ANL-7176, p. 67) is continuing. The monocarbide is produced by carburization of hydrided uranium or uranium-plutonium alloy with hydrogen-hydrocarbon gas mixtures. A fluidized-bed reactor (designated "Demonstration Reactor" in ANL-7176, p. 70) that is small enough to be manipulated in a glovebox (e.g., for product removal) is now being operated to produce (U,Pu)C on a 300- to 600-g scale. Three preparations of (U, 15 w/o Pu)C on a 300-g scale have been completed thus far under the following conditions: gas concentrations, 10 v/o CH₄-90 v/o H₂; total pressure, 4 atm; bed temperature, 750°C.

The rate of reaction for the formation of the (U,Pu)C was determined continuously by monitoring the methane concentration of the incoming and outgoing fluidizing gas streams with an infrared analyzer. The extent of reaction during an experiment was obtained by integrating the rate of reaction over the time interval of interest. Under the conditions used, about 10 to 12 hr were required to produce stoichiometric monocarbide (4.80 w/o carbon). After an initial reaction period of ~6 hr, the rate of reaction became relatively low at a carbon content of about 4.1 w/o. An additional hydriding of the residual U-15 w/o Pu alloy and cooling to room temperature were used to increase the reaction rate. The additional hydriding was carried out in all runs except the first one.

The product from the first run, which was terminated after 6 hr, was hypostoichiometric in carbon content, whereas the other two products were close to the stoichiometric composition. The carbon contents of the products of the three runs were 4.11, 4.96, and 4.89 w/o, respectively. The oxygen impurity levels were very low for these runs, averaging about 500 ppm or 0.05 w/o. One sample of the product from the second run contained only 160 ppm oxygen. It is now believed that, in general, the carbide produced by this process has a low oxygen content (<500 ppm) when it is removed from the reactor but that significant amounts of oxygen may be introduced during sampling and subsequent analysis.

The effect of temperature on the reaction rate was explored. The results indicated that the rate was significantly lower at 550 to 650°C than at 750°C. This is in agreement with previous findings.¹²

F. Plutonium Recycle Experiment (EBWR)

1. Temperature Coefficients of Reactivity

The average uniform temperature coefficient of reactivity between 79 and 153°F for the full loading when poisoned with approximately 4.4 g/gal H₃BO₃ was determined as $3.9 \pm 0.7 \times 10^{-5} \Delta k/^{\circ}\text{F}$ from the multiplication constant as a function of temperature with five control rods withdrawn. The

¹²Chemical Engineering Division Semiannual Report, January-June 1965, ANL-7055, pp. 90-94.

multiplication constant was obtained from the integral worth of that portion of a corner control rod (No. 4 rod) that must be inserted to maintain criticality with the three other corner rods and the center rod fully withdrawn and with the other four rods fully inserted. The results of the experiment are given in Table XVIII.

TABLE XVIII. Multiplication Constant of EBWR with Five Control Rods Withdrawn

Temperature (°F)	Critical Height of No. 4 Rod (in.)	Worth of Withdrawing No. 4 Rod (%k)	k when No. 4 Rod Withdrawn
78.7 ± 0.6	23.52 ± 0.03	0.229 ± 0.018	1.00229 ± 0.00018
121.5 ± 0.8	16.14 ± 0.03	0.406 ± 0.022	1.00406 ± 0.00022
153.5 ± 0.7	7.50 ± 0.03	0.531 ± 0.026	1.00531 ± 0.00026

This method for determining the temperature coefficient assumes that the rod worths do not change with temperature. The validity of this assumption was confirmed when the reactivity effect of withdrawing No. 4 rod from 23.5 in. at two temperatures was compared. The reactivity effects at 153 and 79°F were $0.236 \pm 0.018\% \Delta k$ and $0.229 \pm 0.018\% \Delta k$, respectively. The difference between these values lies well within the experimental error.

2. Transfer Functions

The zero-power transfer function of EBWR has been measured with a full core loading consisting of 36 plutonium assemblies, 60 enriched uranium assemblies, and 49 natural uranium assemblies. The transfer function was measured by sinusoidally oscillating the center control rod $\pm 1/8$ in. from 0.1 to 1.0 radian/sec and $\pm 1/4$ in. from 1.0 to 55 radian/sec. The reactor conditions corresponded to an eight-rod bank position of 18.6-19.2 in., a mean center rod position of 18.5 in., and 6.78 g/gal of boric acid in the reactor water. Data were taken simultaneously with analog and digital data-handling equipment.

III. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. General Plant Design

a. Architect-Engineer. The Architect-Engineer, Burns and Roe of New York, has submitted the drawings and text of the Title I report for review. The Title I cost estimate and the development of the Title II Pert fragnets and construction schedules are in preparation for incorporation in the Title I report to be issued on September 1, 1966.

2. Reactor Physics Experiments

The peripheral control blade No. 6 has been calibrated at room temperature (24.2°C) and at 53°C. To within the accuracy of the measurement, the reactivity-worth curve is independent of temperature. The integrated reactivity-control worth is 1.2%.

To obtain an estimate of the change in control effectiveness of the peripheral control blades during core operation, some fuel adjacent to peripheral control blade No. 6 was removed and the blade was recalibrated. This perturbation simulates a situation where most of the U^{235} in the graded fuel zone at the outer radius of the reactor core has been consumed. Details of the perturbation follow:

Fuel foils were removed from the outermost three fuel foils in each of 7 assemblies of the 1215/1620 loading. Four of these 7 assemblies were aligned with their plates parallel to blade No. 6. The other three assemblies flanked these four immediately adjacent fuel assemblies.

Assuming that the control worths of the control blades in unperturbed regions of the reactor are unchanged by this removal of fuel foils, it is estimated that the replacement of the total of 21 fuel foils by stainless steel resulted in a net reactivity loss of ~0.9%. The estimated loss in control worth of control blade No. 6 is 0.37%, or approximately 30 percentage points; this is based upon a recalibration of peripheral blade No. 6 over its full range of effectiveness.

The Internal Thermal Column (ITC) has been enlarged in lateral area by removing fuel plates, steel strips, and boron-stainless steel (B-SS) strips from the inner 7 fuel-plate positions of the 6 fuel assemblies bordering on the ITC. A total of 7 fuel foils and 9 B-SS strips were removed from each of the assemblies, leaving a core loading of 1173 fuel foils and 1566 poison plates. This loading was subcritical, which means that the perturbation caused a reactivity loss of more than 2.2%. To restore an effective operating

margin of reactivity, an additional 219 B-SS poison strips were removed. The reactor was made critical with control blades Nos. 2 and 5 fully inserted, and No. 6 at 36.15 cm (i.e., slightly inserted).

The new ITC is somewhat oversized in comparison with the reference design ITC. Measurements of the reactivity worths of massive ITC voids will be made, to provide more data in the vicinity of the reference ITC size. Such measurements were made with the original ITC of the Criticality Facility (see Monthly Report for December 1965, ANL-7132).

3. Theoretical Reactor Physics

Additional calculations of reactivity worths of soluble neutron poisons in the reference reactor have been completed for boron, for gadolinium, and for europium (see Progress Report for May 1966, ANL-7219, pp. 57-58). The results are presented in Figs. 10 and 11. The effects of poisoning of the top and bottom reflectors have not been included. However, this is a second-order correction.

