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MASTER

Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

November 1966

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

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ANL-7279
Reactor Technology
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AEC Research and
Development Report

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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

November 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:

May 1966	ANL-7219
June 1966	ANL-7230
July 1966	ANL-7245
August 1966	ANL-7249
September 1966	ANL-7255
October 1966	ANL-7267

REACTOR DEVELOPMENT PROGRAM

Highlights of Project Activities for November 1966

EBWR Plutonium Recycle Program

EBWR has reached a power of 42 MWt with the control rods 10 in. below their maximum allowable heights and with the reactor water poisoned with boric acid to a concentration of approximately 5 g/gal. Measurements of the reactor transfer function performed during the approach to 42 MWt indicate that the reactor can be expected to be dynamically stable for powers up to at least 90 MWt.

EBR-II

Run No. 22 was completed on November 14 with the accumulation of 1045 MWDt (10,925 MWDt and 68,757,000 kWHe total to date). Run No. 23, scheduled for 690 MWD, has started. Thirty fuel-subassembly transfers were completed in 18 hr, and the reactor was made critical for control rod calibrations on November 15. Minor maintenance was also performed during this shutdown, and the reactor resumed power operation on November 16, a total shutdown of less than 3 days.

The cleaning operations performed on the rotating plug seals to remove oxide dross were apparently very effective. Operation was very smooth throughout the fuel-handling operation, with no indication of drag or resistance at any time.

ZPR-3

Experiments are in progress with Assembly 48, a 400-liter, plutonium-fueled core. Radial traverses of neutron detectors were completed, and a series of heterogeneity experiments to measure reactivity changes, resulting from bunched and unbunched fuels, and sodium voids is in progress.

ZPPR

Fifty-one of the 54 piling foundation holes have been drilled. The south wall of the support wing has been poured, as well as parts of the walls for the vault-workroom area and the service wing. Reactor bed and tables were inspected at the vendor's plant, shipped, and had arrived at the Idaho site by the end of the month.

AARR

Supplement 1 to the PSAR, answering AEC questions on the facility, was issued on November 9, 1966.

Modifications of the reactor vessel internals to accommodate the HFIR core have been initiated.

Analysis of flow coastdown of the primary system indicates that the flow will be down to 13% of rated flow in 20 sec with 2 pony motors operating.

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I. PLUTONIUM UTILIZATION

A. Research and Development

1. EBWR Plutonium Recycle Program

EBWR has reached a power of 42 MWt with the control rods 10 in. below their maximum allowable heights and with the reactor water poisoned with boric acid to a concentration of approximately 5 g/gal. If the boric acid were removed and if the control rods were withdrawn to their maximum allowable height, it is estimated that a power of approximately 70 MWt could be attained. Measurements of reactor transfer functions performed during the approach to 42 MWt indicate that the reactor can be expected to be dynamically stable for powers up to at least 90 MWt. As significantly higher powers can be attained without compromising safety, permission for steady-state operation at powers up to 70 MWt has been requested.

2. Transfer Functions for Plutonium-loaded EBWR

Transfer functions were measured for power levels of 15, 28, and 38 MW. Extrapolation to phase margin stability indicates the reactor will be stable at power levels over 90 MW.

A comparison of the phase margin information obtained so far and that obtained during the approach to 100-MW power of EBWR indicates the reactor may be stable to power levels higher than the 120 MW predicted for the 100-MW core.

II. LIQUID-METAL FAST BREEDER REACTORS

A. EBR-II

1. Operations

Routine 45-MW operation continued throughout the month except for a 3-day refueling shutdown between Runs No. 22 and 23. Run No. 22 terminated November 14 after accumulating the scheduled 1045 MWd. Run No. 23 started November 17. After the step-wise escalation to power to obtain power-coefficient data, operation continued.

The reactivity decrement at 45 MW was measured as 65 lh for Run No. 23 (overall power coefficient of $65/45 = 1.44$ lh/MW). This is 4 lh lower than the value measured during Run No. 22. There is some evidence that this reduction may be due to a more loosely structured core as shown by a less pronounced change in power coefficient in the range of 20 to 25 MW and the near absence of the 10-cps oscillation.

Fifteen subassemblies were removed from the core, including XO09 which had obtained its goal exposure of 5100 MWd, and 15 subassemblies were loaded into the core during an 18-hr period after the rotating plug seals reached the molten condition. Experimental subassemblies XO15 and XO17 were included in the loading for Run No. 23; the experiment goals are 11,000 and 6,500 MWd, respectively.

Materials irradiation subassembly XO13, which had been removed from the storage basket in October, was disassembled in a hot cell prior to sodium removal. The inlet filter and orifice were removed, inspected, and found to be clean.

An MI-type electrical penetration (No. 23) and a coaxial-type electrical penetration (No. 35) were modified to provide permanent double flanges for annual leak-rate testing. Both penetrations were satisfactorily leak-rate tested. Six other electrical penetrations and the reactor building vacuum breaker valve were satisfactorily leak-rate tested.

a. Rotating Plugs. On November 14, the rotating plug seals were fully molten. The small plug rotated freely $3\frac{3}{4}$ hr after the heater control was set for fully molten operation, and the large plug rotated freely after 4 hr. Actually, alloy temperatures indicated molten conditions within 2 hr after the heaters were energized for melting, but it was not possible to attempt rotation at that time because of conflicting work on instrumentation. No "jogging" was required to initiate plug rotation, and operation was very smooth, with no indication of seizing or increased resistance at any point in rotation. Ammeter indications of drive-motor armature current showed normal operation of each plug drive. Subsequent plug performance during fuel handling was normal in all respects.

Measurements of alloy temperature profiles in both troughs after completion of fuel handling showed uniform temperature distribution throughout each trough. In addition to the major components, emission spectrographic analysis of two alloy samples taken on October 6 (see Progress Report for October 1966, ANL-7267, p. 6) showed trace amounts of silver, copper, iron, magnesium, nickel, lead, and silicon. No chromium was found.

b. Experimental Irradiations. The status of all experimental irradiations at the start of Run No. 23 and the approximate exposure at month's end is as follows.

Subassembly	Date Loaded	Capsule Content and Number of Capsules ()	Experimenter	Approximate Accumulated Exposure (MWd)	Goal Exposure (MWd)
XG02	7/16/65	UO ₂ -20 w/o PuO ₂ (1) Stainless Dummies (18)	GE	9,004	13,600
XG03	7/16/65	UO ₂ -20 w/o PuO ₂ (2) Stainless Dummies (17)	GE	9,004	19,450
XG04	7/16/65	UO ₂ -20 w/o PuO ₂ (2) Stainless Dummies (17)	GE	9,004	39,000
XG05	9/3/65	UO ₂ -20 w/o PuO ₂ (9) U-15 w/o Pu-10 w/o Zr (1) U-15 w/o Pu-10 w/o Ti (1) UC-20 w/o PuC (3) Structural (5)	GE ANL ANL ANL GE	8,577	10,300
XG06	9/3/65	UO ₂ -20 w/o PuO ₂ (12) U-15 w/o Pu-10 w/o Zr (1) U-15 w/o Pu-10 w/o Ti (1) Structural (5)	GE ANL ANL ANL	8,577	20,600
XA07	10/27/65	U-14 w/o Pu-11 w/o Zr (16) Structural (3)	ANL ANL	7,840	a
XA08	12/13/65	UC-20 w/o PuC (8) Structural (11)	ANL	6,755	19,800
XO10	3/24/66	UO ₂ -20 w/o PuO ₂ (4) Structural (11) Structural (4)	GE ANL PNWL	5,935	19,600
XO11	5/9/66	UO ₂ -20 w/o PuO ₂ (7) UO ₂ -20 w/o PuO ₂ (9) 304 SS-30 v/o PuO ₂ (1) 304 SS-20 v/o PuO ₂ (1) 304 SS-20 v/o UO ₂ (1)	ANL GE PNWL PNWL PNWL	4,354	8,300
XO12	8/10/66	UO ₂ -20 w/o PuO ₂ (19)	NuMEC	2,235	20,600
XO14	7/17/66	Structural (17) Graphite (1) Graphite (1)	PNWL NRL GE	2,934	b
XO15	11/15/66	PuO ₂ -UO ₂ (11) PuO ₂ -UO ₂ (2) (Pu,U)C (4) Mk-1A (Metal) (2)	NuMEC GE ANL ANL	580	11,000

Subassembly	Date Loaded	Capsule Content and Number of Capsules ()	Experimenter	Approximate Accumulated Exposure (MWd)	Goal Exposure (MWd)
XO17	11/15/66	PuO ₂ -UO ₂ (11) (Pu,U)C (3) Mk-1A (Metal) (5)	NuMEC UNC ANL	580	6,500
U-1550X	5/6/65	Alpha Monitor	ANL	9,875	9,120 ^c
U-1551X	5/6/65	Alpha Monitor	ANL	9,875	9,120 ^c

^aTo be removed at the end of Run No. 23 at experimenter's request.

^bMaximum attainable before core has reached terminal size.

^cMinimum acceptable.

Subassembly XO09 was removed during the shutdown preceding startup of Run No. 23. The subassembly contents and exposures are as follows:

3 caps UNC (Pu,U)C	~3.0 a/o burnup
3 caps ANL (Pu,U)C	~3.1 a/o burnup
2 caps ANL PuO ₂ -UO ₂	~2.8 a/o burnup
2 caps PNWL PuO ₂ -SS (cermet)	~3.3 a/o burnup
2 caps GE Structural Materials	~2.0 x 10 ²² nvt ^a
4 caps ANL Structural Materials	~2.0 x 10 ²² nvt ^a

^aTotal integrated flux.

Subassemblies XO15 and XO17, containing fuel capsules as shown in the status table above, were loaded into grid positions 4A2 and 4C3, respectively.

Thirteen UO₂-PuO₂ (Group-8A) capsules from General Electric were received at Illinois and are undergoing nondestructive testing inspection.

2. Reactor Improvements

a. Oscillator Rod. Alignment of the Mark-II oscillator-rod assembly during reactor operation at 45 MW was completed. There is still a binding problem associated with oscillator rotation, but it is apparently caused by temperature-induced distortion of the oscillator rod in its thimble. The rotation is free below a reactor power level of 30 MW and free at 45 MW if the rotational speed is great enough to equalize temperatures in the rod itself.

The oscillator was calibrated statically as a function of angular rotation at zero power. The wave shape appears to be symmetric, with very little harmonic distortion. The wave shape will be analyzed in greater detail prior to use of the oscillator. The peak-to-peak reactivity worth

measured by control-rod calibration was 22.7 Ih, and by period calibration 21.8 Ih. This agrees well with the measurements performed on ZPR-3 with the EBR-II mockup assembly.¹ The rod was also statically calibrated at 45 MW using a calibrated control rod and measured as 23.2 Ih. This agreement is well within the accuracy of reactivity measurements at power. It does appear that the worth of the rod is not a function of power.

b. Improved Pump for Secondary System. The pump manufacturer has completed the design of an improved pump (see Progress Reports for September 1965, ANL-7105, p. 2; and January 1966, ANL-7152, p. 2) and its construction has been underway for several months. Work is severely hampered by a 7-week strike at the plant.

Construction had been started on all major parts of the pump, with such items as the stator windings and water-cooling system near completion. The pump duct, transition pieces, and expansion joints are not as far advanced as the above items. The vendor has estimated that the pump will be completed approximately 12 weeks after the strike is terminated.

c. Instrumented Subassemblies. A program is under way for the development of EBR-II instrumented subassemblies to enhance EBR-II as a fast test reactor facility for the development of fuels and materials for the AEC's Liquid Metal Fast Breeder Program. According to the basic concept, one or more control rods will be replaced with instrumented test subassemblies, employing the normal EBR-II fuel-handling system.

The status of the work to date is as follows:

(i) A fuel-subassembly arrangement similar to the EBR-II Mark-B subassembly can be used. There would be flexibility for incorporating 7, 19, or 37 capsules within a 1.907-in. hexagonal tube.

(ii) The number of sensing instruments (sensors) possible for installation within a particular subassembly would depend largely on the number of leads that may reasonably be installed within the available area of the subassembly. In the preliminary study, twelve 1/16-in.-dia leads have been prescribed.

(iii) A lead-severance scheme in which sheathed leads would be cut at the top of the instrumented subassembly is being pursued and appears practicable.

(iv) Installation procedures have been investigated. The currently favored method for inserting an instrumented subassembly is that in which a completely preassembled and checked-out subassembly, extension leads, and extension tube are inserted into the reactor as one unit directly through the rotating plug.

¹ANL-6301, Idaho Division Summary Report, July-Sept. 1960, pp. 54-57.

(v) Space relations above the reactor and within the control-rod-mechanism configurations have been studied. It appears that a drive mechanism may be installed within the space left after removing a control mechanism. This drive mechanism would raise the instrumented subassembly approximately 8 ft prior to a fuel-handling operation. When the instrumented subassembly is in the raised position, it will not interfere with the standard fuel-handling operation.

(vi) Currently, the favored removal scheme is to provide a mechanical tube connection between the top of the instrumented subassembly and the extension tube. This connection would be engaged positively at all times, except that it would be disconnected after the instrument leads are cut and prior to removal of the lead extension tube. Positive means are planned for ascertaining that leads are cut completely and the tube is actually separated from the subassembly prior to removal of the extension tube, to preclude inadvertent withdrawal of the subassembly. Removal of the irradiated instrumented subassembly would be done only by the existing fuel-handling mechanisms. The instrumented subassembly will have identical fuel-handling fixtures such that it can be handled by the existing fuel-handling apparatus.

(vii) Procedures for the withdrawal of the radioactive severed extension tube (excluding instrumented subassembly) through the rotating plug have been investigated. These will be much like the existing procedures for the withdrawal of the control rod extension shafts. There is adequate space above the reactor plug for the removal of the long tube sections.

(viii) A successful cutting operation of instrument leads was simulated in a laboratory mockup with the lead cut-off sections exposed to sodium for approximately 12 hr of which 2 hr were at 900°F.

(ix) The status of the art of in-core sensors has been reviewed from the standpoint of availability for and adaptability to a prototype EBR-II instrumented subassembly. On the basis of availability, in-core sensors selected for a prototype subassembly are: an EM flowmeter, coolant thermocouples, and flux detector. These devices are considered sufficiently advanced so that they offer reasonable prospects for installations by January 1969.

3. Reactor Physics

A code has been written to calculate the volume fraction of all material in the various regions of the core, using as input the same reactor-loading data cards as used for burnup calculations. Calculated data have been obtained for the hypothetical case of a 91-subassembly core in which experiments in Row 4 are moved to Row 6 and the reactor is made critical by loading either more experiments or partially fueled subassemblies in Row 4. This would correspond to the core loading which might be expected

if an increase of power to 60-62.5 MW was initiated, requiring the relocation of experiments. As would be expected, the reduced density of fuel in Row 6 due to experiments causes the fission rate to fall off faster than with the reference 91-subassembly core with experiments in Row 4.

4. Neutron Radiography

An experimental evaluation of an antimony-beryllium neutron source for neutron radiography has been completed. This source would be used for nondestructive testing of materials which had been irradiated in EBR-II. The results show that an antimony-beryllium neutron source can be successfully used for neutron radiographic inspection of nonthick specimens.

For the experiment, an antimony oxide source was activated in Row 8 of EBR-II to about 22% of saturation activity. With this source in the center of an approximate cylinder of beryllium ($R = 35$ cm and $Z = 40$ cm), a maximum total flux of 5.2×10^7 nv was measured. The beryllium was about 80% of theoretical density.

A neutron radiograph of a natural uranium penetrometer on a 3/8-in.-thick natural uranium test piece was made using a 0.010-in. dysprosium transfer onto medium-speed X-ray film. The neutrons were collimated by inserting a cadmium tube, 2 x 2 in. in cross section and 22 in. long, into the beryllium to the point at which the maximum flux was observed. The resulting radiograph had a 2-4-T penetrometer sensitivity, and the collimated neutron flux intensity at the test specimen was about 4×10^4 nv. The collimated neutron flux at the test specimen was 6.6×10^4 at the beginning of the experiment.

5. Surveillance

a. Direct Determination of Maximum Burnup Achievable in Mark-IA Design. The maximum burnup achievable in the Mark-IA design will be directly determined by the irradiation of seven encapsulated Mark-IA fuel rods that were placed into EBR-II during November for irradiation in two Mark-A subassemblies. These rods are from a regular manufacturing run intended for EBR-II, and therefore are 14 in. in length and consist of U-5 w/o Fz in Type 304 stainless steel jackets and have a sodium annulus. The irradiation of these rods will be to various levels in excess of 1.5 a/o burnup, a level significantly higher than that presently allowed for the core fuel rods, and some of the rods are to be irradiated to the point of jacket failure. Table I summarizes the status of the irradiations.

b. Segregation Phenomena in the Bismuth-Tin Eutectic Alloy. A quantitative investigation of the Soret effect in the bismuth-tin eutectic alloy has been undertaken as part of a program to provide basic metallurgical information on properties of the alloy in the EBR-II rotating-plug seal.

TABLE I. Status of Encapsulated Driver-fuel Irradiations in EBR-II

EBR-II S/A No.	Rod No.	Fuel Rod Type	Effective Density (%)	Cladding Thickness (in.)	Max Clad Temp (°C)	Burnup to Date	
						U + Pu (a/o)	fiss/cc x 10 ⁻²⁰ ^a
XO15	BF-02	Mark IA	85	0.009	497	0.18	0.65
XO15	BF-03	Mark IA	85	0.009	497	0.18	0.65
XO17	BF-04	Mark IA	85	0.009	501	0.19	0.69
XO17	BF-05	Mark IA	85	0.009	501	0.19	0.69
XO17	BF-08	Mark IA	85	0.009	500	0.19	0.68
XO17	BF-09	Mark IA	85	0.009	501	0.19	0.69
XO17	BF-11	Mark IA	85	0.009	500	0.19	0.68

^aBased on effective density.

Segregated masses of a bismuth-rich phase (99.9 w/o Bi, M.P. ~271°C), which can form as a result of thermal diffusion in the seal alloy during the half-molten condition, have been postulated as a possible cause of the problems of plug rotation. These bismuth masses, if present in the seal, would probably be located at the trough walls and at the eutectic solid-liquid interface (cooler regions in the seal). Plug rotation could be impaired until these masses dissolve through sufficient soaking time in the molten mode of operation. Confirmation of the presence of bismuth-rich material and significant segregation of the liquid alloy has not been possible because of difficulties in obtaining representative samples from the seal.