Fig. 10. Reactivity-control Worths of Boron in Various Regions of AARR

Fig. 11. Reactivity-control Worths of Boron, Gadolinium, and Europium as Soluble Neutron Poisons in AARR

The conclusions of this study of soluble poison are:

1. To control 1-2% in reactivity, it appears to be feasible to use boron, either in the form of boric acid or perhaps a borate (e.g., sodium pentaborate). (This feasibility is argued solely in terms of being able to add a concentrated solution sufficient to poison the entire primary coolant system.) The use of a rare-earth poison is neither indicated nor justified for this purpose.
2. To supply enough soluble poison to make up for the entire reactivity worth of the system of 12 control blades, roughly 10 g of natural boron per liter of water would be needed. This amount is considerable, considering that there are of the order of 10^5 liters of water in the entire primary system. It would be difficult to obtain such a uniform concentration in this system. Moreover, if the primary-coolant water were not isolated from the water of the reactor pool, the total mass of poison required would be multiplied. To accomplish this much reactivity control, even a larger mass of rare-earth elements would be needed, at an utterly unreasonable total cost.

In the Progress Report for October 1965, ANL-7115, pp. 69-70, some problems associated with use of a neutron window were discussed. The principal difficulty is to meet the mutually incompatible requirements of a high thermal-neutron flux beyond the pressure vessel and a low high-energy neutron flux in the pressure vessel, so as to reduce radiation damage of the vessel at the window location. These concepts have been studied with the aid of the program MAC on the CDC-3600 computer.

In one concept, most of the water in the region just beyond the pressure vessel wall, at the window location, where the neutron detectors are to be located, is removed, replacing it either with aluminum or with beryllium. This water replacement greatly increases the average thermal-neutron flux in the detector region, for water attenuates thermals very efficiently. The average level of the thermal-neutron flux may be increased by a factor of ten in this way.

In a second concept, some of the aluminum in the window is replaced by a material which is more effective in attenuating the high-energy neutrons in the range from ~ 0.1 to 10 MeV, the principal range of radiation-damage neutrons. Although water might be used, water reduces the level of the thermal-neutron flux in the detectors twice as much as the corresponding reduction in the flux of radiation-damage neutrons in the pressure vessel. Another possibility is steel. The replacement of an 8-in.-thickness of aluminum containing 4 v/o water by steel with 4 v/o water would reduce the radiation-damage rate of the pressure vessel to approximately $1/4$ of its level. The thermal flux in the neutron detectors would be lowered to $\sim 1/3$ of normal level by this substitution. Thus, while it is an easier

engineering solution to introduce water gaps near the inside surface of the vessel, introduction of some steel into the neutron window seems to be a very much more effective way to resolve the problem. A preliminary survey of the engineering problems has not revealed any substantial obstacles to the use of steel in this manner.

Calculations have been completed for the variation of the effective delayed-neutron fraction, β_{eff} , as a function of the loading of the reactor core in the AARR Criticality Facility. These calculations were made with the zero-order perturbation-theory program 1188/RP. The value of β_{eff} is largest in the lightest loading (315/0), and it decreases monotonically as the loading is increased. In the 315/0 loading, $\beta_{\text{eff}} \approx 1.1 \beta$. In the current loading (1215/1620), the computed β_{eff} is only 2% less than in the 315/0 system (see Table XIX).

TABLE XIX. Computed Effective Delayed-neutron Fraction, β_{eff} , in the AARR Criticals

Core Loading	Mass of U ²³⁵ (kg)	Mass of Boron (g)	Composition of Beryllium Reflector	β_{eff}
315/0	16.1	0	10% Plexiglas 90% Be	0.00727
615/420	31.5	220	10% Plexiglas 90% Be	0.00723
615/420	31.5	220	100% Be	0.00721
810/810	41.4	425	100% Be	0.00718
1215/1620 ^(a)	62.1	850	100% Be ^(a)	0.00712

(a) These calculations were for a 1215/2000 loading with a 100% beryllium reflector. The actual core loading was 1215/1620, and the reflector contained the aluminum of the walls of 3 beam tubes.

4. Fuel Materials

The 6 to 18 w/o UO₂-SS cermet dispersion fabrication studies and the UO₂ characterization and evaluation studies at the Battelle Memorial Institute (BMI) have been completed.

The work on the low-inventory fuel-dispersion studies indicated that the fuel plates requiring an equivalent fuel loading in the range from 5 to 15 w/o UO₂-SS should be made by a special blending procedure which departs only slightly from that used in the preparation of the higher-inventory plates but which must be used if the homogeneity of dispersion is to be

maintained at the desired high level. Studies of various fuel concentrations (i.e., 6 w/o UO_2 in a 0.030-in.-thick cermet, 12 w/o UO_2 in a 0.015-in.-thick cermet, 37 w/o UO_2 in a 0.006-in.-thick cermet) demonstrated that the best quality of fuel particle spacing and overall uniformity of fuel dispersion was obtained with a 0.030-in.-thick cermet.

The BMI studies on UO_2 characterization and evaluation were conducted with twelve different commercial and laboratory produced oxides. These studies included determination of the physical properties of the UO_2 particles and evaluation of their respective behavior during fabrication into a finished fuel plate. Two important characteristics were observed for the UO_2 particle that possessed densities of interest to AARR (i.e., 75 to 85% nominal theoretical density):

(a) The UO_2 that had 2 to 3 w/o addition of CaO or ZrO_2 displayed a crushing strength approximately 10 to 15% higher than comparable UO_2 without the additions. The oxide with the additions maintained a higher strength-brittle characteristic that resulted in 20 to 30% of the particles being crushed, but there appeared to be very little densification of the UO_2 . The amount of stringing in most of the crushed particles was no greater than that observed in the earlier irradiation specimens. Examination of samples removed from the test run after 4:1 reduction (i.e., before going on to the final 6:1 reduction) showed very little evidence of change in shape of the UO_2 .

(b) The UO_2 that had no additive displayed was ductile at the fabrication rolling temperatures (1000 to 1100°C); also, rolling reductions of 6:1 produced high densification and deformation (footballing) of the UO_2 .

In the AARR Phase II irradiations test program conducted by Phillips at NRTS, Idaho, the three-plate microassembly were subjected to burnup close to twice the AARR average burnup and to heat fluxes two to three times higher than the AARR 100-MW average. Similar in-pile tests have been completed for the 37 w/o UO_2 -SS with 1.3 w/o Nb-coated ZrB_2 plate specimens.

5. Postirradiation Examination of UO_2 -Stainless Steel Cermet Fuel Specimens

The postirradiation examination of UO_2 -stainless steel cermet Advanced Army Reactor fuel specimens irradiated in the Oak Ridge Research Reactor (ORR) pressurized-water loop under contract with ANL, has been virtually completed; only the final analysis and some supplementary work remain. The characteristics of these specimens are shown in Table XX. The fuel temperatures and the amount of burnup varied considerably among the specimens. All specimens had been produced by the Martin Company, Nuclear Division at the Baltimore plant, and irradiated at ORNL from

November 1963 to May 1964 as part of the Army Reactors (AEC) fuel development program for their plants. No specimen received the planned burnup because of curtailment of the Army development program.

TABLE XX. Nominal Physical Characteristics of the UO_2 -Stainless Steel Cermet Fuel Specimens for the Army Fuel Experiment in the ORR Pressurized-water Loop

Cermet Characteristics									
Specimen		Powder Type ^b	UO ₂	Particle Size (μ)	SS Composition ^a (%)			Cladding Composition (AISI Type)	Fabrication Process
Form	Type		w/o		Cr	Ni	Fe		
Plate ^c	1	MSC	30	105-149	17.5	12.5	70.0	347	d
Plate ^c	2	MSC	35	105-149	17.5	12.5	70.0	347	d
Plate ^c	3	MSC	40	105-149	17.5	12.5	70.0	347	d
Plate ^c	4	MSC	45	105-149	17.5	12.5	70.0	347	d
Plate ^c	5	MSC	50	105-149	17.5	12.5	70.0	347	d
Plate ^c	6	ASC	40	105-149	17.5	12.5	70.0	347	d
Plate ^c	7	AAC	40	105-149	17.5	12.5	70.0	347	d
Plate ^c	8	MSC	40	105-149	20.0	10.0	70.0	347	d
Plate ^c	10	MSC	40	105-149	17.5	12.5	70.0	318	d
Plate ^c	11	MSU	40	105-149	Type 318 prealloyed			347	d
Plate ^c	12	ASC	40	149-177	17.5	12.5	70.0	347	d
Plate ^c	13	MSCP	40	105-149	17.5	12.5	70.0	347	d
Tube ^e	1	AAU	28	44-105	Type 304L prealloyed			347	f
Tube ^e	2	MSC	35	105-149	17.5	12.5	70.0	347	d
Tube ^e	3	MSC	40	105-149	17.5	12.5	70.0	347	d
Tube ^e	4	MSC	45	105-149	17.5	12.5	70.0	347	d

^aElemental powder unless noted otherwise.

^bMSC for Martin-produced spherical coated powder; ASC for alternate-process spherical coated powder; AAC for alternate-process angular coated powder; MSU for Martin-produced spherical uncoated powder; MSCP for Martin-produced spherical coated powder having induced porosity; AAU for alternate-process angular uncoated powder.

^cNominal fueled dimensions are 0.69 x 1.00 x 0.090 in.; nominal overall dimensions are 1.00 x 1.25 x 0.106 in.

^dCold press, sinter, and isostatic press.

^eNominal dimensions in inches were

Type	Overall			Clad Thickness	Fuel	
	OD	ID	Length		Thickness	Length
1	0.506	0.416	10.77	0.0075	0.030	9.0
2-4	0.540	0.330	10.77	0.0075	0.090	9.0

^fMartin process for PM Core-1 fuel.

All specimens except the Type 2 No. 9 tube were stored in the ORR pool from September 1964 to May 1966. The results of this earlier examination had not been formally reported.

The work which was performed is summarized in Table XXI. No examinations of plates No. 8 and No. 10 were made because they had little bearing on the AARR fuel application.

TABLE XXI. Postirradiation Examination Program for Advanced Fuel Specimens

I. Nondestructive Examinations	II. Destructive Examinations
A. Visual examination	A. Burnup analyses (isotopic)
B. Dimensional examination	B. Blister tests
C. Density determination	C. Metallography
D. Gamma scanning	D. Hardness measurements
	E. Microprobe samples to be sent to the National Reactor Testing Station in Idaho

The results of the nondestructive examinations were reported by ORNL as follows:

1. Visual Examination. The specimens all appear to be in good condition. No obvious defects could be seen.
2. Dimensional Examination. Dimensional measurements on the flat-plate specimens showed no evidence of significant changes in specimen length or width. However, preliminary results indicate that the thickness of the specimens has increased slightly (0.3-0.9%) as a result of irradiation. The magnitude of this increase appears to be related to the estimated specimen burnup.

Measurements on the tubular specimens showed no evidence of diameter changes greater than 0.002 in. However, even within this small range, the diameter change appears to be burnup dependent. Fuel tubes Type 2 No. 7, Type 3 No. 7, and Type 3 No. 9 were found to have bowed ~0.025, 0.030, and 0.060 in., respectively, as a result of irradiation. The tubes also appear to have increased in length by 0.003 to 0.018 in.

3. Density Determination. Density measurements on the flat-plate fuel specimens indicate that the coupons increased in volume from 0.1 to 0.5%. Such volume changes appear to be reasonably consistent with the specimen thickness changes. Density measurements on the Type 2, 3, and 4 tubular fuel specimens indicate volume changes of 0.1 to 0.7%. The density measurements on the Type 1 tubular fuel specimens indicate volume increases of 1.1 and 1.5%.

4. Gamma Scanning. These measurements were initiated by performing γ -energy spectra analyses using a 256-channel γ spectrometer. This was done to verify that we could select an energy "window" which would give us burnup profiles.

The flat-plate specimens were scanned for γ activity by moving them in a stacked (end-to-end) column past a 0.020 x 1.0 x 18-in. detector slot and recording the gamma activity having energies between 0.55 and 0.95 MeV on a strip-chart recorder. This activity is predominantly from Ba^{137} (Cs^{137} daughter) and from the Zr-Nb decay chain; thus the γ activity observed should be a good indicator of relative fission density. This was followed by a similar scan after restacking the specimens turned 90° (side-to-side). The resulting scans indicate considerable peaking of the activity near the cermet-dead edge interface. Burnup of a particular portion of flat-plate specimens will be estimated from isotopic analyses from a few specimens, the relative magnitude of these activity scans, and the loading of the specimens.

Tubular specimens were scanned axially for γ activity by the same general techniques as for the flat-plate specimens. Circumferential γ scanning of similar tubes nondestructively has not been very satisfactory and, thus, we have made no attempt to do this. We plan to perform circumferential γ scans on sections cut from these tubes (see destructive examination section).

The results of destructive examinations were as follows:

a. Sectioning and Metallography. The specimens all appeared to show typical irradiation effects. Since there were a large number of samples that could be prepared and evaluated on the basis of burnup and temperature (some of the samples had sizable fission rate gradients across the specimens), a general pattern of irradiation effects in both the matrix and the UO_2 was established.

b. Burnup Analyses. Six of the flat-plate specimens and samples from each of the four tubes were selected for isotopic analyses. These include samples from high, low, and intermediate burnup specimens and high, low, and intermediate loadings. The burnup samples chosen from the flat-plate specimens are as follows:

Type	No.	Nominal Loading (w/o)
1	1	30
4	3	45
5	2	50
6	1	40
12	2	40
13	1	40

Only a few of the samples possessed areas where the burnup exceeded 10 w/o U^{235} .

c. Blister-Anneal Tests. Flat-Plate Specimens—Blister-anneal tests were conducted on sections of fuel samples removed from all except flat-plate coupons No. 8 and No. 10. These tests were performed in a manner similar to that used by Phillips in the blister-anneal tests performed on ANL UO_2 -SS dispersion fuel plates.