Winter and Drickamer² have reported that bismuth preferentially diffuses to the cold end of an alloy with an initial composition 50 w/o Bi-50 w/o Sn for an average temperature of 295°C and a ΔT of 50°C (temperature gradient of 40°C/cm).

In our first experiment, a bismuth layer of substantial thickness formed at the original solid eutectic-liquid interface after 4 days at an average temperature of 190°C and a ΔT of 100°C (temperature gradient 53°C/cm). Further experiments will be made to define the maximum layer thickness as a function of average temperature and temperature gradient. An electron microprobe is being used to determine composition changes in the liquid range associated with the layer growth.

Investigations of the solubility, as a function of temperature, of sodium in the eutectic alloy and of the effect of sodium on the oxidation characteristics of the liquid alloy, are being planned.

6. Fuel Cycle Facility (FCF)

a. Production Operations in the FCF. FCF production during November is summarized in the following table:

² Winter, F. R., and Drickamer, H. G., J. Phys. Chem. 59, 1229-1230 (1955).

Production Summary for November 1966

Subassemblies received:	12	
Subassemblies dismantled:	11	
Subassemblies fabricated:	8 ^a	
Subassemblies transferred to reactor:	9	
Pins decanned:	1143	
Melt Refining:	8 irradiated	1 recycle
Pour Yield (av):	92.9	88.8
Injection casting runs:	12	
Pin processing:		
Accepts:	1107	
Rejects:	79	
Pins welded:	1163	
Leak testing:		
Accepts:	1164	
Rejects:	17	
Bond testing (completed runs):		
Accepts:	1036	
Rejects:	48	
Surveillance:	L-425, B-322, B-328, B-330	

^aFive of the fourteen subassemblies reported fabricated in Progress Report for October 1966, ANL-7267, p. 11, are high in iron content and are being withheld pending a decision on further use.

b. Eddy-current Tests of a Modified Fuel-element Restrainer.

A group of 20 fuel elements containing a proposed modified restrainer was fabricated for evaluation of the feasibility of eddy-current sodium-level measurement using the present in-cell equipment. The restrainer used had a reduced stem diameter (0.040 in. compared to 0.080 in. presently used) with no knob at the lower end (see Progress Report for October 1966, ANL-7267, p. 8). A sodium level of 0.35 ± 0.10 in. above the fuel element was aimed for, as compared to a level of 0.65 ± 0.15 in. currently used as a specification.

Element-fabrication operations revealed no unusual problems. Eddy-current testing, X radiography, and destructive examinations of the elements revealed the following: (1) sodium levels within the specification range are discernible with the bond-testing machine; (2) the restrainer stem assumed a position considerably removed from the axial centerline of the element in greater than one-half of the elements fabricated. In a number of elements this misalignment was sufficient to allow the stem to touch the inside surface of the jacket tubing.

c. EBR-II Vertical Assembler-Dismantler (VAD). Design and detailing work on all units of the VAD machine have now been completed, except for the welding head and the Fuel Element Remover and Loader (FERL). An experimental unit of the FERL unit has been built and will be further tested before designs are completed.

The overall construction schedule for the VAD machine is delayed because of difficulties in obtaining on-time commitments from fabricators and suppliers. Based on current delivery promises from contracted vendors, we estimate this delay to be from 5 to 8 weeks. From our findings, heavy work loads and order backlogs are now prevalent throughout the metal-working industry.

In addition to this general procurement situation some problems arose with equipment already on hand. Difficulty has been experienced in obtaining satisfactory welds during trials of the new Miller ES welder, and the unit has been returned to the factory for readjustment. Also, during disassembly of the harmonic drive-speed reducers for machining of keyways, a bent output support web was noticed. This apparently occurred during factory testing; the reducers were returned to the factory for repairs. A six-week delay in the assembly and testing of the vertical drive gear boxes will result from this problem.

Final engineering and fabrication work of the welding head and the FERL unit should be on schedule, since it is planned to complete them just prior to installation of the VAD in the air cell.

Preliminary work on installation drawings, wiring diagrams, and control console has begun.

B. Physics Development

1. ZPR-3

Experiments continued with Assembly 48, the study of a 400-liter, plutonium-fueled, physics core with a degraded neutron spectrum typical of the anticipated large power-reactor cores. This loading contains 334 drawers, including 10 control plus safety drawers with fuel-spiking for safety requirements. The October Monthly Report (ANL-7267) includes all the pertinent specifications for a just-critical "clean" core derived from the experimental results.

Radial traverses of neutron detectors through the assembly were completed. The traverse channel ran through the P-row drawer of Half No. 1 (see Fig. 1) at about 1 in. from the interface. Scans were made through the tube with U^{235} , U^{238} , and plutonium fission chambers, and with a $B^{10}F_3$ proportional counter. The relative fission rates versus radius

for U^{235} and U^{238} are plotted in Fig. 2. Except near the region of the blanket boundary, the profile of fission rates for Pu and B^{10} (n, α) is similar to that measured for U^{235} .

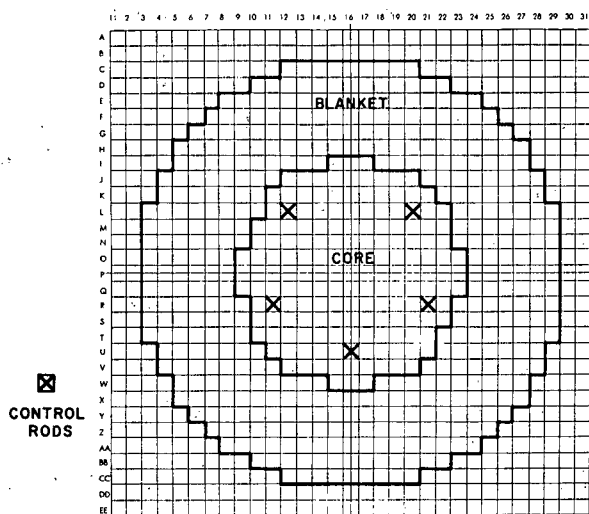


Fig. 1. Half No. 1 of ZPR-3 Assembly 48

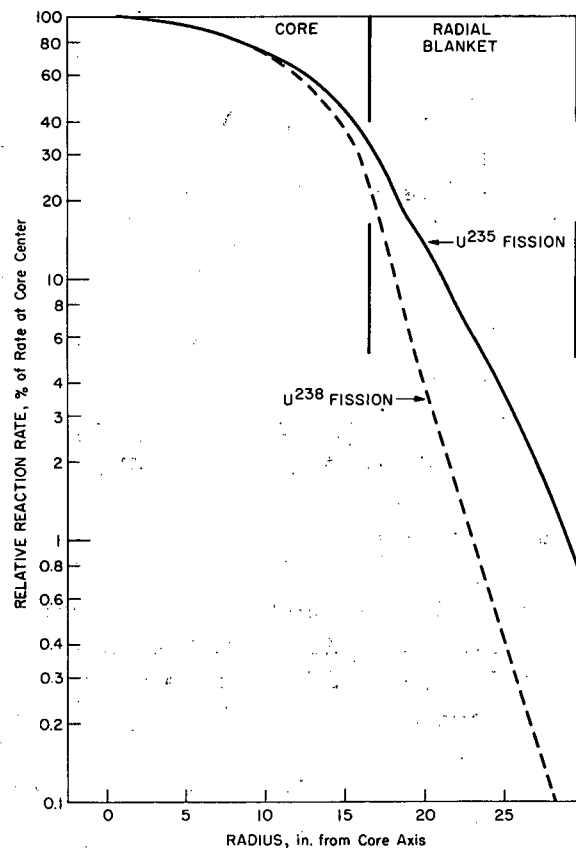


Fig. 2. Radial Profiles of U^{235} and U^{238} Fission Rates in ZPR-3 Assembly 48

A series of heterogeneity experiments is under way to study the effect on the reactivity of the system and on the worth of sodium due to the relative placement of fuel and diluent columns in a core drawer. The experiments include the measurements of the change in reactivity due to bunching and unbunching of the fuel columns, and the voiding of the sodium in the various configurations. Three regions in the core are being examined: the center, the axial edge, and the radial edge. Results will be given when the series of experiments is completed.

2. ZPR-6

a. Sodium-void Measurements. The sodium-void coefficient was remeasured as a function of axial position in Assembly 5. The value previously obtained at the center of the core was found to be about +0.25 lh/kg higher than the presently accepted value. Other axial coefficients changed slightly but were within estimated experimental uncertainties. Figure 3 shows a revised plot of the axial sodium-void coefficients for Assembly 5

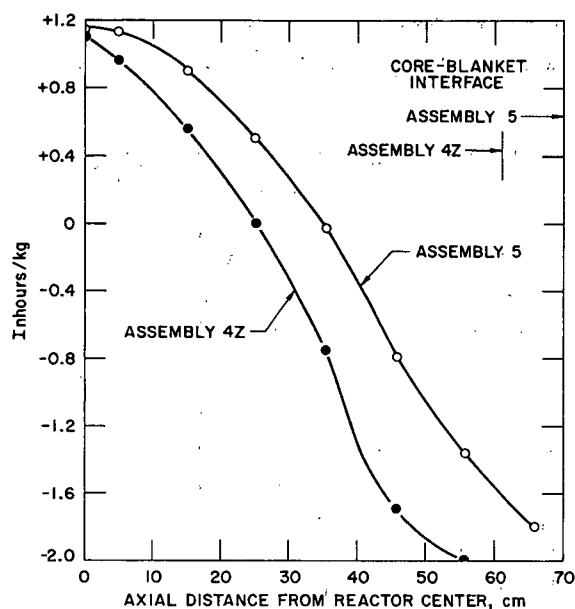


Fig. 3. Reactivity Change due to Sodium Removal

performed in the central 18 drawers (9 in each half). In one set of measurements, the central 8 in. in each half of the core was voided, and in another set the entire 28 in. in each half of the core was voided. Measurements were made for the following configurations:

(i) Normal drawer loading (see Fig. 4).

(ii) A uniform loading using 1/4-in. sodium cans; the remaining reactor materials were retained although their relative positions changed (see Fig. 5).

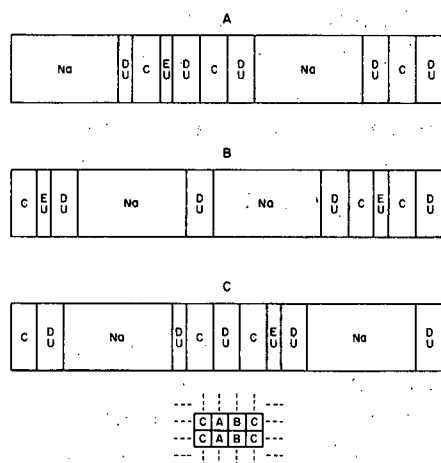


Fig. 4. Normal Drawer Loadings for ZPR-6 Assembly 5

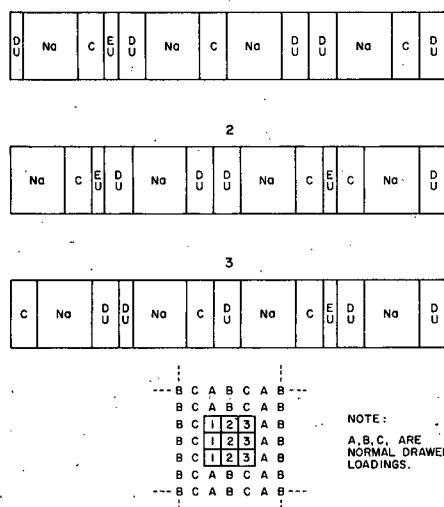


Fig. 5. Uniform Drawer Loadings but with Sodium Cans Reduced to 1/4-in. Thickness

and the data previously obtained for Assembly 4Z. Both sets of measurements have an assigned uncertainty of 0.1 Ih/kg.

Agreement between the Assembly-5 and Assembly-4Z measurements was obtained at the center of the cores. However, the greater axial leakage, due to the reduced core height of Assembly 4Z, results in more negative sodium void coefficients in 4Z at locations away from the axial center of the core.

Other measurements were made to investigate heterogeneity and/or the relative influence of U^{238} on sodium-void coefficients.

These measurements were per-

(iii) A loading which retained the basic Assembly-5 composition but bunched the sodium in a 1-in.-wide column in the center of the drawer (see Fig. 6).

(iv) A loading which bunched the carbon in one-half of the drawer and the depleted uranium in the other half; sodium was then selectively voided from either the carbon region or the depleted uranium region (see Fig. 7).

(v) A loading which eliminated depleted uranium from a five-drawer by five-drawer array in the center of the reactor. The loadings were similar to those of (iv) above except for the elimination of the depleted uranium. In order to retain the spatial positions of the drawer materials, 1/8-in.-thick, 45% density aluminum replaced the 1/8-in.-thick depleted uranium, and 1/16-in.-thick Type 304 SS replaced the 1/16-in.-thick depleted-uranium pieces. (These materials were the best spacer materials available at the time of the experiment.)

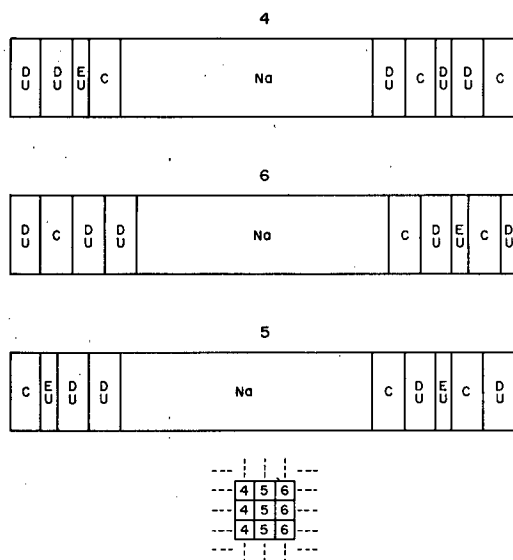


Fig. 6. Arrangement of Core Materials and Drawers to Provide a 1-in. Column of Sodium in the Center of the Drawers

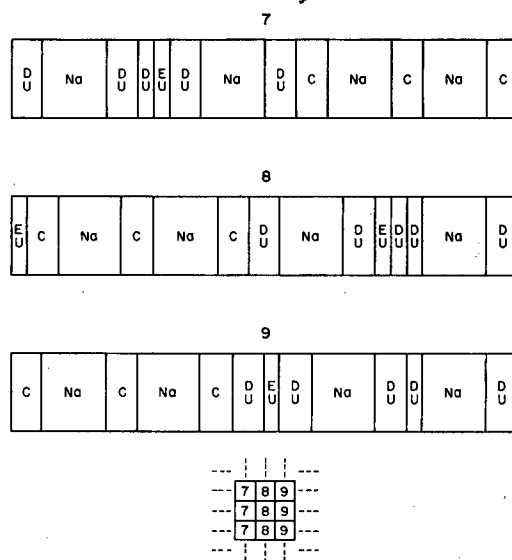


Fig. 7. Rearrangement of Materials in Drawers so that Carbon Is Segregated in One-half and Depleted Uranium in the Other Half. Sodium may be voided from either the carbon or the depleted-uranium region.

The sodium-void measurements were conducted in a three-drawer by three-drawer array so that a one-drawer buffer region (without depleted uranium) existed between the original core and the experimental region.

(vi) A loading using the same basic zone as in (v), i.e., a five-by-five drawer zone without depleted uranium. In this case the sodium was bunched in a one-in.-wide column at the center of the drawer. Again voiding was done in the central 18 drawers, i.e., 9 in each half of the reactor.

The results of these measurements are listed in Table II.

TABLE II. Sodium-void Measurements in ZPR-6 Assembly 5

Configuration	Comments	Central 16 in. Voided		Entire 56 in. Voided	
		Ih/kg	Ih	Ih/kg	Ih
(i) Normal loading (see Fig. 4)	Sodium contiguous to depleted uranium	+1.08	+4.01 ± 0.2	-0.29	-3.95 ± 0.3
(ii) Uniform loading with 1/4-in. sodium cans (see Fig. 5)	Sodium contiguous to depleted uranium and to carbon	+0.97	3.35 ± 0.3	-	-
(iii) 1-in. column of sodium loaded in center of drawer (see Fig. 6)	Void full 1-in. width	+0.70	2.33 ± 0.3	-0.70	-9.52 ± 0.03
	Void two 1/4-in. outside columns	+0.65	+1.10 ± 0.3	-0.49	-2.90 ± 0.30
(iv) Drawer loadings arranged to bunch carbon on one side of drawer and depleted uranium on the other side	All sodium voided in region indicated	+0.65	+2.18 ± 0.3	-	-
	Sodium bounded by carbon voided	-0.28	-0.48 ± 0.3	-1.02	-6.01 ± 0.3
	Sodium bounded by depleted uranium voided	+1.60	+2.67 ± 0.3	+0.14	+0.82 ± 0.3
(v) Central 25 drawers in each half loaded as in (iv), except 1/8-in., 45% density aluminum replaces depleted uranium and 1/16-in. SS replaces 1/16-in. depleted uranium	Voided sodium by carbon in central 18 drawers. 9 in each half.	-5.90	-9.9 ± 0.3	-4.68	-27.7 ± 0.3
	Voided sodium by aluminum in central 18 drawers	-4.95	-8.3 ± 0.3	-4.83	-28.6 ± 0.3
(vi) Same as (v) except sodium arranged as in (iii)	Entire 28- by 1-in. sodium column voided in 18 drawers	-	-	-1.91	-25.9 ± 0.3

Measurements with configuration (iv) yields several bits of information. Firstly, the sodium-void coefficient for that sodium bounded by carbon is negative when the central 16 in. are voided, as well as when the entire axial height of 56 in. is voided. Secondly, the sodium-void coefficient for that sodium bounded by depleted uranium is positive when the entire 56-in. axial height is voided, as well as when the central 16 in. is voided. Thus quite different results are obtained for the two cases. Thirdly, the average of the two sodium-void coefficients, i.e., that in the carbon region and that in the depleted-uranium region, is $(1.60 - 0.28)/2 = +0.61$ Ih/kg, whereas a measurement which voided the same configuration

between the carbon and depleted uranium simultaneously yielded +0.65 lh/kg. Thus the average of the two separate experiments yields the same result within experimental uncertainty as when all of the sodium is voided at once. This is not the same result as the sodium-void coefficient obtained for the normal loading of configuration (i), which gave +1.08 lh/kg. The number of depleted uranium surfaces contiguous to the sodium is the same for configurations (i) and (iv).