The blister-anneal tests were run in 100°F increments from 700 to 1600°F. All but one of the samples failed above 1200°F. Most failed at 1400°F, and one did not fail at 1600°F.

d. Hardness Measurements. Hardness measurements were performed on the clad material of a limited number of specimens (3 flat-plate specimens).

e. Electron-microprobe Examination. This will be conducted pending the analysis of the work completed in the hot-cell examinations. Such analyses would be performed by NRTS personnel at the Idaho facility on selected samples.

6. AARR Vessel

The preliminary stress analysis has been completed for the Inconel alloy 600. If Inconel is selected as the vessel material, the solid Inconel sections at the main shell will be 4 in. or less in thickness at the welds. A welding development program will be needed for Inconel material thickness exceeding 4 in.; however this need can be eliminated if clad carbon steel 212B is used instead of Inconel for the top head closure (8 in. in thickness). Mechanical stresses as well as thermal stresses at both the inner and outer surfaces of the vessel wall are shown in Fig. 12. The thermal stress is relatively high. The outside vessel wall should be partially insulated to reduce the differential temperature across the wall.

The top and bottom head closures were analyzed as if they were perforated plates. A number of different penetration configurations were analyzed in an effort to arrive at an efficient arrangement to meet both the user's requirement and the stress limitations. The stresses shown in Fig. 12 are equivalent stresses. The average stresses in the minimum ligament section and peak stresses have also been computed.

Local stresses at the inlet and outlet nozzles due to external loads have been investigated. The preliminary calculation shows that the maximum stress intensity is about 3600 psi, which is acceptable to the vessel wall.

B. Liquid Metal Fast Breeder Reactor (LMFBR)

1. Test Facility for Control-rod Drive

A facility designed to simulate the 1150°F operating environment of a sodium-cooled fast reactor has been assembled for testing a control-rod drive assembly. The system consists of an insulated and electrically heated, sodium-filled vessel containing a simulated control rod supported by guide bushings. The upper adapter of the control rod may be engaged or released by a gripper mechanism containing an integral sensing device. The gripper, fabricated from Haynes No. 25 alloy and Haynes Stellite No. 6B, is attached to a vertical drive shaft which is housed in a support tube fitted with guide bushings.

The drive shaft penetration at the top of the sodium vessel is sealed by a welded-disc nesting bellows enclosure which permits vertical movement of the shaft and eliminates the usual dynamic seal configuration.

The gripper jaws are manually operated by an actuating mechanism located on the drive shaft above the bellows. A ball-jointed armature, just above the jaw operator, passes through a toroidal electromagnet which is mounted on a pair of ball-nut lead screws. The electromagnet may be raised or lowered over a distance of 23 in. by rotation of the lead screws, which in turn, are driven by a polyphase motor and a torque-limiting clutch arrangement.

The shaft-mounted armature is held in position by the electromagnet during normal raising and lowering operations but may be released by de-energizing the magnet when scram is initiated.

Above the armature, the shaft terminates in the pneumatic piston and cylinder assembly of the scram accelerator and shock-absorber mechanism.

Scram data are transmitted to an oscillograph by a set of electrical contacts spaced 1 in. apart on the shaft armature which mate with a matching set of contacts on the electromagnet housing.

The facility will be used to plot time versus displacement curves at scram accelerator pressures ranging from 10 to 20 psi gauge and also to determine elapsed time between scram initiation and start of shaft movement.

C. Magnetohydrodynamics

1. Condensing Injector Studies

Preliminary attempts to establish high-velocity flow in the condensing injector have not been successful. The reason for the difficulty lies in the fact that it has not been possible to maintain the discharge pressure of the vapor nozzle at a low level for a length of time sufficient to establish supersonic flow. However, a large-capacity reservoir has been added to the test loop so that on injector startup, the downstream pressure can be maintained slightly above atmospheric until proper flow conditions are established. The necessary modifications have been made, and trial runs will be made shortly.

IV. NUCLEAR SAFETY

A. Reactor Kinetics

1. Studies of Penetration by Liquid Metals

The studies of the penetration of Type 304 stainless steel tubes by molten U-15 w/o Pu-10 w/o Zr alloy are useful in safety analyses. Initial measure-

Fig. 13. Penetration Rate vs. Temperature of Type 304 Stainless Steel in Molten U-15 w/o Pu-10 w/o Zr Alloy for Specimens of Different Wall Thickness

ments of the penetration rates indicated an increase with temperature. Further measurements showed that the results were widely scattered at all temperatures and wall thicknesses, for which there is no known reason. A plot is shown in Fig. 13 of the penetration rate versus temperature of Type 304 stainless steel in molten U-15 w/o Pu-10 w/o Zr alloy for specimens of different wall thicknesses. There is a general increase of the average penetration rate from about 0.04 mm/sec at 1220°C to about 0.12 mm/sec at 1350°C.

The mechanism is apparently a dissolution process wherein the liquid penetrates the solid and particles of stainless steel move from the interface by solution and convection. Many of the products are retained near the interface as shown by microprobe analyses of a specimen failed at 1350°C.

As shown in Fig. 14, the dissolution proceeds through a solid zone about 0.5 mm thick and a liquid zone about 1.5 mm thick. The boundary between these two zones is delineated by shrinkage pores and contrast in the electron current image. In the solid zone, the stainless steel is penetrated by the U-Pu-Zr along subgrain boundaries effectively isolating small particles of the stainless steel. The former gamma grain boundaries are shown as dashed lines. In the liquid zone, these particles gradually decrease in size until a liquid melt of uniform composition is reached. The stainless steel particles are shown as dark areas (2, 4, and 6) in Fig. 14, which correspond to their appearance on the electron current image. The microprobe analyses listed in Table XXII are uncorrected for absorption or intensification and can be used only as a relative measure of composition. (For example, the chromium value in area 1 should be 18.5 not 23.9.) All

the areas examined were 10 to 20 μ in width so that good analyses could be expected. The analyses of the stainless particles (2, 4, and 6) may be compared with the unaffected stainless steel analysis (area 1). The iron content is essentially constant in 1, 2, and 4, and decreases somewhat in 6, where an appreciable amount of uranium has finally diffused into the stain-

Fig. 14. Schematic of the Type 304 Stainless Steel Specimen Penetrated by U-15 w/o Pu-10 w/o Zr. The numbers correspond to the numbered areas in Table XXII.

less particle. The nickel diffuses rapidly out of these particles into the light areas of uranium and plutonium that surround them. The chromium content of the particles seems to build up (area 4) and then decrease. The light boundary areas (3, 5, and 7) change in composition as stainless steel is dissolved and as the proportion of liquid fuel increases in the boundary. The liquid fuel areas (8, 9, and 10) are probably indicative of the phases present in rapidly solidified uniform composition material.

2. Irradiated U-5 w/o Fs

An injection-cast U-5 w/o Fs alloy of reactor-grade material annealed at 500°C for 28 days has been examined by electron microscopy. This examination has shown that the intermediate phases in this alloy are considerably smaller in size than the phases formed in high-purity U-5 w/o Fs after the same heat treatment. The distribution of

TABLE XXII. Compositions of Phases Determined by Microprobe Intensity Ratios in the Couple of Type 304 Stainless Steel Against Molten U-15 w/o Pu-10 w/o Zr Alloy

Area: (a)	Compositions of Phases (w/o)									
	1	2	3	4	5	6	7	8	9	10
Electron Current Image:	Grey Continuous	Dark Subgrains	Light Boundary	Dark Subgrains	Light Boundary	Dark Subgrains	Light Areas	Light Areas	Grey Areas	Dark Areas
Element										
Ni	8.6	4.7	12.4	2.7	1.1	4.0	7.3	2.5	4.1	3.0
U	0	0	23.3 30.3(b)	0.3	31.7	10.3	22.9	60.4	42.9	25.4
Fe	62.3	62.3	38.9	65.1	37.9	55.6	26.5	10.0	27.2	31.0
Pu	0	0.1	5.0 6.5(b)	0.1	5.2	1.4	12.2	15.0	5.1	3.6
Cr	23.9	28.8	6.4	32.6	3.6	28.3	7.1	0.2	8.9	10.4
Zr	0	0	2.9	0	5.6	1.3	7.8	0.1	3.7	13.0
Total Percentage	94.8	95.8	88.9(c)	100.8	85.1	100.9	83.8	88.2	91.9	86.4

(a) Area 1: unaffected SS matrix; Areas 2 and 3: solid-state penetrated region; Areas 4, 5, 6, 7, 8, 9, and 10: molten region.

(b) Corrected for absorption and atomic number.

(c) Corrected total is 101.3%.

the intermediate phases in the annealed injection-cast sample is less uniform than in the high-purity U-5 w/o Fs alloy. An examination of injection-cast, irradiated (at 500°C for four weeks) U-5 w/o Fs showed no evidence of the intermediate phases, indicating that the intermediate phases may be put into a metastable solid solution as a result of irradiation.

3. Fast Reactor Safety

a. Coolant (Water) Expulsion Studies. All of the expulsion data, with the exception of the film information, have been reduced. Analysis and correlation have been started. Work is continuing on the computer program which will smooth and differentiate the data of height versus time from the high-speed films.

No additional experimental work in this area is planned. The apparatus has been disassembled.

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

Assembly of the superheat experiment is continuing. Welding of the sodium piping and valves is complete. The initial resistance heater has been assembled around the superheat vessel. The cooling coils and the auxiliary heating wire have also been assembled.

c. Sodium Expulsion Experiment. Fabrication of some of the components to be used are underway. Investigations pertaining to the instrumentation for the experiment are continuing.

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

The construction of experiments to study critical flow of different fluids is continuing. The fabrication of the argon gas cleaning system, the boiler, blowdown vessel, and test sections for the sodium loop should be completed shortly. Final assembly of the loop components will then be initiated.

The subatmospheric water loop has been put into operation. Preliminary tests indicate considerable superheat in the water during blow-down. Attempts are now being made to measure the amount of superheat necessary to create two-phase flow during the blowdown process.

4. Irradiated Fast Reactor Oxide Fuel Pins

Four UO_2 pins irradiated in MTR (see Progress Report for May 1966, ANL-7219, p. 77) were subjected to nuclear heating experiments in TREAT, and high-speed color photography was used to record sample behavior. Each sample pin was exposed in a standard TREAT transparent meltdown capsule having an inert gas atmosphere. Two Chromel-Alumel fast-response thermocouples were spotwelded to the cladding surface of each pin. A camera speed of 2000 frames per second was used. Each power burst was of about a half-second duration. Sample energy input in a fuel transient ranged from about 165 cal/g to about 300 cal/g. Sample fuel heating rates were as high as $\sim 2000^\circ\text{C}/\text{sec}$.

Each pin was given a low-energy preliminary burst of about 103 cal/g, to produce a cladding temperature rise of $\sim 800^\circ\text{C}$ to provide a check on the calculated dependence of sample temperature with reactor energy release (this relationship is dependent on sample burnup) and on the magnitude and consistency of heat transfer rates from fuel to cladding. Because of the absence of a liquid metal environment it was possible to observe the fuel pins and the pin fragments directly during and after failure, obtain fast-response cladding temperatures, and avoid the complexities of indirect analyses and reconstruction of events caused by large radial temperature gradients.

The first sample, a 0.7 a/o burnup pin, survived a transient that produced a maximum cladding temperature of 1275°C without failure. The other 0.7 a/o burnup sample and one 3 a/o burnup sample were exposed to transients calculated to produce temperatures in excess of those attained in steady-state irradiation, but not melt the fuel; the cladding on both pins failed, however. The remaining 3 a/o burnup pin was exposed to the highest energy transient, one programmed to attain fuel melting. In none of the four experiments which were characterized by temperature rise of short duration was there any photographic evidence of destructive fragmentation of the fuel due to internally trapped fission gases. Even in the case of the excursion of highest energy input, the high-speed photographs showed cylindrical fuel pieces, uncovered as cladding broke away or melted and ran down. In subsequent movement of the fuel, the fuel sections tended to maintain shape without disintegrating, splitting, frothing, or undergoing significant changes from cylindrical geometry. Although the fission gases inside the cladding cans constitute, in principle, a source of stored energy capable of producing rapid motion of fuel upon failure, they were not effective as such under the conditions of these experiments.

5. Modifications of ARGUS Code

The ARGUS computer code (1089/B134) was written¹³ to calculate the transient temperature distribution in multiregion, concentric cylindrical configurations with space- and time-dependent heat generation, temperature-dependent thermophysical properties (phase changes can be included), and flowing coolants with time-dependent velocities and appropriate film coefficients of heat transfer.

Since the principal application of this code is for the analysis of TREAT transient experiments it is being modified to include graphical output. Some minor changes also will be made to improve running time.

A subroutine will permit calculation, as an option, of the internal pressure, pressure stresses, and thermal stresses for a single clad pin. In this subroutine volume changes due to thermal expansion of the materials from the temperatures at the nodes as a function of time may be calculated. Temperature dependence of thermal expansion coefficients and phase changes will be included, but the geometrical position of the node points will be considered fixed (although in principle, changes due to radial and axial expansion could be incorporated by suitable modification of the basic code structure).

The net expansion of the fuel pin and bond material with respect to the cladding will be used to calculate the decrease in volume of the gas space initially present. From the initial gas pressure and volume, which will be input data, the pressure and the hoop stress due to pressure will be calculated by means of the approximations of the ideal gas law, assuming the arrangement to be that of a thin-walled cylinder.

From the temperatures in the cladding (the subroutine will emphasize this region), the thermal hoop stress from the temperature-dependent elastic and thermal properties will be calculated. The pressure and thermal stresses will be combined and compared to the temperature-dependent yield stress as a function of axial position and time. When the combined stress is greater than the yield stress, a simple linear interpolation will be used to give the time and position of failure. The thermal stresses, combined stresses, yield stress, pressure, pressure stress, and position and time of failure can be obtained as output by suitable input specification. Reduction of cladding strength due to melting or eutectic formation is neglected at this time but may be introduced in future revisions.