In configuration (ii), 1/4-in. sodium-filled cans were utilized; about half (10) of the sodium surfaces were contiguous to carbon and the remainder (12) were contiguous to depleted uranium. The results show the central sodium-void coefficient was only slightly less than that obtained for the normal loading. This indicates that the Assembly 5 loading with 1/2-in.-thick sodium cans yields sodium-void coefficients which are quite close to those obtained using a somewhat more homogeneous configuration utilizing 1/4-in. sodium-filled cans. It is noted that the normal drawer loadings have 11 sodium can surfaces contiguous to depleted uranium. Results for configuration (iii) indicate that at least for the central region the sodium-void coefficient is, within experimental uncertainty, the same regardless of whether 50% or all of the sodium in the region is voided. The results obtained for voiding the entire core length do, however, appear to be dependent on the quantity of sodium removed. This is probably a streaming effect for which neutrons traverse the void from regions of higher to lower importance.

Configuration (v) was arranged to duplicate the bunched-carbon, bunched-depleted-uranium experiment, but with the depleted uranium removed. The results graphically highlight the role of depleted uranium in the results for the sodium-void coefficient. The central sodium-void coefficient is more negative than that obtained by voiding the entire 56 in., both in the vicinity of the carbon and in the vicinity of the aluminum. This is attributed in part to the influence of the depleted uranium in the axial blanket.

Configuration (vi) duplicates configuration (iii) but with the depleted uranium removed as for configuration (v). Again voiding all of the sodium results in a substantially more negative sodium-void coefficient than when depleted uranium is present.

b. Pulsed Measurements. The Kaman pulsed neutron generator was installed in ZPR-6, the drift tube entering the central matrix tube of the stationary half. The tritium target is located in the center of the reactor. Prompt-neutron decay measurements will be made at different locations in the core and reflector of Assembly 5 with a U^{235} fission chamber, a BF_3 counter, and possibly a U^{238} fission chamber and proton-recoil counters. Also, intensity measurements are planned to establish the feasibility of time-of-flight measurements. The prompt decay measurements with the U^{235} chamber are now in progress.

3. ZPR-9

ZPR-9 Assembly 12 is a zoned assembly designed to improve the matching of the central flux and, particularly, the adjoint matching with the reference core, ZPR-6 Assembly 5, over that obtained when using the ZPR-9 Assembly 11. ZPR-9 Assembly 12 was loaded to criticality during the month. Measurements of central reactivity worth were made with various materials as a test of the flux and adjoint spectra. Preliminary analysis of the central reactivity worths obtained indicates that the desired improvement in spectral matching has been achieved.

The assembly is presently cooling to permit the gamma-ray intensity to decay and allow a neutron-spectrum measurement to be made in the core with a proton-recoil counter.

4. ZPPR

At present 51 of 54 post holes for the reactor cell have been finished. The south wall of the support wing has been poured and parts of the remaining walls for the vault-workroom area and the service wing have been poured.

The present status of the reactor components is as follows:

a. Bed and Tables: Two Idaho Division project representatives visited the Giddings and Lewis Machine Tool Co., Fond du Lac, Wisconsin, to witness operating tests with the ZPPR bed and tables. All normal functions of the machine were performed with no load on the tables. The machine was then loaded with 77 tons of concrete blocks on each table, simulating the load distribution of an assembled reactor core. The specification for load capability of the machine is 180 tons, with a stretch capability of 240 tons. Each table is 9 ft long and 14 ft wide. The center of gravity of each load was approximately 3 ft from the front edge of each table. Measurements were made before and during loading to determine deflection and levelness of the tables due to the various loads on the machine. Special tests were performed, which included coasting of the movable table with and without load, slipping each in-drive clutch with the electric scram motor system, checking the table-closure safety clutch, and driving off the hot-rail system.

During the tests the following difficulties were encountered: (1) the slow-speed clutch (1/6-in./min drive) did not perform according to the manufacturer's literature and is being replaced (a clutch identical to the replacement was used during the checkout); (2) the low-speed gear motor heated slightly and required excessive current on one phase to operate, and was sent to the manufacturer's repair shop for correction; (3) the air motor rotated in a direction opposite to that required, and the internals for reversing the rotation have now been supplied by the manufacturer. The surface of the movable tables was manufactured so that it

would be 0.003 in. above the stationary table, since it was assumed that there would be a small amount of compression of the bearings and hardened strips on the runways when the movable table was loaded. Measurements indicated that compression was only 0.0005 in. with respect to the stationary table when the machine was loaded with 154 tons, which is 85% of the full load specified. To bring the two tables into better alignment, the flange on each roller-bearing assembly was ground down 0.002 in.

In summary, after minor adjustments to correct the observed difficulties, the bed and tables were judged to be acceptable.

The machine was boxed and shipped to the Idaho site. The loads shifted approximately 2 to 4 in. during shipping. Although there is no apparent damage, the railroad company was notified accordingly, and the pieces will be opened up and inspected after bringing them into the storage building.

b. Matrix Drawers: The vendor, Mechanical Products Mfg. Co., Seattle, Washington, shipped 800 drawers during the month of November. Of these, 791 were acceptable. At present, approximately 1900 back drawers, 1330 front drawers, and 100 special front drawers have been accepted and are being stored. Because of the number of rejected drawers, the current lot of stainless steel has been exhausted and another lot will be required to fill the order for the remaining drawers.

c. Nuclear Instrumentation and Area Gamma Monitors: The nuclear instrumentation and area gamma monitors have been received and are being stored.

d. Poison Safety Rod Drives: Delivery of two bearings and a microswitch is holding up the assembly and checkout of the first rod drive.

e. Reactor Knees: The four reactor knees have been cast and are now at the machine shop for machining.

f. Personnel Shields: Bids on the personnel shields are due November 30.

g. Matrix Alignment: Review of the alignment-plate requirement indicated some additional drawings were required. These are now being prepared for the bid package.

h. Source Drives: The vendor, Teleflex, Inc., North Wales, Pennsylvania, has submitted drawings for approval. ANL reviewed the drawings and sent back the marked-up drawings. The revised drawings have been reviewed by ZPPR project personnel and have been approved.

i. Rod-drive Mounting Plates: The two rod-drive mounting plates have been machined and are being checked for integrity before drilling the required holes.

j. Beta-Gamma Air-particulate Monitors: The beta-gamma air-particulate monitors have been received. The vendor sent the improper instruction manual. ANL has deferred payment until the proper manuals have been received and the units have been checked out.

k. Drawers Springs: The spring samples submitted by its manufacturer were too long and were rejected. The vendor was instructed to correct the deficiency and to resubmit samples for approval.

l. Matrix Tie-down: All materials have been received and assembly is progressing.

m. Loading Platform: The platform has been welded by the vendor. The remaining items to be finished are drilling the holes and painting.

n. Reactor Frame: The reactor frame is being stored.

o. Reactor Plenum Assemblies: Delivery for the reactor plenum assemblies has been revised to February 1967, due to a large work load in the vendor's shop. Samples of the Neoprene plugs have been received and are being examined.

p. Fuel-control and Safety-rod Drawers and Ball-joint Assemblies: The drawers have been fabricated and are now being Scottsonized to harden the surfaces. The delay of delivery of the monoballs from the manufacturer will delay final assembly of the units.

q. Poison Safety Blades and Sheaths: Work is continuing on the safety blades and sheaths.

r. Control Consoles and Cabinets: The vendor requested substitutions for several control cabinets. The requested substitutions were not approved, so the vendor is fabricating the required units. All consoles and cabinets are ready for shipping. Two Plexiglas paneled doors have not been finished.

s. Boron Fabrication: The bid packages for the boron fabrication are now being prepared.

C. Other Fast Reactor Physics

1. Determination of Burnup of Fast Reactor Fuels

The program for development of analytical methods for the determination of burnup of fast reactor fuels and for the measurement of fast fission yields is continuing. The development of a burnup method using the rare

earth group of fission products as the monitor was reported in the Progress Report for September 1966, ANL-7255, p. 19.

Praseodymium-141 (stable) is a potentially useful burnup monitor; its fission yield is high, the half-lives of its volatile precursors are short, and it is not expected to be a constituent of virgin or reprocessed fuels. However, praseodymium has not been previously considered as a burnup monitor nor has its fission yield been determined because there were no analytical methods with sufficient sensitivity, selectivity, and precision to determine this nuclide in irradiated fuel. Neutron activation would provide the sensitivity and selectivity but is ordinarily not a sufficiently precise technique. However, by incorporating an internal flux monitor in the irradiation, a precision of better than $\pm 1\%$ is attainable.

A procedure for the determination of praseodymium-141 in nuclear fuels has been developed in which manganese-55 is used as the internal flux monitor. The procedure, as presently conceived, consists of the following basic steps:

- a. fluoride precipitation of the rare earths to effect a separation from the bulk of the uranium;
- b. hydroxide precipitation of the rare earths with ammonium hydroxide to remove such fission products as cesium and strontium.
- c. anion exchange separation to remove the last traces of uranium and such fission products as zirconium, niobium, and molybdenum;
- d. perchloric acid fuming to volatilize ruthenium;
- e. cation-exchange separation of cerium and praseodymium from the other rare earths using α -hydroxyisobutyric acid as the elutrient;
- f. separation of cerium and praseodymium from each other on a column containing a mixture of Dowex-1 and lead dioxide;
- g. neutron activation of praseodymium-141 along with the internal flux monitor, manganese-55;
- h. hydroxide precipitation of praseodymium and manganese to separate them from reagent impurities, primarily sodium;
- i. gamma spectrometry of praseodymium-142 and manganese-56.

An evaluation of the procedure has been made using a solution of EBR-I, Mark-III fuel (burnup $\sim 0.3\%$). The relative standard deviation of four determinations was $\pm 1.7\%$. No activities other than praseodymium-142 and manganese-56 were present in the gamma spectrum.

To establish the fast-fission yield of praseodymium it will be necessary to relate the praseodymium content of the fuel to that of a fission product whose yield is known, namely, cesium-137. The cesium-137 analyses, as well as additional praseodymium analyses, are now in progress.

D. Component Development

1. Sodium Technology and Development

a. Dispersion of Carbon in Liquid Sodium. It is believed that almost all of the elemental carbon present in sodium exists as particulates. Accordingly, attempts are being made to prepare carbon dispersions in liquid sodium, controlled as to particle size and concentration, so as to enable study of the behavior of carbon in liquid sodium.

Another attempt (see Progress Report for September 1966, ANL-7255, p. 25) was made to employ decomposition of Na_2CO_3 , added to molten sodium in much smaller amounts than previously, as a means of dispersing carbon. Finely ground, C^{14} -labeled Na_2CO_3 was added incrementally over several days to reactor-grade sodium in a stainless steel crucible. The sodium was stirred continuously at $\sim 550^\circ\text{C}$, a temperature at which Na_2CO_3 is known to decompose. Both filtered (with a 5-micron porosity frit) and unfiltered samples of the liquid were periodically withdrawn and analyzed for total carbon and carbon-14. The analytical results showed that no significant degree of carbon dispersion had occurred.

The results of the two experiments indicate that the decomposition of Na_2CO_3 in liquid sodium, under the conditions used, does not yield carbon dispersions because the carbon forms in an agglomerated state. Visual evidence of this was obtained at the end of the recent experiment by decanting the bulk of the sodium from the crucible and treating the residue with ethyl alcohol: as the sodium reacted, clumps of carbon about 1 to 3 mm in diameter could be seen. Dispersal of carbon black (9 to 250 millimicrons) in liquid sodium will be attempted.

2. Materials Evaluation

a. Physicochemical Mechanics of Metals. Work on the investigation of sodium effects on 304 stainless steel has continued. Measurements were made of samples exposed to 10 ppm and 100 ppm sodium oxide in sodium at 594°C and at 732°C . The volumetric and weight methods were used for determining electrical resistivity. The independent variable in all cases is the duration of the Strauss Test.

In Fig. 8, the upper left-hand graph shows the electrical resistivity (by weight) after a 3-mo exposure in 594°C sodium with 10 ppm and 100 ppm of sodium oxide. The lower left-hand graph gives the electrical resistivity under these same conditions by the usual volumetric method. By either method it is apparent that the lower concentration of sodium oxide has a detrimental effect on the rate of Strauss solution attack on the 304 stainless steel. Characteristically, an electrical resistivity of greater than $200 \mu\Omega\text{-cm}$ after the Strauss test indicates a severely deteriorated specimen which can be retained intact only with great care.

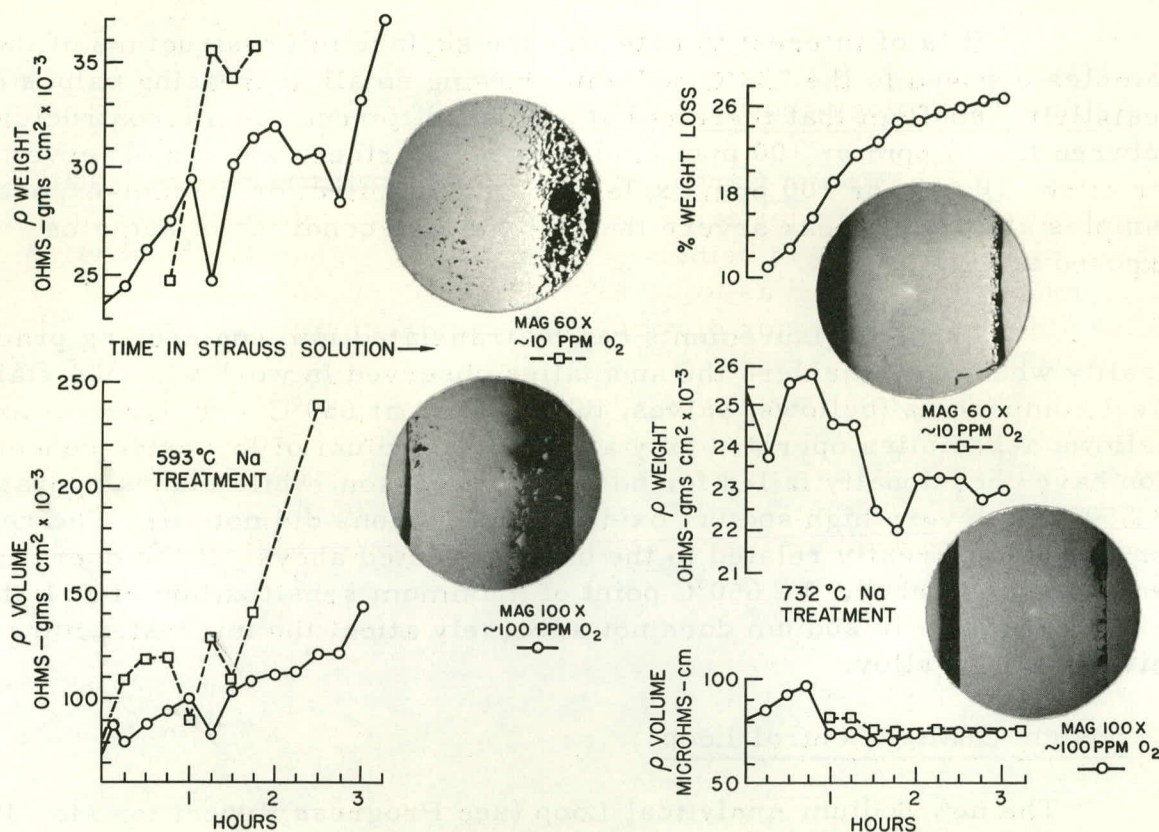


Fig. 8. 304 SS Electrical Resistivity Values after Strauss Test. Reduced 55% in reproduction.

The graphs on the right side of Fig. 8 are similar to those on the left except that the exposure to sodium has been for 3 months at 732°C. The data points on the middle graph show that the rate of attack, as indicated by the weight-method resistivity values, decreases with increased exposure to Strauss solution for the 100 ppm sodium oxide in sodium case. This phenomenon should be compared to the increasing rate values for 100 ppm sodium oxide illustrated at 594°C.

This phenomenon implies an improved microstructure of 304 SS at the higher temperature which may inhibit the infusion of sodium into the grain boundaries. The extreme, initial reaction of sodium-soaked 304 SS with Strauss solution indicates reaction with sodium, hence sodium penetration. The data of the lower right-hand side indicate that at 732°C there is little or no difference in electrical resistivity values by the volumetric method whether the oxide content is 10 ppm or 100 ppm.

Additional data in the upper right-hand corner of Fig. 8 indicate that the weighed (measured) percent weight loss of material is large ($\sim 11\%$) for the first hour of exposure to Strauss solution. Diminishing percent weight losses correspond well to the reduced weight-resistivity values shown in the middle right-hand curve.

It is of interest to note that the surface microstructure of the samples exposed to the 732°C sodium, showing small decreasing values of resistivity, confirm that there is but a small difference in microstructure between the 10 ppm or 100 ppm oxide cases. Further, one can observe that, for either 10 ppm or 100 ppm oxide, the surface effect on sodium-exposed samples at 732°C is less severe than the surface condition of samples exposed at 594°C.

These measurements can be translated into engineering practicality when one considers the anomalies observed in work with 304 stainless steel components (bellows, valves, tubing, etc.) at 650°C and higher in sodium. Bellows assemblies operated only at 650°C in sodium of low oxide concentration have occasionally failed for no apparent reason, while others operated at 850°C with very high sodium oxide concentrations did not fail. The reason for this is apparently related to the data presented above. If the operating temperature is above the 650°C point of maximum sensitization of 304 stainless steel, oxide in sodium does not adversely affect the microstructure of this particular alloy.

3. Sodium Quality Control Loop

The new Sodium Analytical Loop (see Progress Report for May 1966, ANL-7219, pp. 25-26) has been successfully helium mass spectrometer leak tested.

Design work has started on models of sample cups for test in the line sampler. These models will include resistance-heated types and fixed-volume types. The anticipated favorable test results should produce an improved replacement sample cup for EBR-II.

4. Sodium Quality Control Laboratory

Installation of the needed electrical service for the standard, evacuable glovebox and associated gas-purification system has been completed. Approximately 24 hr after initial startup, with a helium atmosphere in the box, the oxygen and moisture monitors read 4 ppm and 9 ppm, respectively. Equipment can be transferred into or out of the glovebox through the evacuable transfer chamber, without causing a noticeable change in the glovebox atmosphere. After two weeks of glovebox use with the purification system in operation, the oxygen and moisture in the system had leveled off at 1-2 ppm and 2-3 ppm, respectively. The common complaint of moisture diffusing through the rubber gloves, when in use, was the greatest factor in atmosphere contamination.

Vacuum distillation runs with sodium obtained from bypass freeze-line sampling have been started. The object is to determine the specific sampling and analytical procedure that affords the highest precision and accuracy for oxide determination by this method.

5. Modified Falex Materials Wear Tester

A modified Falex wear-testing device has been prepared to study the effects of wear testing in sodium under EBR-II conditions.

Vee-block samples of Stellite 19B are being prepared and will be operated (under liquid sodium) in contact with a shaft of Carboloy 78B. Hardness measurements are being made of the material by means of the ultrasonic hardness tester.

Three of the Falex machines were mounted in position on the component and materials evaluation loop (CAMEL), which is to be operated with sodium under EBR-II conditions. This will supply a dynamic test environment which will allow wear test assessment of the materials under simulated operational conditions.

E. Fuel Development

1. Metallic Fuels

a. Irradiation of Uranium-Plutonium Alloys. A total of 20 full-length (14 in.) uranium-plutonium alloy fuel rods are undergoing irradiation in the EBR-II reactor. 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 tubing. These rods are operating at maximum jacket temperatures of 630°C and at present have reached a calculated maximum 4.7 a/o burnup. Two additional rods are fueled with U-Pu-Ti alloy and are jacketed in V-20 w/o Ti alloy tubing. The rods are being irradiated at maximum jacket temperatures of 540°C and have attained a maximum burnup of 4.8 a/o. Additional details on the design of the fuel rods and the irradiation conditions are summarized in Table III.

A postirradiation examination of 19 experimental U-Pu-Fs rods irradiated in XA01 subassembly in the EBR-II reactor is being conducted. The pins measured 0.144 in. in diameter and were jacketed in 0.009-in.-thick Nb-1 w/o Zr, Hastelloy-X, or Nb-4 w/o V alloy. Removal of the rods from the capsules confirmed the previously reported observation that the radiographs indicated the presence of collapsed tubing on three rods (see Progress Report for September 1966, ANL-7255, pp. 25-26). The collapsed sections occurred in the gas plenum, which is above the fuel pin. It is presumed that the failure occurred when the subassembly was lowered into the hot reactor sodium before the sodium in the capsules had fully melted.

Dimensional measurements of the fuel rods failed to show significant diameter or length changes. The measured densities also failed to show significant changes. Exceptions were the three rods with the collapsed tubing, which showed an average density increase of 4%.

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

Test Reactor	Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions				
			Fuel Composition (w/o)	Effective Density (%)	Cladding Composition	Cladding OD (in.)	Cladding Thickness (in.)	Power Density (kW/cc) ^a	Max Cladding Temp (°C)	Burnup to Date	
										a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ (a)
EBR-II	XG06	ND23	U-15 Pu-10 Zr	66	V-20 w/o Ti	0.209	0.016	1.7	540	4.7	11.2
EBR-II	XG05	ND24	U-15 Pu-10 Zr	66	V-20 w/o Ti	0.209	0.016	1.7	535	4.5	10.8
EBR-II	XA07	ND28	U-15 Pu-9 Zr	75	304 SS	0.208	0.021	2.0	630	4.5	12.7
EBR-II	XA07	ND41	U-15 Pu-9 Zr	75	304 SS	0.208	0.021	2.0	625	4.6	12.8
EBR-II	XA07	ND32	U-15 Pu-9 Zr	75	316 SS	0.196	0.015	2.0	605	4.4	12.2
EBR-II	XA07	ND43	U-15 Pu-9 Zr	75	Hastelloy-X	0.196	0.015	2.0	615	4.5	12.6
EBR-II	XA07	ND25	U-14 Pu-12 Zr	76	304 SS	0.208	0.021	1.9	600	4.1	10.7
EBR-II	XA07	ND27	U-14 Pu-12 Zr	76	304 SS	0.208	0.021	1.9	605	4.2	10.8
EBR-II	XA07	ND26	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	1.8	590	3.9	10.2
EBR-II	XA07	ND29	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	1.8	595	4.1	10.7
EBR-II	XA07	ND30	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	1.9	615	4.5	11.7
EBR-II	XA07	ND31	U-14 Pu-12 Zr	76	316 SS	0.196	0.015	1.9	610	4.3	11.3
EBR-II	XA07	ND33	U-14 Pu-12 Zr	76	Hastelloy-X	0.196	0.015	1.9	605	4.3	11.1
EBR-II	XA07	ND34	U-14 Pu-12 Zr	76	Hastelloy-X	0.196	0.015	1.9	610	4.4	11.4
EBR-II	XA07	ND35	U-14 Pu-12 Zr	76	Hastelloy-X	0.196	0.015	1.9	615	4.5	11.7
EBR-II	XA07	ND37	U-14 Pu-12 Zr	66	Hastelloy-X-280	0.208	0.015	1.8	610	4.5	11.2
EBR-II	XA07	ND39	U-14 Pu-12 Zr	66	Hastelloy-X-280	0.208	0.015	1.8	610	4.5	11.2
EBR-II	XA07	ND44	U-14 Pu-12 Zr	66	Hastelloy-X-280	0.208	0.015	1.8	600	4.3	10.8
EBR-II	XG05	NC17	U-15 Pu-10 Ti	63	V-20 w/o Ti	0.209	0.016	1.6	540	4.6	10.1
EBR-II	XG06	NC23	U-15 Pu-10 Ti	63	V-20 w/o Ti	0.209	0.016	1.6	540	4.8	10.7
CP-5	CP-50	1N16	Th-20 U	75	V-20 w/o Ti	0.196	0.015	1.8	590	11.5	5.4
CP-5	CP-50	4N19	Th-20 U	75	V-20 w/o Ti	0.196	0.015	1.8	560	11.5	5.4
CP-5	CP-50	2N17	Th-10 Pu-10 U	75	V-20 w/o Ti	0.196	0.015	1.9	590	13.3	6.2
CP-5	CP-50	5N20	Th-10 Pu-10 U	75	V-20 w/o Ti	0.196	0.015	1.9	590	13.3	6.2
CP-5	CP-50	3N18	Th-10 Pu-20 U	75	V-20 w/o Ti	0.196	0.015	1.8	560	7.8	5.8
CP-5	CP-50	6N21	Th-10 Pu-20 U	75	V-20 w/o Ti	0.196	0.015	1.8	560	7.8	5.8

^aBased on effective density.

The most significant variations noted were the change in fuel pin length. All pins elongated except the three with collapsed jackets. It has not yet been ascertained whether the crimping of the jacket just above the fuel produced a mechanical restraint or if the calculated pressure increase of 20 psi above the fuel (due to the collapsed plenum) contributed to this observed behavior. Data pertaining to these specimens are included in Table IV.

TABLE IV. Particulars and Irradiation Variables of XA01 Fuel Rods

Rod No.	Fuel Composition (w/o)	Jacket		Max Fuel Temp (°C)	Max Jacket Temp (°C)	Burnup (a/o)	Fuel Elongation (%)
		Composition	Condition				
C93	U-20 Pu-10 Fs	Nb-1 w/o Zr	Stress Relieved	450	435	0.57	2.8
C97	U-20 Pu-10 Fs	Nb-1 w/o Zr	Stress Relieved	445	432	0.56	2.1
C98	U-20 Pu-10 Fs	Nb-1 w/o Zr	Stress Relieved	445	432	0.55	2.7
C99	U-20 Pu-10 Fs	Nb-1 w/o Zr	Stress Relieved	445	432	0.56	2.9
C100	U-20 Pu-10 Fs	Nb-1 w/o Zr	Stress Relieved	445	432	0.56	1.8
C101	U-20 Pu-10 Fs	Nb-1 w/o Zr	Stress Relieved	450	435	0.56	2.8
CM01	U-20 Pu-10 Fz	Nb-1 w/o Zr	Stress Relieved	450	435	0.56	4.7
CA01	U-15 Pu-10 Fz	Nb-1 w/o Zr	Stress Relieved	470	445	0.57	3.6
CB02	U-15 Pu-10 Fz	Nb-1 w/o Zr	Stress Relieved	465	443	0.58	4.6
CB03	U-15 Pu-10 Fz	Nb-1 w/o Zr	Stress Relieved	460	440	0.57	3.9
CB04	U-15 Pu-10 Fz	Nb-1 w/o Zr	Stress Relieved	460	440	0.57	4.5
CD01*	U-15 Pu-10 Fz	Nb-1 w/o Zr	Annealed	455	437	0.57	-0.6
CD02*	U-15 Pu-10 Fz	Nb-1 w/o Zr	Annealed	445	432	0.56	-0.43
LA02	U-15 Pu-10 Fz	Hastelloy-X	Annealed	450	435	0.56	3.5
PA01	U-15 Pu-10 Fz	Nb-4 w/o V	Annealed	450	435	0.56	3.5
CG02	U-10 Pu-10 Fz	Nb-1 w/o Zr	Stress Relieved	465	443	0.58	4.1
CG03	U-10 Pu-10 Fz	Nb-1 w/o Zr	Stress Relieved	445	432	0.57	3.7
CJ01*	U-10 Pu-10 Fz	Nb-1 w/o Zr	Annealed	450	435	0.56	-0.4
PB02	U-10 Pu-10 Fz	Nb-4 w/o V	Annealed	455	437	0.57	4.8

*Jacket was accidentally collapsed prior to irradiation.

Six Th-U and Th-U-Pu fuel alloys, jacketed in V-20 w/o Ti tubing, are being irradiated in CP-5. The objectives of the experiment are to determine the relative swelling behavior of the fuels, the restraint characteristics of the jacket, and the maximum attainable burnup before jacket failure. These specimens were recently neutron radiographed, and it was noted that the Th-U pins had not elongated, whereas the Th-U-Pu pins had elongated an average of 8%.

2. Oxide and Carbide Fuels

a. Ceramic and Cermet Fuel Irradiations in EBR-II. The status of mixed-oxide, mixed-carbide, and stainless steel-PuO₂ cermet rods under irradiation in EBR-II is shown in Tables V, VI, and VII. The cermets were manufactured by Battelle Northwest Laboratories and prepared for irradiation by Argonne National Laboratory.

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

Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions			
		Effective Density (%)	Cladding Composition	Cladding OD (in.)	Cladding Thickness (in.)	Power Density (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	4.5	12.4
XG05	HMV-5	80	Hastelloy-X	0.297	0.015	2.2	670	4.6	12.1
XG05	NMV-11	84	Nb-1 w/o Zr	0.281	0.012	2.1	645	4.6	12.9
XO08	NMP-2	82	Nb-1 w/o Zr	0.281	0.012	1.6	545	2.7	7.3
XO08	NMV-4	80	Nb-1 w/o Zr	0.281	0.012	2.4	635	3.9	10.3
XO08	NMV-7	80	Nb-1 w/o Zr	0.281	0.012	2.1	605	3.6	9.4
XO08	NMV-12	86	Nb-1 w/o Zr	0.281	0.012	2.4	635	3.9	11.0
XO08	HMV-1	80	Hastelloy-X	0.297	0.015	2.1	640	3.5	9.2
XO08	HMV-4	80	Hastelloy-X	0.297	0.015	2.3	670	3.8	10.1
XO08	HWMP-1	82	Hastelloy-X + W	0.297	0.020	1.6	555	2.7	7.0
XO08	HWMP-1	83	Hastelloy-X + W	0.297	0.020	2.4	685	3.9	10.6
XO09	SMV-1	82	316 SS	0.306	0.024	1.6	570	2.7	7.3
XO09	SMP-1	80	316 SS	0.306	0.024	2.1	640	2.1	5.5
XO09	VMV-1	86	Vanadium	0.301	0.022	2.5	640	3.1	8.9
XO15	NMP-1	80	Nb-1 w/o Zr	0.281	0.012	1.6	535	0.23	0.61
XO15	NMV-3	80	Nb-1 w/o Zr	0.281	0.012	2.4	625	0.35	0.91
XO15	HMV-2	79	Hastelloy-X	0.297	0.015	2.3	670	0.38	0.98
XO15	TVMV-1	82	V-20 w/o Ti	0.297	0.020	2.1	620	0.30	0.82

^aBased on effective density.

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

Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions			
		Effective Density (%)	Cladding Composition	Cladding OD (in.)	Cladding Thickness (in.)	Power Density (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	2.7	5.6
XO09	SOV-6	82	304 SS	0.297	0.020	1.7	565	2.9	6.0
XO11	SOV-3	83	304 SS	0.296	0.019	1.9	610	2.7	5.6
XO11	HOV-4	80	Hastelloy-X	0.295	0.014	1.8	600	2.6	5.2
XO11	TVOV-1	77	V-20 w/o Ti	0.297	0.020	1.7	575	2.6	4.9
XO11	SOV-7	85	304 SS	0.296	0.019	2.0	630	2.6	5.5
XO11	SOV-1	80	304 SS	0.296	0.019	1.7	590	2.6	5.1
XO11	HOV-10	86	Hastelloy-X	0.295	0.014	1.9	615	2.5	5.4
XO11	HOV-15	80	Hastelloy-X	0.295	0.014	1.7	590	2.5	5.0

^aBased on effective density.

TABLE VII. Status of Type 304 Stainless Steel-clad Cermet Fuel Irradiations in Capsule XO11 in EBR-II

Specimen Number	Fuel Composition (w/o)	Design Parameters			Operating Conditions			
		Effective Density (%)	Cladding OD (in.)	Cladding Thickness (in.)	Power Density (kW/cc) ^a	Max Cladding Temp (°C)	Burnup to Date	
							a/o (U + Pu)	fiss/cc x 10 ^{-20(a)}
5P-9	SS-40PuO ₂	98	0.301	0.015	0.88	495	3.6	2.7
5P-12	SS-27PuO ₂	99	0.294	0.015	0.59	450	3.6	1.8
5U-14	SS-27UO ₂	98	0.298	0.013	0.45	435	2.8	1.4

^aBased on effective density.

Included in the list of specimens for the first time is a group of four mixed-carbide rods in experimental subassembly XO15, charged into the reactor during the month. These rods will yield performance information at linear heat ratings of 16 to 25 kW/ft and a nominal burnup level of 5 a/o. Similar rods have been undergoing irradiation in experimental subassembly XOO8, which is planned for discharge at 10 a/o burnup. Rods in subassembly XOO8 will have attained a burnup of about 8 a/o when XO15 is discharged.

Experimental subassembly XOO9 was discharged from EBR-II during the month. Three mixed-carbide and two mixed-oxide rods reached a nominal burnup of about 2.7 a/o in this subassembly. Following a suitable cooling period the rods will be shipped to the Metallurgy Alpha-Gamma Hot Cell for examination.

b. Compatibility of (U,Pu)C with Potential Cladding Materials.

(i) Compatibility Couples. Eight compatibility couples have been removed from the heat-treatment furnaces and are being prepared for metallographic examination. Included are two specimens of vanadium-20 w/o titanium heat-treated for 42 days at 700°C, the lowest temperature at which this material has been tested with (U,Pu) carbides at Argonne National Laboratory. Appreciable grain-boundary penetration of this material, when in contact with (U,Pu)C, has previously been reported at 800°C.

The remaining six are combinations of cladding materials heat-treated for 42 days in contact with pellets that contain $(U,Pu)_2C_3$ as the major phase. Chemical analysis of these pellets shows 6.75 w/o equivalent carbon. [Carbon content of $(U_{0.8}Pu_{0.2})_2C_3 = 7.03$ w/o.] The cladding materials that have been heat-treated with this material were Type 304 stainless steel (800°C testing temperature), Hastelloy-X (700°C and 800°C), Inconel 625 (700°C), 16-25-6 Cr-Ni-Mo steel (800°C), and Haynes 56 (800°C).

(ii) Specimen Preparation

(a) Uranium Sulfide Additions to Hyperstoichiometric (U,Pu) Carbides. As previously reported (see Progress Report for August 1966, ANL-7249, p. 5), a preliminary study of the effects of US additions to hyperstoichiometric (U,Pu)C was begun as a possible approach to improve compatibility with cladding materials. Although first attempts to fabricate pellets containing 15 w/o and 30 w/o US attained geometrical densities of only 80% and 77% of X-ray density, respectively, when fired to 2000°C, recrushing these materials to -44μ , repressing, and firing to 2000°C resulted in pellets approximately 90% dense. [Full density was estimated from the X-ray densities of $(U_{0.8}Pu_{0.2})C$, $(U_{0.8}Pu_{0.2})_2C_3$ and US and on the assumption of no interaction.]

(b) $(U_{0.8}Pu_{0.2})Fe_2$. Iron additions to hypostoichiometric UC are reported³ to result in improved compatibility with stainless steel through the reaction with the excess uranium metal phase to form UFe_2 . No direct compatibility test with this compound has been reported, however.

Because of the extreme brittleness of the $(UPu)Fe_2$ compound, an attempt was made to prepare pellets by powder-metallurgy techniques. Two previously arc-melted buttons (U-35.7 w/o Pu and U-51.7 w/o Fe) were melted together in an yttria-coated zirconia crucible heated to 1500°C in argon. The resulting button was crushed to $-44\ \mu$ and pressed at 3.18 metric tons/cm² with 0.5 w/o Carbowax 4000 as binder. Sintering these pellets to 1050°C resulted in an average geometrical density of 9.34 g/cm³ (71% of X-ray density); resintering to 1100°C increased the density to 9.74 g/cm³ (75% of X-ray density). A further attempt will be made to achieve greater densification of these pellets.

(c) Sodium-bonded Test Carbides. Investigations are in progress to study the contact compatibility of nuclear carbides with various jacket materials, and the irradiation behavior of carbides in gas-bonded capsules. The gas bonds limit the heat transfer, and thus it is essential that liquid-metal-bonded carbides be evaluated. Since it is desirable to know the compatibility behavior prior to irradiation testing, a series of capsule tests has been scheduled for the purpose of obtaining compatibility data for $U_{0.85}Pu_{0.15}$ -carbides in a sodium-bonded system. The capsule geometry is essentially identical to the current design of irradiation test capsule. Initial tests will include the more compatible cladding materials as determined by the contact tests. The initial tests will be conducted at 650°C and 800°C for a time interval of 500 or 1000 hr. Capsule components are being prepared and the carbide is being evaluated and characterized.

(d) Fabrication of Powdered (U, Pu)C by the Fluidized-bed Technique. The fabrication and characterization of mixed $(U, Pu)C_{1+x}$ powders produced by the fluidized-bed techniques developed by the Chemical Engineering Division are being studied. This work has as one objective the preparation of samples for compatibility studies, and a second objective of fabricating fuel samples of a rather low (85% of X-ray) density.