¹³D. F. Schoeberle, L. B. Miller, and J. Heestand, A Method of Calculating Transient Temperatures in a Multiregion Axisymmetric Cylindrical Configuration. The ARGUS Program 1089/RE248, Written in FORTRAN II, ANL-6654 (1963).

Because of the increased emphasis on ceramic fuels in the TREAT program, consideration is being given to modification of the basic logic of ARGUS for evaluation of the coefficients of the finite-difference equations used for the temperature calculations. Due to the large temperature gradients that can be generated in low-conductivity materials, significant errors can occur in the overall heat balance with the present method of computing the coefficients.

6. Convective Heat or Mass Transfer with Phase Changes

Many problems in nuclear safety and reactor technology, such as fuel melting and redistribution, or trapping of sodium oxide from sodium, have in common the mathematical problem of convective heat or mass transfer with a moving boundary. The transfer process and moving boundary are coupled by a boundary condition, whereas the convection and boundary movement are coupled through the basic partial differential equations.

An idealized version of the general problem has been formulated to develop the appropriate methods of solution and generate relations to use for design or analysis of this class of problems.

The equations of conservation of mass, momentum, and energy have been combined by the usual assumption of a stream function and, with appropriate boundary conditions, are put in nondimensional coordinates. Preliminary studies are concentrating only on convection and mass transfer for flow in a two-dimensional channel. Use of the stream function on the continuity and two Navier-Stokes equations leads to a fourth-order nonlinear partial differential equation, whereas the diffusion equation leads to a second-order nonlinear equation which includes the stream function. This set of partial differential equations can be reduced to a pair of coupled nonlinear ordinary differential equations by a new version of the widely used similarity transformation.

Unique features of this similarity transformation are its definition for a finite interval and the fact that one of the boundary conditions is specified for changing values of the similarity variable. The fourth-order "velocity" equation has four boundary conditions: two at the channel axis and two at the channel wall, one of which depends on the "diffusion" at the wall. The second-order "diffusion" equation contains the "velocity" and has a boundary condition at the axis and at the wall. The system has the necessary conditions for a solution, but it is not yet known whether these are sufficient for the existence of a solution.

7. Mark-II Integral Sodium Loop

A simplified layout of the loop is shown in Fig. 15.

Fig. 15. The Mark-II Sodium Integral Loop

The need for a larger test section and higher pressure rating made it impossible to retain the volume required for the facility within that available in a single TREAT fuel element. By housing the loop complex within a shell equivalent to two TREAT elements, a simplification of the loop geometry was achieved, resulting in a closed-circuit, direct-flow recirculating system. The simplified loop affords an uncluttered view of the fuel-containing test section, lower hydraulic losses in the sodium flow, ease of instrumenting the inlet, interior and outlet of the test section, and less difficulty in assembly and disassembly of successive handling.

The Mark-II loop has been designed according to the ASME Boiler and Pressure Vessel Code specification of a Class A Nuclear Vessel,¹⁴ to exceed requirements for a rating of 1500 psi at 1000°F. The loop body, piping, hydraulic attachments, and accessory components are made entirely of Type 316 stainless steel, which is the highest strength material at 1000°F readily available, and provides full compatibility with sodium and high weldability. The exception in the loop system is the sodium pump, and closure and joining flanges, which are fully heat-treated Inconel X-750. Although the Inconel X-750 material has not been qualified for the ASME code, the maximum allowable design stress for the superalloy was calculated according to the most conservative method used by the Code, and an over-design factor added in the design of the pump tube.

The sodium pump is a miniaturized, modified version of the Annular Linear Induction Pump (ALIP) designed and constructed by the British.¹⁵ It is a 4-pole, 3-phase, 60-cycle, 230-V a.c., approximately 50-amp/phase, 12-yoke induction pump, with a central core of high-Curie-point magnetic material defining a 0.150-in. annulus of driven sodium. The pump walls are 0.100 in. thick and are surrounded by an evacuated multilayered stainless steel Dewar heat barrier, which insulates the magnetic yokes and driving coils from the loop heat. Yokes and coils are cooled by air blast. The pump tube is joined to the loop body with Inconel X-750 autoclave flanges, sealed with Type 316 stainless steel solid O-rings.

The flow cross section of the loop will be equivalent to a 0.750-in.-ID tube.

Sodium flow is measured, at test section inlet and outlet, by high-gauss electromagnetic flowmeters. These consist of multiturn electromagnets, energized by high-capacity, constant-current power supplies. Variable impedance pressure transducers (cooled to prevent unacceptable thermal gradients) are located on the standard NaK-filled standoff assemblies at inlet and outlet.

¹⁴Section III, "Nuclear Vessels," ASME Boiler and Pressure Vessel Code, 1965 edition.

¹⁵L. R. Blake, Conduction and Induction Pumps for Liquid Metals, J. British Nuclear Energy Conf., 2, 306 (1957).

The fuel-holder consists of a rigid tubular member, preassembled with the blind closure plate at the top of the loop, and inserted into the test section through the single top port. A seal between fuel holder and test section is made at the bottom of the test section, with a hemispherical contour stressed into a conical section by stacked Inconel Belleville washers compressed by the camming action of the Conoseal autoclave top closure flange. Fuel thermocouples are brought through the closure plate with a miniature version of the thermocouple exit assembly previously designed and used in the large TREAT loop test section.

The test section is sufficiently simple to ensure ready preassembly, either on the work bench or within a hot lab cave when remote handling is necessary. Interchangeable fuel holders will be designed for experiments covering the full range of fuel-sample configurations and types.

The test section of the loop is to be located on the centerline of the reactor during experiments, with the longer axis of the 4 x 8-in. dummy-element cross section perpendicular to the north-south viewing slot. Inlet and outlet instrumentation will be clear of the test section to prevent nuclear shadowing.

The loop sodium is contained in a separate reservoir integral with the loop system and connected to the loop proper by valved high-pressure lines. Reservoir capacity is approximately twice the fluid volume of the loop, and a minimum of 150% loop volume of sodium total will be contained initially. Designed to the same pressure-temperature criteria as the loop, the sodium reservoir serves as a multipurpose chamber: as a supply and dump tank, and as a safety expansion chamber.

Connecting sodium flow lines between the reservoir and loop are controlled by remotely operated bellows-sealed high-pressure valves, backed up by sodium freeze plugs. The valves are actuated by double-acting pneumatic cylinders. The freeze valves are designed to be gas-cooled if necessary to ensure closure, and are electrically heated for open operation.

The bottom fitting is simply two standard TREAT fuel-element bottom fittings. The top has been changed from the standard TREAT gripper cap to an enlarged flange extended into the upper plenum of the reactor. The loop has been designed to use the large TREAT loop coffin when highly irradiated fuel is inside the test section. A shipping container for samples without prior irradiation is to be designed.

B. TREAT

1. Operations

Four preirradiated EBR-II-sized UO_2 samples clad with stainless steel were subjected to transient irradiations in transparent capsules to obtain data for comparison with that previously obtained on similar unirradiated samples.

Transient testing of SEFOR sample 41-D for the General Electric Company was completed. This sample contained several fuel thermocouples. The main purpose of the test was to investigate the transient performance of these thermocouples.