Two techniques were investigated to reduce the density of the sintered pellets, namely, (1) increasing the binder content, and (2) pre-firing the powder in an attempt to reduce its sinterability. The first technique has been used to reduce pellet density in sintering PuC (Ref. 4) and with mixed carbides.⁵ These sintering studies, which were made in vacuo,

³ Herbst, R. J., and Jordan, K. R., Compatibility of Modified Monocarbide Fuels with Stainless Steel, Trans. Am. Nucl. Soc., p. 17 (June 1966).

⁴ Kruger, O. L., Preparation of PuC Pellets by Vacuum-Sintering, Nucl. Applications 1(4) 1965.

⁵ Horspool, J. M., Mowat, J. A. S., and Pickles, S., "Development of Mixed Carbide Fuel Preparation and Fabrication Routes at DERE" Carbides of Nuclear Energy, Vol. II, Macmillan Co., London, 1964.

differ from the present sintering studies, which are being made in an argon atmosphere. The second technique has been used to reduce the sinterability of oxide powders in the fabrication of UO_2 fuels.

As shown in Tables VIII and IX negligible reductions in pellet density were obtained by either of these techniques, under the fabrication conditions employed (all compacts pressed at 4.23 metric tons/cm², with 0.5% Carbowax, unless denoted otherwise). The data represent an average taken from four pellets at each condition. A slight trend of decreasing pellet density with increased binder content was noted for pellets sintered at 1200°C; however, if the trend continued, a very large amount of binder would be required to attain densities of ~85% of the so-called theoretical density (T.D.). This would probably seriously increase the carbon content of the product. Compacts pressed with 2% binder, then sintered at 1650°C, were severely cracked (horizontal laminations), apparently owing to insufficient presintering time to remove this amount of binder safely; in the subsequent 1900°C run the time to heat ~700°C was increased from ~2.5 to 4 hr and the resulting pellets were free of cracks.

TABLE VIII. Effect of Variable Binder (Carbowax) Content on the Fabrication of $(\text{U,Pu})\text{C}_{1+x}$ Pellets from Mixed-carbide Powders Produced by the Hydride Cycle-Hydrocarbon Route

Binder Content (%)	Green Density (g/cc)	Temp (°C)	Weight Loss (%)	Volume Shrinkage (%)	Density	
					g/cc	% T.D. ^a
0.5	7.91	1650	0.58	36.9	12.43	91.2
1.0	7.99	1650	0.83	36.6	12.50	91.7
2.0	8.10	1650	2.04 ^b	33.3 ^b	11.89 ^b	87.3 ^b
0.5 ^c	7.89	1650	1.11	34.3	11.88	87.2
0.5	7.92	1900	0.58	36.6	12.41	91.1
1.0	8.01	1900	1.00	35.7	12.36	90.7
2.0	8.13	1900	1.80	34.9	12.27	90.0
0.5 ^c	8.06 ^d	1900	1.28	33.7	12.01	88.1 ^d

^aBased upon 13.61 g/cc, a "weighted" average of the theoretical monocarbide densities of mixed carbides containing 15% Pu.

^bNot representative data since pellets severely cracked and distorted.

^cThese single compacts prepared from exposed powder sample (~0.7% weight gain).

^dCompact repressed (resulting in higher than normal green and sintered density) after bottom chipped severely during identification marking.

TABLE IX. Effect of Powder Prefire Temperature on the Fabrication of $(U,Pu)C_{1+x}$ Pellets from Mixed-carbide Powders Produced by the Hydride Cycle-Hydrocarbon Route

Powder Prefire Temp (°C)	Green Density (g/cc)	Sinter Data				
		Temp (°C)	Weight Loss (%)	Volume Shrinkage (%)	Density	
					g/cc	% T.D.
None ^a	8.06	1650	0.77	34.4	12.19	89.6
1000	8.04	1650	0.84	35.6	12.45	91.4
1200	8.26	1650	0.66	33.7	12.39	91.0
None ^a	7.95	1900	0.88	34.9	12.10	88.9
1000	8.04	1900	0.88	34.7	12.19	89.6
1200	8.21	1900	0.73	33.8	12.31	90.4

^aBut prepared at 800°C, with CH_4-H_2 , during synthesis.

The deleterious effects of powder contamination, due to O_2 and/or H_2O vapor "pickup" by exposure, are evident, resulting in approximately twice the weight loss and ~3% to 4% reduction in density. The remaining powder used in the binder test, stored (without binder) in a vacuum desiccator for the same time (~5 weeks), was apparently "preserved" (despite many openings of the desiccator for access), since similar data (weight loss, sintered density, etc.) were obtained in comparison to previously reported fabrication results.

Though compacts of reasonable green strength can be obtained, the compacted powder is difficult to remove from the die. Unless the die cavity is swabbed out with lubricant (stearic acid) each time, excess ejection pressures are required to remove the compact (~one-half that required for compaction). A thorough coating of the powder with lubricant would probably prove beneficial.

Preheating the powders for 2 hr under vacuum (~5 μ Hg) at 1000 or 1200°C did not result in significantly lower pellet densities. The data from these tests, carried out approximately one month after the binder test, imply some deterioration in the original powders, since higher weight losses and slightly lower pellet densities were observed. Slightly higher pellet densities from the preheated powders suggest that vacuum heating may have reduced some of the deleterious effects resulting from long exposure, i.e., O_2 , H_2O vapor, and/or N_2 "pickup."

Pellets sintered at 1900°C again resulted in slightly lower densities than were obtained when sintering was carried out at 1650°C. Data obtained to date indicate that the only convenient technique to achieve

pellets of ~85% T.D. from these powders is to add nickel (0.2 w/o) at the binder-addition stage, compact at 4.2 to 5.6 metric tonnes/cm², then sinter at 1650°C; slight contamination of the product (as yet not determined) by nickel would have to be considered. Alternative potential techniques to achieve such densities may include vacuum sintering of compacts containing approximately 2% binder or prefiring the powders to higher temperatures than 1200°C.

Metallographic examination was completed on typical pellets sintered at 1550, 1650, and 1990°C, and on pellets produced from the preheated powders. Appreciable second-phase sesquicarbide was evident in all pellets. This is in apparent agreement with the 5.14% equivalent carbon content indicated by chemical analyses. A change is evident in the structure obtained at 1900°C, in contrast to those obtained at 1550 and 1650°C, i.e., the sesquicarbide diffuses to the monocarbide grain boundaries, completely encircling the monocarbide phase and sharply defining it. Structures obtained from the preheated powders were no different than those obtained from unheated powders. Specimens have been prepared for microprobe analysis, in an attempt to determine U/Pu homogeneity between the MC and M₂C₃ phases, for pellets sintered at the various temperatures.

A few plutonium analyses have been obtained. Analyses for carbon, oxygen, and nitrogen will be run as soon as scheduling permits. As shown in Table X plutonium analyses, obtained in duplicate, indicate no losses of plutonium (at the lowest or highest sintering temperature used) when sintering is carried out by the present techniques (flowing argon at slight positive pressure).

TABLE X. Typical Plutonium Analyses Occurring during the Fabrication of Hydride-derived Mixed Carbides

Original U-Pu Metal Alloy (w/o Pu)	As-prepared Mixed Carbide Powder (w/o Pu)	In Pellets Sintered at Indicated Temperatures (w/o Pu)	
		1550°C	1900°C
15.1	14.15	14.23	14.23

3. Fuel Cladding and Structure

a. Corrosion of Vanadium-base Alloys in Sodium. A continuing dynamic test (at 650°C and 6.1-m/sec velocity) in sodium containing about 10 ppm oxygen has been interrupted for specimen examination. A brief summary of the latest data is given in Table XI. All samples acquired a very thin stain or film on the surface. No dimensional changes were noted nor any trace of loss of corrosion product. It is believed that the reported weight gains fairly represent the relative resistance of the alloys to the pickup of impurities (mostly oxygen) from the sodium. Thus, V-5 w/o Cr is

more resistant by about an order of magnitude than the other alloys. V-15 w/o Ti-7.5 w/o Cr gained 80%, and unalloyed vanadium about 33% of the V-20 w/o Ti reference weight gains at equivalent exposure. Taken together, these data suggest that in sodium with a relatively low oxygen level, chromium is a beneficial addition and titanium is not. However, in other tests, in which the oxygen level of the sodium exceeded about 15 wt ppm, titanium was shown to be one of the better alloying additions.

TABLE XI. Corrosion of Vanadium Alloys in Sodium at 650°C

Alloy (Composition in w/o)	Test Time (days)	Actual Weight Gain (mg/cm ²)	Weight Gain of V-20 Ti at Equivalent Time (mg/cm ²)
V-20 Ti	173	1.04	1.04
V-15 Ti-7.5 Cr	145	0.67 0.68	0.85
V-5 Cr	57	0.02 0.05	0.69
V	41	0.21	0.63

An explanation for the apparently contradictory behavior of titanium as an alloying additive for vanadium lies in the relative affinity of the metals for oxygen. At 650°C titanium is capable of forming titanium oxide in sodium containing <1 wt ppm oxygen, whereas vanadium does not form an oxide phase until the oxygen level in the sodium exceeds about 10 wt ppm. Chromium has shown no oxide in sodium containing ~20 wt ppm oxygen.

Thus, in sodium whose oxygen concentration is less than 10 wt ppm, the greater oxygen affinity of titanium in the V-Ti alloys provides an added driving force for the pickup of oxygen. Above an oxygen level of about 10 wt ppm in the sodium, the vanadium itself reacts rather rapidly to form an oxide layer. The presence of titanium in the alloy and subsequently of titanium oxide in corrosion coating apparently improves the protective character of the rather loosely adherent oxide layer and provides a better diffusion barrier to the corrosion reaction.

In both cases, the diffusion of oxygen into the bulk of the specimen is retarded by the titanium addition, resulting in a highly oxygen-enriched zone near the surface.

Chromium has exhibited some of the same propensity for retarding oxygen diffusion in V-Cr alloys and, because of its lower chemical affinity for oxygen, it does not provide an increased driving force for oxygen pickup from reactor sodium. Other binary alloys of vanadium have been

prepared and will be exposed to sodium containing <10 wt ppm oxygen to determine if a greater improvement can be achieved with another alloying addition.

b. Effect of Sodium Environment on Creep Properties of V-20 w/o Ti. Creep tests of V-20 w/o Ti tensile specimens, which had been exposed to 650°C sodium were conducted. These specimens were made from 900°C annealed sheet, of 0.4-mm thickness, comparable to the wall thickness of fuel jackets in EBR-II. During the sodium exposure, the specimens were stressed in a loading fixture by means of a stress-relaxation method, which takes advantage of the difference in linear thermal expansion between the Type 304 stainless steel fixture and the vanadium alloy specimens in 650°C sodium. In one experiment (see Progress Report for June 1966, ANL-7230, pp. 27-29) a specimen was stressed to 1.3% strain for 7 days in sodium containing about 20 wt ppm oxygen and flowing at approximately 15 ft/sec. In another experiment, two specimens were stressed to 0.7% and 1.8% strain for 7 and 21 days, respectively, in static sodium containing less than 10 wt ppm oxygen. The control specimens were not stressed, but heated to 650°C in vacuum for 7 days. Finally, uniaxial creep tests in vacuum (4×10^{-7} to 5×10^{-6} mm Hg) at 650°C temperature and 15 kg/mm² stress were conducted on all the above-mentioned specimens.

All sodium-exposed specimens showed an increase in hardness at the surface; however, the specimen exposed to sodium of lower purity (~20 wt ppm oxygen) showed a weight loss of 1.81 mg/cm² and 20% reduction in metal cross section in 7 days, while the specimens exposed to higher purity sodium (<10 wt ppm oxygen) resulted in weight gain of 1 to 2 mg/cm² and about 1% reduction in metal cross section in 7 to 21 days. All these observations are in agreement with previous data.⁶ The weight change and microhardness data are summarized in Table XII.

The hardness profiles of these test specimens and other specimens of related experiments in Table XII are represented in Fig. 9. From the data of Fig. 9, a parabolic relationship between the depth of the hardened layer and duration of sodium exposure was plotted as shown in Fig. 10. In addition, the hardened layer was found to be deeper in lower purity sodium (~20 wt ppm oxygen) as compared to higher purity sodium (<10 wt ppm oxygen) for the same duration of exposure.

⁶ Greenberg, S., Ruther, W. E., and Levin, H. A., Corrosion of Vanadium-base Alloys in Sodium at 550°C to 750°C, to be presented at the Symposium on Alkali-metal Coolants--Corrosion Studies and System Operating Experience, Vienna, 1966.

TABLE XII. Corrosion of V-20 w/o Ti in 650°C Sodium

Test Conditions		<10 ppm				~20 ppm	
Oxygen Content in Sodium:		Static Autoclave,			Flow Loop,	Flowing Loop,	
System Used:		Building #212			Bldg. #212	Building #308	
Location:		Building #212			Bldg. #212	Building #308	
Exposure (days):		7	14	21	139	7	15
Specimen		Tensile Strip			Coupon	Tensile Strip	
Type							
No.		F	G	H	#5	R7	#51
Wt Change (mg/cm ²) (W _{final} - W _{initial})		0.096	0.131	0.171	0.87	-1.81	-17.32
Adherent Oxide Layer, μ		~2	4	6	~2	5	30
Metal Cross Section, μ		406	400	398	-	325	625 ^a
Distance from Surface (exclude loose scale)		Hardness Measurement KNOOP--15-g Load					
10 μ		511	556	624	888	567	-
15 μ		453	508	534	765	505	-
20 μ		342	433	462	722	473	-
25 μ		342	342	387	550	466	-
30 μ		-	-	342	528	410	635
35 μ		-	-	-	493	342	437
40 μ		-	-	-	441	342	378
45 μ		-	-	-	386	342	364
50 μ		342	342	342	377	342	342
55 μ		-	-	-	-	-	342
60 μ		-	-	-	-	-	342
80 μ		-	-	-	-	-	342
100 μ		342	342	342	377	342	342

^aOriginal cross section of 0.8 instead of 0.4 mm.

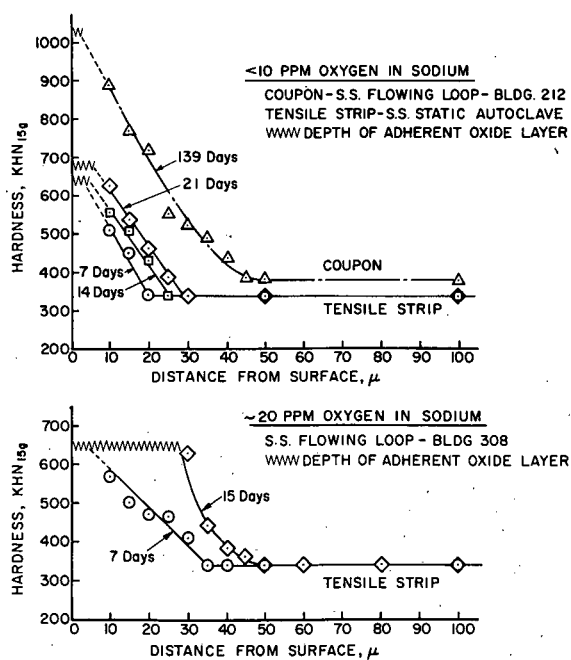


Fig. 9. Hardness Profiles of V-20 w/o Ti after Exposure to Sodium at 650°C

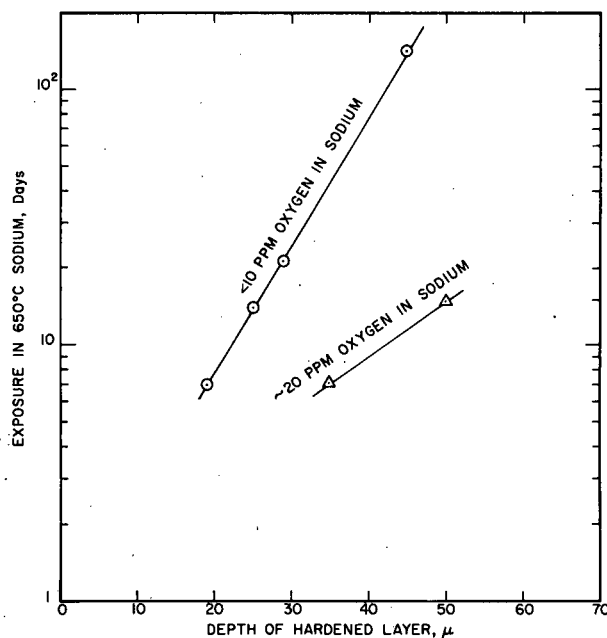


Fig. 10. Depth of Hardened Layer vs. Sodium Exposure for Material-annealed V-20 w/o Ti

The strain-time curves of Fig. 11 indicate that the hardened layer degrades the creep properties of V-20 w/o Ti at 650°C. The strain rate is higher for the specimen exposed to lower-purity sodium (~20 wt ppm oxygen) as compared to control specimens that had not been in sodium. However, consideration should be given to 20% reduction in metal cross section due to sodium corrosion. Therefore, a calculated strain-time curve to compensate for the reduction in specimen cross section is also plotted in Fig. 11. As expected, the strain rate from this curve is somewhat lower than the curve based on original cross section. Nevertheless, both rates are much higher than the rates for controlled specimens. On the other hand, there is no significant change in strain rates between specimens that had been exposed to higher-purity sodium (<10 wt ppm oxygen) for 7 or 21 days and control specimens in vacuum only. Thus it appears that the creep properties of V-20 w/o Ti sheet specimens (0.4 mm thick) will be degraded when the hardened layer reaches a finite depth (~35 μ). For specimens exposed to higher-purity sodium (<10 wt ppm oxygen) to develop such a depth of hardened layer, a duration of 42 days is estimated from Fig. 10. Additional experiments with about 50 exposure days in higher-purity sodium (<10 wt ppm oxygen) have been planned to confirm this prediction.