Several steady-state runs were made to obtain calibration data for future metal-water reaction experiments involving stainless steel and Zircaloy-clad UO_2 fuel samples in transparent capsules. These experiments are designed to obtain photographic data on the physical behavior of the samples while the metal-water reaction is in progress.

2. Large TREAT Loop

Because of the undesirability of having water in the system, in place of a hydrostatic test prior to filling with sodium, the loop was remotely pneumatically pressure-tested at 180 psig to verify that all components, pipe anchors, etc., were adequate for operation at the loop design pressure of 150 psig. Normal steady-state operating pressure will be less than 75 psig. No components failed during the test. However, inspection following the tests showed that one expansion joint had experienced excessive elongation. This joint was replaced, and tie rods were installed to limit the extension of the new bellows.

A critical review of the characteristics of the thermal expansion of the interconnecting piping between the main system and the purification system indicated that the capabilities of two expansion joints installed in this piping were marginal. New units have been received, and installation is in progress.

A destructive test was performed upon a 3-in., three-ply bellows to determine its performance capability for service in the loop crossover piping. A permanent distortion of the bellows did occur with the application of 1170 psi for 70 sec. However, a massive rupture of the bellows did not occur. The only breach of containment that was encountered resulted from a pin-hole leak caused by forcing of the bellows convolution against a restrainer bracket. These conditions are considerably in excess of the calculated maximum pressures for the maximum credible accident. Consequently, the test demonstrates the capability of the unit for the service intended.

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

1. Interaction of Fuel and Cladding with Liquid Sodium

Experiments have been conducted to determine the nature and magnitude of safety problems that might arise in the event that abnormally hot fuel and cladding materials come in contact with the coolant (usually sodium) in a fast reactor. Small heated samples of fuel or cladding materials were dropped into heated sodium in a survey study which identified a fragmentation mechanism that results in extensive subdivision of molten metals of interest to fast reactor technology (see Progress Report for May 1965, ANL-7046, p. 81, and for January 1966, ANL-7152, p. 90).

Fig. 16. Particle-size Distribution of Type 304 Stainless Steel Samples Dropped into 250°C Sodium

More recently, this investigation has been concerned with determining the mean particle sizes of the various metal residues, particularly those of the Type 304 stainless steel samples, resulting from the fragmentation phenomenon. The metal residues were first sieved, using sieve screen sizes from 1 to 500 mils, and then the several fractions were weighed. From the weight-size distribution of each "drop," the mean size was calculated by the relationship

$$d_{sv} = \frac{1}{\sum \frac{w_i}{d_i}}$$

where d_{sv} is the Sauter mean diameter and w_i the fraction of particles by weight which have the diameter d_i .

Figure 16 gives the mean particle sizes of the residues of Type 304 stainless steel samples heated to various temperatures and dropped into 250°C sodium. The temperatures of the samples ranged from 1425 to 2400°C when they were dropped. It can be seen from Fig. 16 that, when the drop temperature exceeds 1600°C, total fragmentation takes place with a mean particle size for the residues which approaches about 10 mils for sample temperatures greater than 2000°C.

Samples of Type 304 stainless steel were also dropped into liquid sodium which had been heated to temperatures as high as 820°C. The mean particle sizes determined for the residues from these experiments were as follows:

Sodium Temperature (°C)	Sample Drop Temperature (°C)	Mean Particle Size, d_{sv} (mils)
250	2150-2400	~10
720	2200	6
800	2200	4
820	2300	3

Thus, the effect of the higher sodium temperature was a more thorough fragmentation of the residue.

Determinations of mean particle size were also carried out for Type 304 stainless steel residues from drops into molten lithium and potassium. These measurements indicated that fragmentation in lithium was slightly less extensive than in the sodium under similar conditions, whereas fragmentation in potassium was substantially the same.

The fragmentation mechanism resulting in the extensive subdivision of molten metals described above was previously delineated (see ANL-7152, p. 90). A quantitative measure of the degree of fragmentation is now available to accompany this mechanistic understanding.

2. Calculations for the Sodium-Air Reaction

Rational safety analyses for fast reactors require knowledge of the temperatures and pressures generated during sodium combustion in air. Combustion of both pools and sprays of sodium will have to be considered. Baker and Tevebaugh¹⁶ pointed out that, regardless of the burning mechanism, the pressures and temperatures are limited by the thermodynamic properties of the system. Calculations have been carried out for equilibrium conditions in sodium-air and sodium-air-water mixtures by means of a computer program designed to minimize free energy.

The method used is that originated by White, Johnson, and Dantzig¹⁷ and extended by Oliver, Stephanou, and Baier¹⁸ to include condensed phases. The computer program was designed to use thermodynamic data from the JANAF¹⁹ tables. Briefly, the equilibrium distribution of species in a system of fixed elemental composition at a specified pressure (or volume) and temperature is determined by minimizing free energy subject to constraints imposed by chemical balance. At constant pressure, the Gibbs free

¹⁶Chemical Engineering Division Semiannual Report, January-June 1965, ANL-7055, p. 208.

¹⁷W. B. White, S. M. Johnson, and G. B. Dantzig, J. Chem. Phys. **28**, 751 (1958).

¹⁸R. C. Oliver, S. E. Stephanou, and R. W. Baier, Chem. Eng. **69**(4), 121 (1962).

¹⁹JANAF Thermochemical Tables, The Dow Chemical Co., Midland, Mich. (1965).

energy is minimized, while at constant volume, the Helmholtz free energy is minimized. (This is an extension of the work reported by Oliver, Stephanou, and Baier¹⁸ in that they describe only constant-pressure calculations.) The equilibrium temperature is then found from a heat balance assuming adiabatic conditions. The composition of the mixture at the equilibrium temperature can then be computed by interpolating the thermodynamic data to find the composition giving the minimum free energy at that temperature.

A computer program to minimize the free energy was developed and used to repeat the calculations of Baker and Tevebaugh,¹⁶ who found the compositions which simultaneously satisfied equilibrium constants and the material balance equations. The equilibrium adiabatic temperature was found graphically using a heat balance similar to that used herein. A comparison of results is shown in Table XXIII. Very good agreement was found, as expected.

TABLE XXIII. Comparison of the Baker and Tevebaugh (B-T) and Free-energy-minimization (FEM) Calculations of Adiabatic Pressures and Temperatures for Sodium-Air Reaction

Initial Conditions: Air: 12.2 psia; 25°C
Sodium: 427°C; liquid

Z Sodium-Oxygen Ratio (g atoms Na/mole O ₂)	Adiabatic Temperature (°C)		Total Pressure (atm)	
	B-T	FEM	B-T	FEM
1	1257	1259	4.18	4.18
2	1707	1689	5.40	5.34
4	1902	1918	6.35	6.53
5	1947	1943	7.05	7.04
6	1877	1863	7.49	7.47
7	1660	1607	7.77	7.57
Air initially at 14.7 psia				
4	1930	1943	7.47	7.68

Since the values of equilibrium temperature and pressure are to be used in conjunction with safety analyses, the machine calculation was repeated for an initial pressure of 14.7 psia and a nitrogen-to-oxygen ratio more truly representative of air (namely, 3.7733). The calculations were done for dry air and for air saturated with water at 25°C; the results are being given in Tables XXIV and XXV.