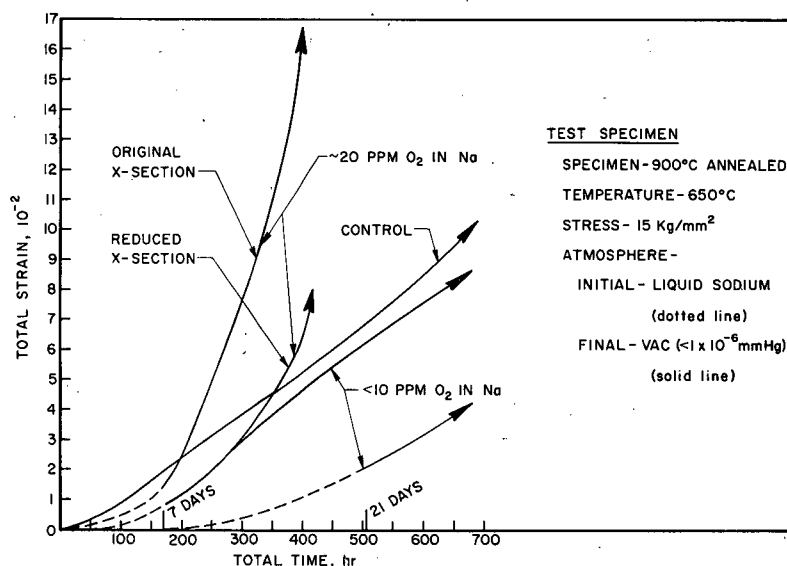


Fig. 11. Creep of V-20 w/o Ti (0.4-mm Sheet Specimen)

According to Mathews *et al.*,⁷ nitrogen and carbon in sodium are more potent strengtheners of vanadium than oxygen in sodium. In our case, carbon pickup by the V-20 w/o Ti alloy is not expected to be very important since the equipment itself is constructed with carbon getter materials--austenitic stainless steel. As for nitrogen, its presence in

⁷ Mathews, D. R., Keith, G. H., and Loria, F. A., Effect of Interstitial Impurities on the Mechanical Properties of Electrorefined Vanadium at Low Temperature, Bureau of Mines Report 6637 (1965).

lower-purity sodium (~ 20 wt ppm oxygen) has been noted by increase in nitrogen content at the surface of stainless steel loop components.⁸ Whether similar nitrogen content is present in the higher-purity sodium (< 10 wt ppm oxygen) is not known.

4. Fuel Reprocessing

a. Processes for Fast Reactor Fuels. The pyrochemical processing of fast reactor fuels of the ceramic type (e.g., oxide or carbide) clad with stainless steel is being investigated. A conceptual flowsheet currently under investigation has been previously described (see Progress Report for September 1966, ANL-7255, pp. 30-31). The process utilizes liquid metal-molten salt extractions and salt-transport separations⁹ for the separation of fissile and fertile constituents of the fuel from the fission products. Recent laboratory and engineering investigations of the current flowsheet are discussed below.

In the salt-transport separation (see ANL-7255, pp. 30-31), a molten salt (e.g., 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl) is cycled between a liquid donor alloy (e.g., Cu-33 w/o Mg) and a liquid acceptor alloy (e.g., Zn-5 w/o Mg). This procedure results in selective transfer of the plutonium from the donor alloy to the acceptor alloy, whereas uranium and the noble and refractory metal fission products remain in the donor alloy. Laboratory studies in support of this step have included a determination of the dependence of the plutonium distribution coefficient ($K_d = \text{w/o Pu in salt} / \text{w/o Pu in metal}$) between 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl and Cu-33 w/o Mg alloy on the concentration of plutonium in the donor alloy. The measurements were conducted at 600°C , the plutonium content of the donor alloy being varied from 8×10^{-3} w/o to 5 w/o. The distribution coefficient was independent of the plutonium concentration and had an average value of 0.51 ± 0.05 . These results indicate that a plutonium salt-transport step using the Cu-33 w/o Mg donor alloy should proceed satisfactorily at low plutonium concentrations and should result in high plutonium recoveries.

Other supporting laboratory work was carried out on the separation of zirconium from plutonium and uranium (see Progress Report for August 1966, ANL-7249, p. 8).¹⁰ Distribution coefficients for zirconium between molten 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl and liquid copper-magnesium alloys were redetermined at 600, 700, and 800°C , using tracer

⁸ Ratz, G. A., and Brickner, K. G. Examination of Welded AISI Type 304 Stainless-Steel Pipe Exposed to Liquid Sodium, Project No. 44.012-049(1)--Tech. Report, U. S. Steel, Applied Res. Lab., Monroeville, Pennsylvania, November 30, 1965.

⁹ Salt-transport separations are based upon the selective transfer of fissile and fertile materials from one liquid metal solution (donor) to another (acceptor) by cycling a molten salt phase which acts as a carrier between the two metal solutions. Noble and refractory metal fission products remain in the donor alloy.

¹⁰ In the salt-transport step, zirconium remains in the donor alloy with the uranium.

to increase the sensitivity of the analyses. The magnesium concentrations were as follows: 35.4 w/o at 600°C; 34.5 w/o at 700°C; 6.6 w/o at 800°C. As previously shown, the zirconium distributed strongly in favor of the metal phase. Despite the use of irradiated zirconium, the zirconium concentrations in the ternary salt were near the limit of detection. The zirconium distribution coefficient ($K_d = \text{w/o zirconium in salt} / \text{w/o zirconium in metal}$) at the three temperatures was of the order of 10^{-6} , which may be compared to the value of about 5×10^{-4} obtained in the earlier experiments with unirradiated zirconium. The results of this work indicate that zirconium is not likely to transport with plutonium.

In the distribution experiments, data were also obtained for the solubility of zirconium in the liquid copper-magnesium alloys. The zirconium solubilities in a Cu-~33 w/o Mg alloy were as follows: 0.26 w/o at 600°C; 1.1 w/o at 700°C; 4.6 w/o at 800°C. These values indicate that the solubility of zirconium is large enough in the donor alloy to permit its separation from the uranium by decantation of the donor alloy and washing the uranium with liquid magnesium.

In a further study of the salt-transport step, distribution coefficients were determined at 600 and 700°C for neptunium between liquid Cu-33 w/o Mg alloy and each of the following molten MgCl_2 -based chloride salts: 16 m/o MgCl_2 -42 m/o NaCl-42 m/o CaCl_2 , 50 m/o MgCl_2 -25 m/o NaCl-25 m/o CaCl_2 , and 50 m/o MgCl_2 -30 m/o NaCl-20 m/o KCl. The neptunium distribution coefficients ($K_d = \text{w/o neptunium in salt} / \text{w/o neptunium in metal}$) at 600 and 700°C were as follows:

Salt Phase (m/o)	K_d	
	600°C	700°C
16 MgCl_2 -42 NaCl-42 CaCl_2	0.20	0.12
50 MgCl_2 -25 NaCl-25 CaCl_2	0.80	0.67
50 MgCl_2 -30 NaCl-20 KCl	0.53	0.47

These K_d values are quite similar to those for plutonium and suggest that neptunium will follow plutonium in the present reference flowsheet. It should also be noted that the neptunium K_d is temperature-dependent in the systems studied.

Liquid zinc-magnesium alloys are being considered for use as plutonium acceptor alloys in the salt-transport separation step. Plutonium distribution coefficients between molten 50 m/o MgCl_2 -30 m/o NaCl-20 m/o KCl and zinc-magnesium acceptor alloys and plutonium solubility data at 600, 700, and 800°C were presented previously (see Progress Report for October 1966, ANL-7267, p. 45). A thermodynamic analysis of these data indicates that two plutonium-zinc intermetallic phases exist in the plutonium-zinc-magnesium system (up to about 40 a/o Mg). One phase is PuZn_{12} ,

which is the solid phase in equilibrium with liquid plutonium-zinc solutions below about 710°C and liquid plutonium-zinc-magnesium solutions containing up to 7 w/o Mg at 600°C and 2.4 w/o Mg at 700°C. The other equilibrium solid phase is $\text{Pu}_2\text{Zn}_{17}$ in the binary plutonium-zinc alloys above about 710°C and in the ternary plutonium-zinc-magnesium alloys above the following magnesium concentrations: 7 w/o Mg at 600°C; 2.4 w/o Mg at 700°C; 0 w/o Mg at 800°C. These thermodynamic findings apply up to about 20 w/o Mg, the limit of the available solubility data.

A supporting engineering effort to investigate containment materials for process solutions has continued. In recent corrosion work, specimens of niobium in the form of agitator blades were exposed to a metal-salt system consisting of Cu-33 w/o Mg-1 w/o U and 50 m/o MgCl_2 -30 m/o NaCl-20 m/o KCl at 750°C for 156 hr. The corrosion resistance of the specimens was very good (penetrations of <0.01 mm/156 hr). Another encouraging aspect of this test was the retention of ductility of the niobium specimens after exposure; these specimens could still be bent nearly 180° without cracking. Additional testing of niobium is planned. Material for the fabrication of niobium crucibles has been ordered.

b. Plutonium Salt-transport Separations. Installation of equipment to conduct engineering feasibility experiments of the plutonium salt separations step (see Progress Report for August 1966, ANL-7249, p. 10) was completed. Equipment shakedown runs without plutonium are underway.

F. General Research and Development

1. Parameter Study of Fast Reactor Cores

Blanketed and reflected single-zone cores are being studied as a basis for future work with multizoned cores. Physics calculations are being made primarily via a one-dimensional diffusion code (MACH I). Initially, equivalent bare-core modeling was attempted, but appeared to be unsatisfactory at length-to-diameter (L/D) ratios of 0.2. As a result, self-consistent one-dimensional analysis was applied, and appears to be workable at least down to $L/D = 0.2$. This technique is now being used routinely to generate the physics parameters for the single-zone cores under study. As one indication of its workability, breeding ratios evaluated from it by three different techniques agree to within 2%.

A summary report of the initial portion of the parametric study concerned with the single-zone core is being prepared. This will include a comprehensive description of the physics-analysis methods and of the basis for technical design and resulting core compositions.

2. Steam-generator and Turbine-Generator Islands for LMFBR Power Plants

This study represents a second joint effort with United Engineers and Constructors, Inc. (UEC) to establish design trends for large, sodium-cooled, fast-breeder-reactor-powered central stations. In addition to updating the results of the first study,^{11,12} three more steam conditions were evaluated with respect to net thermal efficiency and capital costs of the turbine-generator island. Also, for two steam conditions, costs of the steam-generator island were estimated.

For each set of steam conditions, engineering flow sheets and equipment layouts were developed. Design of the steam generator was based on the Babcock and Wilcox concept which is being developed under AEC funds. Cost estimates were prepared in accordance with the Federal Power Commission Uniform Code of Accounts. They included all direct and indirect costs related to materials, labor, and equipment. Excluded from the estimates were land, interest during construction, and certain of the purchasers' indirect costs.

a. Turbine-Generator Island. The turbine-generator (T-G) island costs include all equipment from the inlet steam line at the T-G building wall to the high-voltage terminals of the main transformer. Table XIII lists the comparative costs and thermal performance for all the nominal 1000-MW(e) cases with a single tandem compound-turbine arrangement. The net station efficiencies have been adjusted slightly to provide uniformity among the cases considered in the first and second studies.

TABLE XIII. Influence of Steam Conditions on Thermal Efficiency and Capital Costs of the Turbine-Generator Island for 1000-MW(e) Nuclear-powered Central Stations

Case	Steam Condition: Pressure--Initial/Reheat Temp (psig--°F/F)	Net Thermal Efficiency (%)	Capital Cost (\$/Net kW)
A	3500--1000/1000	43.5	44.40
B	2400--1000/1000	42.6	43.15
C	1800--900/900	40.4	47.36
D	1000--Dry and Saturated	33.9	56.99
E	1450--1000	38.5	48.37
F	2400--950/950	41.8	45.48
G	2400--950	39.5	50.31

¹¹First study summarized in ANL-6977, Reactor Development Program Progress Report--November 1964, p. 17.

¹²UEC Report of the first study--Engineering Study for Large Steam-Electric Generating Units Applied to Argonne Fast Breeder Reactor Studies, TID-23257 (Nov 1964).

The results for the 1000-MW(e) cases lead to the following observations:

1. Excluding Case D (1000 psig dry and saturated) of Table XIII, reasonable steam cycles for the LMFBR power plant result in net thermal efficiencies of 38 to 44% and in turbine-generator island costs of \$43 to 50 per net kW.

2. Based on an average energy cost of $27\text{¢}/10^6$ Btu for the steam, an annual capacity factor of 80%, and annual capital charges of 13%, the four steam cycles with sodium reheat (Cases A, B, C, and F) yield costs within ~ 0.2 mill/kW-hr, whereas Cases D and G with no sodium reheat are about 0.3 mill/kW-hr more expensive. Although the assumed average cost of steam energy is a gross simplification, this approach does provide some perspective on design trends for T-G islands.

3. It would appear that the T-G island associated with sodium-cooled fast reactors could exhibit a capital cost advantage of about \$10/kW(e) over islands associated with water-cooled thermal reactors.

- b. Combined Steam Generator and Turbine Generator. Ideally, the total power plant, including the fuel cycle, should be investigated in order to specify the design trends. A step in this direction is to couple the steam-generator and turbine-generator islands. For this purpose, two of the seven steam conditions investigated for the turbine-generator island were selected for estimating costs of the steam-generator island. Engineering estimates were made of the steam-generator island costs for 2400 psig--950/950°F steam with sodium reheat, and 2400 psig--950°F with no sodium reheater. In both cases, the overall battery limits were the high-temperature line for inlet of sodium to the steam generator and the secondary sodium-pump-discharge line at one interface, and the high-voltage transformer output terminals at the other.

The results are listed in Table XIV. Before making any comparisons, a few comments on the 2400 psig--950°F cycle are appropriate. This cycle, as described in the UEC study, has a very high moisture content in the low-pressure cylinder of the turbine. Discussions with turbine manufacturers in the initial phases of the study revealed there was no readily available information on this cycle nor on commercial means of alleviating the moisture content. Basically, neglecting new materials or turbines designed for high moisture, external moisture separation or live-steam reheat are possibilities for lowering moisture content. However, each of these solutions has an effect on the heat rate and capital cost.

TABLE XIV. Estimated Costs of Combined Steam-generator and Turbine-Generator Islands with and without Sodium Reheat

Case	Steam Condition (psig--°F/°F)	Net MW(e)	Cost [\$/kW(e)]		
			S-G	T-G	Combined
F	2400--950/950	1029	36.66	45.48	82.14
G	2400--950	1001	29.69	50.31	80.00

The results of the combined study lead to the following observations:

1. Ignoring the influence of obtaining an allowable moisture level in Case G, and assuming an 80% annual plant capacity factor, 13% charges on capital, with an incremental energy cost of 15¢/10⁶ Btu, one finds a 0.03-mill/kW-hr differential which favors the sodium-reheat cycle.

2. Adjustments for obtaining an allowable moisture content would increase the differential, probably by a slight amount. Plant simplification, however, could increase the reliability of the nonsodium-reheat cycle.

These statements should be tempered by the fact that only two steam conditions were considered along with a particular type of sodium-heated steam generator. A more comprehensive study of alternative engineering and equipment designs could modify the foregoing observations. Finally, it is interesting to note that linking the turbine-generator and steam-generator islands tends to foreshorten the extremes indicated by studies confined to the T-G island. An even greater impact on apparent design trends can be expected upon inclusion of reactor island and fuel-cycle details.

3. 1000-MWe Study

The Chicago Operations Office of the AEC is reviewing the Atomic International contract for correct contract languages, especially language pertaining to patent clauses.

The AEC, Division of Reactor Development and Technology, has reviewed and approved the revised Follow-on Study Proposal submitted by Combustion Engineering. Contract negotiations between CE and ANL is progressing.

A meeting was held at Westinghouse (WAPD) in Pittsburgh on November 9, 1966 to discuss patent rights, auditing rights, and contract content. As a result, Westinghouse suggested revisions to Appendix C of the Request for Proposal. These revisions are being reviewed by ANL.

The five contractors have submitted additional comments on the ground rules and these comments are being reviewed by ANL.

¹³A cascode amplifier fundamentally consists of two triodes connected in series.

The modification shown in Fig. 12 gives a considerably faster response for equal low-frequency gain. The cathode of T_2 is maintained by feedback from T_3 at a very low impedance, and the signal line from T_1 can be terminated at this point with its characteristic impedance in series. Only the input triode need be directly attached to the counter; signals can be delivered to the remainder of the preamplifier without sacrificing rise-time or noise levels. The gain of the circuit in Fig. 12 is the product of g_m of T_1 by the resistance coupling T_2 to T_3 ($2000\ \Omega$ in the diagram). The value of this resistance can be changed to provide any desired value of gain without influencing the operation of the circuit. The choice of T_1 in the figure was an 8254 tube (EC/1000) which has a fair value of g_m (about $14\ \text{mA/V}$) and a low input capacitance (about $5\ \text{pF}$). Any tube not drawing more than $25\ \text{mA}$ can be substituted, depending upon the application. It is difficult at present to find a better tube than the 8254 for overall signal-to-noise optimization for use with low-capacity detectors.

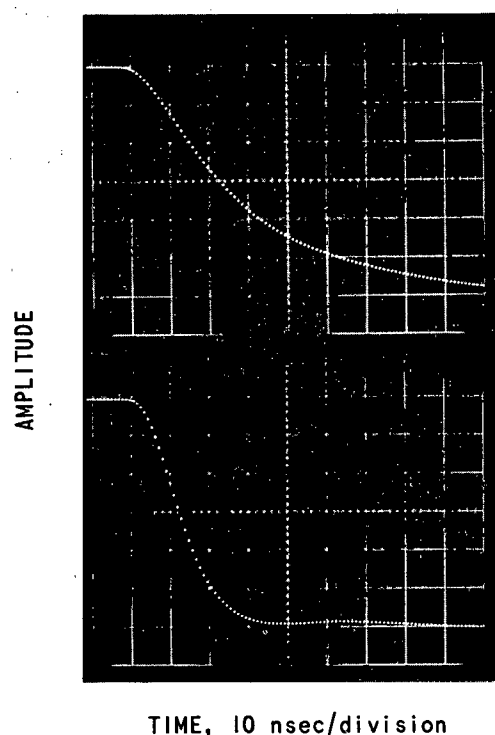


Fig. 13. Comparison of Risetime of a Cascode Preamplifier. (a) Without feedback and (b) with feedback.