TABLE XXIV. Calculated Adiabatic Temperatures and Pressures for Sodium-Air Reaction

Initial Conditions: Air: 14.7 psia; 25°C
Sodium: 427°C; liquid

Z Sodium-Oxygen Ratio (g atoms Na/mole O ₂)	Adiabatic Temperature (°C)	Partial Pressure (atm)				Total Pressure (atm)
		N ₂	O ₂	NaO	Na	
1	1292	4.15	0.82	0.00	0.00	4.97
2	1729	5.31	0.73	0.10	0.21	6.35
3	1882	5.71	0.50	0.21	0.69	7.11
4	1961	5.92	0.26	0.27	1.32	7.78
5	1987	5.99	0.08	0.22	2.11	8.40
6	1901	5.76	0.00	0.06	3.12	8.94
7	1634	5.05	0.00	0.01	4.02	9.08
8	1335	4.26	0.00	0.01	4.53	8.80

TABLE XXV. Calculated Adiabatic Temperatures and Pressures for Sodium-Air-Water Reactions

Initial Conditions: Air: 14.7 psia; 25°C
(saturated with H₂O)
Sodium: 427°C; liquid

Z Sodium-Oxygen Ratio (g atoms Na/mole O ₂)	Adiabatic Temperature (°C)	Partial Pressure ^a (atm)					Total Pressure (atm)
		N ₂	O ₂	NaO	Na	NaOH	
1	1177	3.72	0.67	0.00	0.01	0.30	4.70
2	1766	5.24	0.61	0.05	0.12	0.42	6.44
3	1889	5.56	0.41	0.08	0.67	0.45	7.17
4	1901	5.59	0.22	0.08	1.41	0.45	7.75
5	1836	5.42	0.08	0.05	2.25	0.44	8.24
6	1576	4.75	0.05	0.02	3.11	0.38	8.31
7	1294	4.03	0.03	0.01	3.66	0.32	8.05

^aEquilibrium pressures of H₂ and H₂O were each less than 0.005 atm.

The species considered to be present when water was added were, in addition to those already noted, H₂(g), H₂O(g), and NaOH(g).²⁰ The calculations showed that virtually all of the hydrogen initially present in the water was found in the NaOH.

The maximum pressures and temperatures thermodynamically possible are not very different from those prescribed by Baker and Tevebaugh.¹⁶ The values for dry air and for air saturated with water at 25°C are summarized in Table XXVI. The added water has the effect of lowering both the maximum temperature and pressure attainable.

²⁰No new condensed phases needed to be considered over the range of Na/O₂ ratios under investigation.

TABLE XXVI. Maximum Adiabatic Temperatures and Pressures for Sodium-Air Reaction

Initial Water Content of Air	Peak Temperature (°C)	Sodium-Oxygen Ratio at Peak Temperature (g atoms Na/mole O ₂)	Peak Pressure (atm)	Sodium-Oxygen Ratio at Peak Pressure (g atoms Na/mole O ₂)
0	1987	5	9.08	7
Saturated at 25°C	1901	4	8.31	6

It is interesting to compare these results with those reported by Humphreys²¹ as is done in Fig. 17. It is clear that the experimental pressures found by Humphreys are all below the theoretical pressures calculated here. In a number of cases, however, very nearly the theoretical maximum pressure is reached. The high values calculated by Humphreys seem to have resulted from his assumption of complete reaction. However, the above calculations show that, for a sodium-oxygen ratio of 4, for example, 21.1% of the sodium is unreacted at equilibrium and is present as Na in the vapor phase.

Fig. 17. Comparison of Maximum Theoretical Pressures for Sodium-Air Reaction with Data of Humphreys

D. Plutonium Volatility Safety

1. Chemistry of Tellurium Fluorides

Preliminary studies on the sorption of tellurium hexafluoride on activated alumina have been carried out at 200, 300, and 400°C, in addition to those made at 25 and 100°C (see Progress Report for May 1966, ANL-7219, p. 89). The results of these tests indicate that the rate and extent of TeF₆ sorption on activated alumina is essentially independent of temperature between 25 and 300°C. A decrease in sorption rate observed at 400°C may be attributed to a decrease in surface area of the alumina.

Similar experiments have been performed to determine the sorption characteristics of TeF₆ on activated charcoal at temperatures from 25 to 400°C. Preliminary results indicate that charcoal is a good sorbent for TeF₆ at temperatures below 200°C. At higher temperatures, the data suggest that TeF₆ undergoes a reaction with charcoal.

²¹J. R. Humphreys, Jr., Sodium-Air Reactions as They Pertain to Reactor Safety and Containment, Proc. Second Intern. Conf. on Peaceful Uses of Atomic Energy, Geneva (1958), Vol. 11, p. 177.

2. Disposal of Gaseous Fluoride Volatility Reagents

A factorially designed experiment is under way in a 2-in.-dia fluid-bed reactor to study the effect of temperature, gas concentration, gas velocity, bed particle size, and bed height on the efficiency of removal of fluorine from a gas stream in a fluidized bed of activated alumina. Preliminary analyses of the first 8 of 16 runs indicate that, within the ranges of the variables studied, increases in temperature (from 300 to 400°C) and bed height (L/D from 3 to 6) have positive effects on the capacity of the activated alumina for fluorine. Upon completion of the 16 experiments the effect of particle size and possible interactions of all the independent variables will be determined.

V. PUBLICATIONS

Papers

Absorption Spectrum of Neptunium Hexafluoride

M. J. Steindler and T. J. Gerding

Spectrochim. Acta 22, 1197 (June 1966)

Critical Evaluation of Fast-fission Cross Sections

W. G. Davey

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Techniques for Extrusion of Small Diameter Refractory Metal Tube Blanks

W. R. Burt and R. M. Mayfield

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British Nucl. Energy Soc., Ref:2/2, pp. 1-18

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The Effect of Deadtime on Counting Errors

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Measurement of the Li^6 and B^{10} Neutron-absorption Cross Sections by the Shell Transmission Method

S. A. Cox

Bull. Am. Phys. Soc. 11, 654 (June 1966) Abstract

COINC, Computer Code for Reduction of Coincidence Counting Data

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Nucl. Sci. Eng. 25, 306 (July 1966) Note

Computer Code for Reduction of Coincidence Counting Data

Alexander DeVolpi, K. G. Porges, and G. C. Jensen

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Comparison of Multilevel and Single-level Effects for a Fissionable Isotope

E. M. Pennington

Bull. Am. Phys. Soc. 11, 659 (June 1966) Abstract

Low Flux Determination of Capture-to-fission Ratio

W. C. Redman and M. M. Bretscher

Bull. Am. Phys. Soc. 11, 661 (June 1966) Abstract

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Progress in the Development of Fast-reactor Metal Fuels

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Compatibility Studies of Uranium-Plutonium-Zirconium Fuels with Iron-base Cladding Alloys

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- ANL-7166 SAFETY ANALYSIS FOR THE ZPR-9 FACILITY [An Addendum to ANL-6271, Safety Analysis Report, Argonne Fast Critical Facility (ZPR-6)]
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