2. Relationship of Cross-section Errors to Integral Data

Recent attempts¹⁴ to resolve cross-section errors by use of fast reactor integral data have been marked by a lack of consistency. This arises because there are many correlations and accidental cancellations which exist among the effects of differing errors in microscopic cross sections. This is known and expected in general reactor theory such as has been applied to thermal reactors, and the result has been to relate cross-section errors to such lumped parameters as ϵ , p , η , f , τ , L^2 , or to the corresponding observable

¹⁴Rakavy, G., Use of Integral Measurements as Supplementary Data for Determining Neutron Cross Sections, Proc. Conf. Neutron Cross Section Technology, March 22-24, 1966, CONF-660303.

quantities. In fast reactor physics one attempt to take into account correlated effects has been to develop a mechanical approach based on second-order perturbation theory.¹⁵

An approach with more physical content is to use the lumped-parameter model of the fast reactor spectrum, such as has been most recently suggested by Stacey.¹⁶ In this approach, errors between observed and calculated integral data are related to errors in lumped constants. A collateral effort is to find the effect of microscopic cross-section errors on the lumped constants. This technique has worked well in general reactor physics,¹⁷ and it is reasonable to expect such an approach to be valuable here.

Using Stacey's formalism it is possible to show, for example, that if inelastic-scattering effects are neglected, the error in the reaction rate R , given by ΔR , is related to the lumped constant S (representing the elastic-scattering process) by

$$\frac{\Delta S}{S} = \frac{S(1 - S^2)\Delta R}{(1 - S)^2(R - F)}, \quad (1)$$

where

S = calculated value of S ;

ΔS = error in S which yields the discrepancy between the calculated value of R and the measured value of R ;

F = response of the detector to a pure fission spectrum.

3. Evaluation of Cross Sections

The compilation of cross sections for gadolinium for the ENDF/B library has been completed. The main source of data was KFK-352.¹⁸ Various additions and changes in the data from the primary source were made, as follows:

¹⁵Ceschini, G., Gandini, A., Dal Bono, I., and Faleschini, B., Comparison between Experimental and Theoretical Integral Data on Fast Critical Facilities. CALI, A Program for Generating Effective Nuclear Group Constants by a Perturbation Method, Proc. Int. Conf. on Fast Critical Experiments and Their Analysis, Oct. 10-13, 1966, ANL-7320, to be published.

¹⁶Stacey, W. M., Jr., Spectrum and Importance Function in Spatially Independent Fast Reactor Assemblies, Trans. ANS 9, 474-475 (1966).

¹⁷See, for example, the review article

Price, G., and Hellens, R., Lattice Studies in Light Water Moderated Systems, Geneva Conf., P/Z64 3, 3-10 (1964).

¹⁸Schmidt, J. J., and Siep, I., 26 Gruppen-Wirkungsquerschnitte für Europium, Samarium, Gadolinium und Hafnium, KFK-352 (Aug 1965).

Resonance parameters for the first five resonances are from Möller.¹⁹ Parameters for 24 other resonances are from BNL-325.²⁰ The cross sections are calculated entirely from resonance parameters up to 50 eV.

From 50 eV to 10 keV, the total cross section was read from BNL-325 graphs. It was split into capture and scattering components using Eqs. 39, 40, 43, and 44 of Ref. 18.

Cross sections for (n,2n) and (n,3n) reactions, calculated according to the methods of Pearlstein,²¹ were obtained from Pearlstein.

Cross sections were extended from 10 to 15 MeV. The (n, γ) cross section was extrapolated linearly on a log-log scale. The total cross section was extended using BNL-325 points as a guide, while the reaction cross section was extrapolated to a value at 15 MeV equal to 1.01 times the sum of (n,2n), (n,3n), and (n, γ) cross sections.

The inelastic scattering cross section was found by subtracting other cross sections from the reaction cross section.

Legendre coefficients for elastic scattering obtained from H. Alter of Atomics International²² were used from 0.3 to 1.5 MeV. Below 0.3 MeV, coefficients were found by extrapolation. Above 1.5 MeV, the coefficients are based on Abacus-2 optical-model problems using roughly the same parameters as in Ref. 18.

Values of $\bar{\mu}_L$, ξ , and γ were calculated from the Legendre expansion coefficients.

Parameters for the free-gas thermal scattering law were included.

4. Multilevel Cross Sections for the Fissionable Isotopes

A study of cross sections of the fissionable isotopes is being made by multilevel analysis. In particular, the Reich-Moore²³ multilevel formulas, which assume one neutron and two fission channels, are being explored. Doppler broadening of the multilevel cross sections will be done

¹⁹Möller, H. B., et al., Low Energy Neutron Resonances in Erbium and Gadolinium, Nucl. Sci. Eng. 8, 183-192 (Sept 1960).

²⁰Hughes, D. J., and Schwartz, R. B., Neutron Cross Sections, BNL-325, Second Edition (July 1, 1958).

²¹Pearlstein, S., Analysis of (n,2n) Cross Sections for Medium and Heavy Mass Nuclei, Nucl. Sci. Eng. 23, 238-250 (Nov 1965).

²²Davis, S. K., et al., Compilation, Evaluation and Reduction of Neutron Differential Scattering Data, NAA-SR-11980, Vol. IV (to be published).

²³Reich, C. W., and Moore, M. S., Multilevel Formula for the Fission Process, Phys. Rev. 111, 929-933 (1958).

numerically. The various resonance parameters will be selected by Monte Carlo techniques as suggested by Moore and Simpson.²⁴ In addition the "Lynn doublet"²⁵ effect, which results from two or more closely spaced resonance levels interfering with one another to form a single resonance peak, will also be investigated.

5. The ARC System

a. Burnup and Depletion. The Phase 0 (see Progress Report for August 1966, ANL-7249, pp. 40-41) package is complete, and test runs are being made on enrichment search procedures and recycle calculations necessary to solve equilibrium problems. Although search procedures have been greatly improved, work continues on this portion of the package.

Some successful comparisons have been made with the CYCLE 2 depletion code. Recycle problems will be compared with CYCLE 1 equilibrium calculations in the near future. (CYCLE 1 and CYCLE 2 are unpublished ANL burnup codes.)

The ARC modules for the first production version have been fixed. Upper and lower interface quantities, i.e., the data required and the data produced by each of the modules, have been defined.

b. Expanded Data Handling. The dimension limitations in MC² have been increased to allow more pointwise data to be handled. The code had previously accommodated 2000 data points for the elastic-scattering cross section and 500 data points for all other pointwise data, namely, $\bar{\mu}$, σ_{inel} , σ_f , σ_c , $\sigma_{n,2n}$, $\sigma_{n\alpha}$, σ_{np} , discrete level excitation for σ_{inel} , and nuclear temperatures for σ_{inel} and $\sigma_{n,2n}$. The code now accepts 1400 data points for the inelastic discrete level excitation and nuclear temperature data and 5500 points for all other pointwise data. This change was prompted by the needs of the current ENDF/B cross-section-evaluation effort being coordinated by Brookhaven National Laboratory.

c. Computation of Effective Cross Sections. A computer code, RABBLE, for the computation of effective cross sections and fluxes in slab or circularized lattice cells for the resonance-energy region has been written and tested. The theory upon which the code is based has been described previously (see Progress Report for July 1966, ANL-7245, p. 29). The theory allows for space- and energy-dependent slowing-down sources and is therefore well-suited for application to problems in which resonance overlapping effects are significant.

²⁴Moore, M. S., and Simpson, O. D., Measurement and Analysis of Cross Sections of Fissile Nuclides, Conf. on Neutron Cross Section Technology, March 1966, Washington, D. C., pp. 840-872.

²⁵Lynn, J. E., Analysis and Interpretation of the Slow Neutron Cross Sections of the Fissionable Nuclei, Phys. Rev. Letters 13, 412-415 (1964).

The code allows up to four mixtures or compositions, each of which can have resonant materials and can have different temperatures. The code computes effective cell-averaged fluxes, and macroscopic cross sections and effective composition-averaged fluxes and microscopic cross sections for up to 400 groups. The code allows four resonant materials, each having up to 200 resolved resonances. The groups for which the fluxes and cross sections are edited can have the same width in lethargy as the ultrafine groups in the more versatile MC² cross-section code. Thus RABBLE can be incorporated in MC² to provide precise computation of resonance absorption.

d. Fast Reactor Accident Analysis. An important component of any computer program for analysis of fast reactor behavior during accident conditions is that which computes heat transfer during the incident. Correct treatment of heat transfer is essential if many of the feedback responses are to be correctly computed. This work is a part of the programming effort in fast reactor accident analysis that will become a part of the ARC system (see Progress Report for May 1966, ANL-7219, pp. 32-35). The heat-transfer treatment has been separated into an area of steady state, for initialization, and of the transient state.

(i) Steady State: Heat Transfer in the Fuel Cell. The equations of steady-state heat transfer in the fuel cell have been programmed in Fortran IV for cylindrical geometry. The variation of physical properties and dimensions with temperature is taken into account. A typical fuel cell consists of the fuel pin surrounded by a fluid bond, metallic cladding, and coolant.

The differencing scheme (to approximate the space derivative) used to solve the steady-state heat-conduction equation is basically a central differencing scheme which evolves from the application of heat balance to each spatial increment. Care is taken to avoid having an overdetermined system of equations. The scheme is capable of handling wide variations of the physical properties, such as the variation of thermal conductivity with temperature.

A unique feature of the differencing scheme is that it permits nonuniform space increments. This is intended primarily to account for the (nonuniform) thermal expansions of the space increments and the expansions due to phase changes.

The thermal expansion of the fuel is considered in an "open loop" sense. When a steady distribution of temperature is obtained, each space increment is allowed an unrestricted thermal expansion. However, when the bond consists of a gas such as helium, the temperature drop through the bond is large, and the drop is a strong function of the thickness of the bond. Therefore, if thermal expansion is considered, an iteration of

the computation of the steady state is necessary to obtain compatibility between the bond thickness and the temperature drop through the bond.

Since the temperature of the liquid metal coolant is normally far from its boiling point in the initial state, the energy-balance equation for the coolant is solved easily to obtain the temperature rise in the coolant in any axial segment.

Axial conduction of heat is neglected in the fuel. Therefore, temperatures are coupled axially only in one direction, namely, the direction of the coolant flow. A straightforward matching procedure (starting from the coolant inlet) is adopted to obtain the two-dimensional temperature profile in the fuel cell. When such profiles are obtained, the necessary regional average temperature, volume fractions, and physical dimensions are supplied to the neutronics codes. Changes in dimensions and density as well as changes in cross sections related to fuel temperature through the Doppler effect are important feedback responses.

(ii) Transient State. Coding in Fortran IV of the transient state is in progress. The numerical method adopted for solving the heat-conduction equation is essentially a two-level (in time) implicit scheme. The method does not suffer from any numerical instability and does not require iterations to handle the nonlinearities of the heat-conduction equation. It is believed that the present method is an improvement over the currently used explicit schemes in that the method has no inherent numerical instability, and that it may result in saving computing time in cases where large time increments are used to compute slow transients.

The capabilities of the coding are:

1. It handles the variation of the physical properties with temperature.
2. The thermal expansions and phase changes are taken into account. This is facilitated by using nonuniform space meshes.
3. It evaluates the pressure buildup in the fuel rod when the expansion is constrained by a structural component.

Currently, only the solid and the liquid states of the fuel are considered.

B. Fuels and Cladding

1. Chemistry of Fuel Materials

a. Vaporization of Uranium Monophosphide. Investigations are being made of the high-temperature properties of uranium monophosphide, a potentially useful component of nuclear fuels, because the existing

information (including the U-UP phase diagram) is uncertain as well as incomplete. In this study the basic quantities being determined are the temperature dependences of the partial pressures of vapor species in equilibrium with uranium monophosphide. The experimental technique involves vaporization of uranium monophosphide within an effusion cell, and analysis of the effusing species by means of a Bendix time-of-flight mass spectrometer. Tungsten was chosen as the container material when it was found (by X-ray diffraction analysis of residues) to be inert to uranium monophosphide up to the maximum temperature (2150°C) of the present experiments. Ionizing electron energies of 15 eV were used in order to avoid fragmentation effects which were observed at higher energies. The compositions of uranium phosphide samples were determined by chemical analysis.²⁶

Between 1800 and 2150°C, the primary species vaporizing from uranium monophosphide were found to be U(g), P(g), and P₂(g), as previously reported.²⁷ Also in agreement with earlier reports, the hypothetical species UP(g) was not observed. Although P₄(g) was observed, its presence was attributed to the vaporization, at relatively low temperatures, of phosphorus which had condensed on the heat shields above the effusion cell. A correction was applied for the effect of this secondary vaporization on the observed partial pressure of P₂(g). Minor amounts of UO(g) and UO₂(g), formed because of slight oxygen contamination of the UP, were also detected, during the initial heating. The intensities of these species decreased steadily, generally becoming less than 1/30 that of U(g) after one hour at temperature.

The preliminary mass-spectrometric experiments give evidence that stoichiometric UP changes composition during vaporization. Continuous effusion from a starting composition UP_{1.00±0.03} at 2010°C yielded a much higher concentration of P plus P₂ than of U in the vapor, thereby indicating that the monophosphide phase was becoming hypostoichiometric with respect to phosphorus (i.e., UP_{1-x}). Confirmation of the changing vaporization behavior and composition of the residual solid was also obtained by an effusion experiment in which over 50% of a sample of initially stoichiometric UP was vaporized at ~2120°C. The rate of loss in weight of the sample was observed to increase steadily over successive time periods. Moreover, metallographic analysis of the quenched residue from this experiment revealed the presence of uranium as a minor phase located at monophosphide grain boundaries. From appearance alone it could not be decided whether the uranium had existed as a second phase at the temperature of the experiment, or, alternatively, had precipitated from hypostoichiometric uranium monophosphide on cooling. X-ray diffraction analysis of the residue showed that the lattice parameter of the UP was unchanged from its original value of $a_0 = 5.589 \text{ \AA}$.

²⁶Milner, G. W. C., Rowe, D. H., and Phillips, G., The Analysis of Uranium Phosphides, AERE-R-4906 (April 1965).

²⁷Gingerich, K. A., and Lee, P. K., J. Chem. Phys. **40**, 3520 (1964).

Baskin and Dusek,²⁸ on the basis of lattice parameters and metallographic analyses, reported that uranium monophosphide formed a hypostoichiometric phase which appeared to vaporize congruently at about 2000°C. Gingerich and Lee²⁷ concluded that uranium monophosphide vaporizes congruently between 1477 and 1977°C; however, from extrapolation of their data they inferred that incongruent vaporization would occur at temperatures above 2127°C. Our results, thus far, give no indication of congruent vaporization of monophasic uranium monophosphide. Instead, they suggest that UP, on vaporization, becomes progressively hypostoichiometric until a two-phase mixture, UP_{1-x} (s, saturated with U)²⁹ + U (ℓ , saturated with UP_{1-x}), forms.

Preliminary equations for the partial pressures of U, P, and P_2 over hypostoichiometric UP (possibly the two-phase mixtures) were derived by correlating the data from two types of effusion experiments, mass-spectrometric and weight-loss.

The temperature dependences (from 1800 to 2150°C) of the U^+ , P^+ , and P_2^+ ion currents were obtained in three series of measurements for which the starting materials were: (1) 0.1 g of powdered $UP_{1.00 \pm 0.02}$, preheated for 5 hr at 2000°C; (2) 75 mg of uranium plus 300 mg of a sintered UP disk, preheated 6.7 hr at ~1972°C; and (3) the residue from series (2) plus 36 mg of uranium, preheated 2 hr at ~2040°C. Because the ion currents were found to be reversible with temperature, it may be concluded that either a monophasic material of constant composition was present, or that equilibrium between $U(\ell)$ and UP_{1-x} was always maintained. Furthermore, because of the agreement of the initial ion-current curves for all three series, it is believed that all three came to the same phase composition(s) by vaporization during the preheat periods. The residues from these runs have not been analyzed; however, the residue from a similar run, which gave similar ion-current curves, was found to have a P/U ratio of 0.95 ± 0.02 . The ion-current data ($\log I_i T$ versus $1/T$) for each species were least squared and the three series were combined by normalizing (for different instrumental conditions) at 1961°C, the approximate midrange temperature.

The partial pressure of an effusing species, p_i , is related to the ion current I_i by the equation

$$I_i T = C \sigma_i \gamma_i p_i, \quad (1)$$

where T is the absolute temperature, C is a constant characteristic of the electronics and geometry of the mass spectrometer, σ_i is the ionization

²⁸Baskin, Y., and Dusek, J. T., Annual Progress Report for 1964, Metallurgy Division, ANL-7000 (1964), p. 87.

²⁹Bowman, M., Los Alamos Scientific Laboratory, reports that the hypostoichiometric range may extend down to $UP_{0.96}$ at 1900°C (private communication).

cross section, and γ_i is the multiplier efficiency. Otvos-Stevenson cross sections³⁰ were used in the calculations ($\sigma_U = 55.7$; $\sigma_P = 13.8$; $\sigma_{P_2} = 27.6$). Multiplier efficiencies were estimated on the assumption³¹ that ions of equal velocity have equal multiplier efficiencies, i.e., $\gamma_1/\gamma_2 = (M_2/M_1)^{1/2}$, where M is molecular weight. The constant C may be determined by combining the results of experiments in which total mass loss is measured with results of experiments in which the ratios of species in the vapor are measured.

Total weight loss during effusion at about 2000°C was measured for samples comparable to those used in the temperature-dependence experiments. After normalizing the weight losses to the same effusion conditions (time, temperature, and orifice diameter), they were found to agree within 10%. The actual weight loss was then used to calculate the constant C in Eq. 1.

The combined weight-loss and mass-spectrometric measurements led to the tentative partial-pressure equations (from 2073 to 2423°K):

$$\log p_U (\text{atm}) = 5.8905 - (26,268/T); \quad (2)$$

$$\log p_P (\text{atm}) = 8.2842 - (31,494/T); \quad (3)$$

$$\log p_{P_2} (\text{atm}) = 10.1451 - (37,351/T). \quad (4)$$

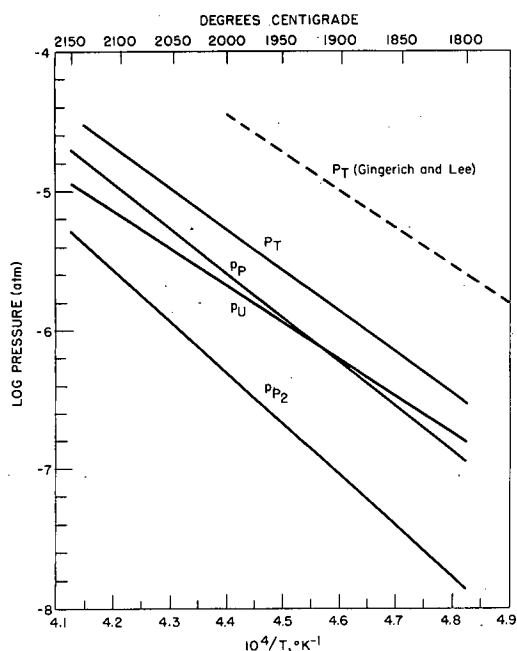


Fig. 14. Vapor Pressures over Hypostoichiometric Uranium Monophosphide vs. Reciprocal Temperature. (2073 to 2423°K)

The combined uncertainties (95% confidence level) of these pressures are $\pm 4.5\%$, $\pm 7.1\%$, and $\pm 10.8\%$, respectively. The corresponding partial molar enthalpies of vaporization are (in kcal/mole) $\Delta H(U) = 120.2 \pm 0.3$, $\Delta H(P) = 144.1 \pm 1.5$, and $\Delta H(P_2) = 170.9 \pm 4.2$. Figure 14 shows a plot of these equations and includes, for comparison, the total pressures P_T from the mass-spectrometric effusion experiments of Gingerich and Lee, and from summation of the partial pressures reported above. The disagreement with Gingerich and Lee was not unexpected inasmuch as they had reported $P(g)/P_2(g)$ ratios which were clearly nonequilibrium values.

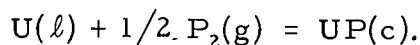
The internal consistency of the present results was found to be good. The second-law Gibbs free energy, enthalpy, and entropy of dissociation of $P_2(g)$, calculated from Eqs. 3 and 4 at

³⁰Otvos, J. W., and Stevenson, D. P., J. Am. Chem. Soc. **78**, 546 (1956).

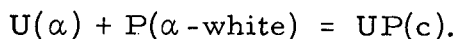
³¹Ploch, W., and Walcher, W., Rev. Sci. Instr. **22**, 1028 (1951).

2234°K, the approximate midrange temperature of the experiments, were 51.6 kcal/mole, 117.3 kcal/mole, and 29.4 eu, respectively. These values are in good agreement with corresponding values, 55.1 kcal/mole, 119.2 kcal/mole, and 28.7 eu, calculated from spectroscopic data.³²

A preliminary value for the enthalpy of formation of uranium monophosphide [$\Delta H_f^\circ(\text{UP}, c)$], which to date is unreported, was estimated from the present results. From Eqs. 2 and 3 and enthalpy-of-formation data³² for $\text{U}(g)$ and $\text{P}(g)$, the value $\Delta H_f^\circ_{2234}(\text{UP}, c) = -99 \pm 4$ kcal/mole was derived for the reaction



By extrapolation of the recently reported mean heat capacity for uranium monophosphide, 13.8 cal/deg/mole³³ for the range 800 to 1200°C, the value $\Delta H_f^\circ_{298}(\text{UP}, c) = -77 \pm 5$ kcal/mole was obtained for the reaction



2. Fabrication and Evaluation

a. Studies of Corrosion in Lithium at Elevated Temperatures.

The initial examination of a tantalum surface that was tested in silicon-containing-lithium at 1200°C (see Progress Report for September 1966, ANL-7255, p. 45) showed a deposition of silicon and a hardened surface layer. The composition and thickness of this layer have been determined. Evidence from X-ray diffraction analysis indicates that the composition of the layer closely corresponds to the compound Ta_2Si . A linear traverse across the sample with an electron microprobe showed that the layer consisted of material containing 5 to 10 w/o silicon in a band about 5 μ wide. This silicon concentration is in good agreement with the stoichiometric value of silicon in Ta_2Si , which is 7.2 w/o Si.

A better understanding of this phenomenon requires further study. It is conceivable that the silicon in the system first diffused through the surface of tantalum with the help of liquid lithium as carrier and subsequently interdiffused to form a protective layer. The observed layer is in fact stable in the testing environment and may be responsible for the corrosion resistance of tantalum.

b. High-temperature Phases and Behavior

(i) (Th-U-Pu)S Bodies. Studies of the electrical properties of the actinide compounds with the NaCl-type structure have shown that there

³²Stull, D. R., and Sinke, G. C., Thermodynamic Properties of the Elements, Advances in Chemistry Series, No. 18, American Chemical Society, Washington, D. C. (1956).

³³Brugger, J. E., Chemical Engineering Division Research Highlights, May 1965-April 1966, ANL-7175 (1966), p. 142.

is a systematic increase in their resistivities progressing from ThX $\rightarrow$ UX $\rightarrow$ PuX. Likewise, the resistivity increases systematically as the non-metal atoms change from Group IVA to VIA.

In order to get a better understanding of the role of the nonmetal atom in controlling the effective valence and electronic configuration of the metal atoms in the various compounds, plots of unit-cell volume of the compound versus the cube of the CN12 metal radius were made for some mononitrides, monophosphides, and monosulfides of the rare-earth metals, transition metals, and actinides. Elements of the rare-earth series form NaCl-type compounds when combined with nitrogen, phosphorus, and sulfur and can therefore be used for a comparison with the isostructural actinide compounds. The same structure is also found in some transition metal compounds and these have been included in the plots. This type of approach was previously used by Nevitt and Rosen³⁴ for the monocarbides. Data needed for calculating the compound volumes and metallic radii were obtained from reliable literature sources.

The data show a consistent relationship between cell dimensions and metal radius for the rare-earth compounds. A smaller number of valence electrons appear to be present in the metal constituent of the compound as compared to its metal structure, thorium excepted. In order for the actinide mononitrides to conform to the curve, it is necessary for them to have about four electrons per atom in the 7s + 6d states. Likewise, between three and four electrons per atom are inferred for the actinide monophosphides and monosulfides. These results appear to be in good agreement with the 4+ state found by Didchenko and Gortsema³⁵ for the metallic component of US, ThS, and ThN.

The valence of the actinide constituent appears to be about the same for a common nonmetal atom. If this is true, then for each group of compounds, progression from ThX $\rightarrow$ UX $\rightarrow$ PuX would result in a higher density of 5f states in the actinide component. In the sulfides a greater degree of ionic bonding would be expected and a separation of f-states could then account for the semiconductor properties of PuS. This model is in agreement with the split in f-states proposed by Kazmierowicz for US.³⁶

In view of the above relationships several other actinide monosulfides may also have semiconductor properties. The two most likely suspects are NpS and AmS; however, the energy separation of the f-states in the former compound may be barely detectable. In compounds of

³⁴Nevitt, M. V., and Rosen, S., IV International Union of Crystallography Congress and Symposia, Rome, Italy, 1963, p. A-19.

³⁵Didchenko, R., and Gortsema, F. P., Magnetic and Electric Properties of Monosulfides and Mononitrides of Thorium and Uranium, *Inorganic Chem.* **2**, 1079 (1963).

³⁶Kazmierowicz, C. W., Galvanomagnetic Properties of Uranium Monosulfide, ANL-6731 (1963).

plutonium with other Group VIA nonmetals (PuSe and PuTe), a greater share of covalent bonding can be expected; therefore, quasimetallic conduction probably occurs.

The nonmetallic elements of Group IVA-VIA combine with the actinide metals to form a theoretically interesting group of inorganic compounds which have unfilled 6d and 5f energy levels and are characterized by their ability to exist as structure with mixed ionic and covalent bonding. Differences in electronegativity between bonding atoms and wide variations in the density of states of the metal components result in electronic structures that are extremely complex. The nonmetallic elements generally form compounds having the NaCl-type crystal structure. Although many of these compounds have properties that are not particularly suitable for nuclear fuels, the entire group of compounds is attractive for theoretical studies. Their common structure provides a basis for comparison of changes in properties associated with systematic changes in electronic configuration.

Previous work on the monocarbides, monophosphides, and monosulfides of uranium and plutonium has been reported. As part of the overall investigation of properties, the remaining Group IVA-VIA uranium and plutonium compound has been prepared. The monogermanides, monoantimonides, monoselenides, and monotellurides were made by arc-fusion of the elements. Compositions of 50, 53, and 55 a/o nonmetal were prepared so that one of the melts would be close to the stoichiometric composition even if slight loss of the nonmetal occurred through vaporization. Uranium monoarsenide has been prepared by reaction of powdered arsenic and decomposed uranium hydride. The same process is not being used to prepare PuAs. The melting points of these compounds will be measured in our tungsten V-filament furnace. X-ray diffraction patterns of the above alloys indicate they are close to the stoichiometric composition. Some of these photograms will be used to calculate precision lattice constants. Others have blurred high-angle doublets, and high-temperature heat treatments are needed in order to obtain good X-ray patterns. These heat treatments are in progress.

(ii) U-S-O System. Eight compositions on the U-S join were prepared from mixtures of U and US_2 powders. The as-mixed compositions included $US_{0.85}$, the known compounds US, U_2S_3 , U_3S_5 , and US_2 , and points midway between adjoining compound pairs. Pellets that had been isostatically pressed at 55,000 psi were sintered at 1460°C for 3 hr in vacuum to densities of about 80% theoretical. Only US_2 exhibited significant weight loss, its composition being reduced to $US_{1.71}$ by the firing. All samples were X rayed and observed metallographically.

The stoichiometric US appeared to be single-phase and had a relatively low lattice parameter of $a_0 = 5.4853 \text{ \AA}$, which was attributed

to nitrogen impurity from the US_2 . Since the range of a_0 extended only from 5.4865 Å, for US in equilibrium with uranium ($US_{0.85}$), to 5.4841 Å, for US in equilibrium with U_2S_3 ($US_{1.25}$), only a small homogeneity range is indicated for US at 1460°C.

The stoichiometric U_2S_3 composition was about 99% pure, the remainder being U_3S_5 . The lattice parameters for its orthorhombic Sb_2S_3 -type structure were determined to be $a_0 = 10.35$ Å, $b_0 = 10.61$ Å, and $c_0 = 3.87$ Å. U_2S_3 has a purple-gray color (in polished section) and can be fairly easily distinguished from U_3S_5 which has a lighter, more pure gray color. The stoichiometric U_3S_5 composition was only about 95% pure, some areas of U_2S_3 being observed toward the center of the pellet. The lattice parameters for the orthorhombic U_3S_5 structure were determined to be $a_0 = 7.44$ Å, $b_0 = 8.11$ Å, and $c_0 = 11.78$ Å. The blue-gray phase, which appears when sulfur is added to UOS, was mistakenly identified as U_3S_5 on the basis of relative line-intensity data found in the literature. Revised intensity data from this recent work established that this blue-gray phase was actually U_2S_3 . It appeared blue- instead of purple-gray on account of a thin oxide film that had formed on the polished surface. As-polished purple-gray U_2S_3 can be readily distinguished from UOS, which has a light, pure gray color almost identical to that of U_3S_5 .

The $US_{2.00}$ pellet had a cored structure, the outer shell being U_3S_5 while the inner core remained predominantly US_2 . The colors of the two phases were identical after polishing; however, the US_2 readily acquired a brownish oxidation stain whereas the U_3S_5 did not. From observation of numerous polished surfaces exposed to moist ambient air, the following order of reactivity can be given: $US_2 > U_3S_5 > U_2S_3 > US > UOS$.

The room-temperature electrical resistivities of the US_x series pellets were measured by a variation of the four-point probe method in which wire loops were tightened circumferentially onto a cylindrical sample for both the current and potential leads. This method was found to give much more stable and reproducible measurements than when end contacts and pointed probes were used on these particular materials. The following values in $\mu\Omega\text{-cm}$ were obtained for the pure (as-mixed) compounds corrected to X-ray density:

US	263
U_2S_3	775
U_3S_5	42,900
US_2	28,400

The resistivity values for the samples containing mixtures of two phases were in all cases intermediate between those for the "pure" phases and quite representative of the microstructural phase placement.

Since the value for US_2 actually represents a two-phase structure, it is meaningful only in the indication that US_2 is a better conductor than U_3S_5 . US has been shown elsewhere to be a metallic-type conductor (the resistivity increases slightly with temperature). The resistivity value for U_2S_3 suggests it may either behave as a metal or semiconductor, whereas U_3S_5 and US_2 would seem more likely to have semiconductor characteristics.

c. Mechanical Properties of Uranium Compounds. Several specimens of UO_2 , approximately 0.31 cm in thickness, were tested at 1000°C and at three strain rates. These specimens were approximately half the thickness of the specimens heretofore used. Values of modulus of rupture were considerably higher for all strain rates than the values obtained previously at room temperature. The increases for slow, medium, and fast strain rates were 54, 74, and 65%, respectively. These results are shown in Table XV.

TABLE XV. Mechanical Properties of UO_2 at 1000°C in Vacuo

Number of Specimens	Density (g/cc)		Modulus of Rupture (kg/cm ²)		Mean Load Rate (g/sec)	Mean Total Strain (mm)	Remarks
	Mean	Standard Deviation	Mean	Standard Deviation			
4	10.66	0.005	3116	232.4	26.0	0.24	Slow Strain
4	10.66	0	2371	308.8	147.2	0.14	Medium Strain
5	10.66	0.004	2171	561.5	342.6	0.15	Fast Strain

A number of electron micrographs were taken of several areas of the various specimens: (1) widened grain boundaries, which appear to be sites for dislocation buildup, (2) dislocation etch pits within the grains, and (3) the "mottled" areas observed in specimens examined with an optical microscope.

d. Irradiation of Prototype Paste Fuel Element. The in-reactor evaluation of a paste fuel element concept is the subject of a joint experiment by the United Nuclear Corporation and Argonne National Laboratory. UNC developed and supplied a fuel specimen consisting of a 50-50 mixture, by volume, of approximately 10-micron UO_2 powder and sodium in an AISI Type 304 stainless steel tube. ANL encapsulated and irradiated the experimental fuel rod in MTR to 2% burnup at a maximum jacket temperature of 600°C.

The fuel mixture had been expected to move upward because of thermal expansion of the sodium as the MTR was brought to power. Apparently, however, very little movement occurred, as determined by a series of thermocouples spaced at small intervals above the top of the fuel column. Initial postirradiation-inspection of the encapsulated fuel specimen by neutron radiography indicates that the fuel mixture is clustered at various

intervals within the specimen and is no longer uniformly distributed within the stainless steel tube. In addition, the tube appears to be bowed and distorted over the original length of the fuel column and may be breached in one area.

e. Irradiation of High-temperature Materials. Ceramic fuel materials being developed under the high-temperature materials program include UC-20 w/o PuC, PuC, and US jacketed in Nb-1 w/o Zr. These materials are under irradiation in instrumented capsules in the MTR. A summary of the irradiations is shown in Table XVI.

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

Capsule or S/A No.	Specimen Number	Design Parameters				Operating Conditions			
		Fuel Composition (w/o)	Effective Density (%)	Cladding OD (in.)	Cladding Thickness (in.)	Power Density (kW/cc) ^a	Max Cladding Temp (°C)	Burnup to Date	
								a/o (U + Pu)	fiss/cc x 10 ^{-20a}
56-11	MV-2	UC-20 PuC	79	0.281	0.021	1.2	470	6.4	16.5
56-8	MV-3	UC-20 PuC	81	0.281	0.012	1.2	715	6.9	18.3
56-8	MV-5	UC-20 PuC	80	0.281	0.012	1.2	705	6.6	17.3
56-11	MV-6	UC-20 PuC	80	0.281	0.012	1.2	480	6.9	18.1
56-13	Z-4	UC-20 PuC	79	0.174	0.015	1.2	665	1.8	4.6
56-13	Z-5	UC-20 PuC	79	0.174	0.015	1.2	585	1.8	4.6
56-13	Z-7	UC-20 PuC	79	0.174	0.015	1.2	570	1.8	4.6
56-13	C-45	PuC	84	0.174	0.009	1.4	700	1.8	7.7
56-8	S-7	US	80	0.281	0.012	1.0	535	5.2	10.1
56-8	S-8	US	89	0.281	0.012	1.0	725	7.2	15.6
56-8	S-9	US	76	0.281	0.012	1.0	750	7.2	13.2
56-8	S-10	US	91	0.281	0.012	1.0	690	7.2	15.9
56-11	S-15	US	82	0.281	0.012	1.0	380	4.3	8.6
56-11	S-16	US	90	0.281	0.012	1.0	510	6.2	13.5
56-11	S-17	US	88	0.281	0.012	1.0	500	4.9	10.4
56-11	S-18	US	77	0.281	0.012	1.0	610	6.7	12.5

^aBased on effective density.

Electron-microscopic studies are in progress on specimens of vibratorily compacted powders of UC-20 w/o PuC irradiated up to 6.9 a/o burnup. Studies by the two-stage carbon-replica method are being continued on high-burnup hypostoichiometric carbide specimens. The distribution of fission-gas bubbles and fission products, and other microstructural features will be compared for hyper and hypostoichiometric carbide specimens.

3. Radiation Damage to Structural Materials

a. Effect of Fast-neutron Irradiation on Jacket Materials. The effects of fast-neutron irradiation coupled with elevated temperature on the tensile, creep, creep-rupture, and tube-rupture properties of V-20 w/o Ti, V-15 w/o Ti-7.5 w/o Cr, AISI Type 304 stainless steel, Hastelloy-X, and Inconel 625 are being studied in the temperature range of 500 to 750°C at total neutron exposures up to 1×10^{23} n/cm² in an argon-helium environment. Twenty-three capsules, containing 60 tube-rupture specimens and 259 tensile-type specimens are being irradiated in EBR-II subassemblies XA07, XA08, and XO10 at temperatures between 500 and 590°C. These specimens have accumulated at the end of this report period maximum total neutron exposures ranging from 1.0×10^{22} to 3.8×10^{22} n/cm². Three capsules containing 26 tensile-type specimens of V-20 w/o Ti, three of

Hastelloy-X, and ten of AISI Type 304 stainless steel were removed from EBR-II subassembly XO09 at the end of run No. 22. The specimens will be shipped to the Lemont site for postirradiation testing. The specimens in XO09 were irradiated to a maximum total neutron exposure of 2.7×10^{22} n/cm² at 670°C. The status and identification of all capsules are given in Table XVII.

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

S/A No.	Capsule Number	Design Parameters			Operating Conditions		
		Cladding Composition	Type of Specimen	No. of Specimens	Specimen Environment	Max Specimen Temp (°C)	Exposure to Date (n/cm ² x 10 ⁻²²)
XA07	AS-9	V-20 w/o Ti	Tensile	16	Argon-helium	590	3.8
XA07	AS-10	Hastelloy-X	Tensile	16	Argon-helium	590	3.5
XA07	AS-11	304 SS	Tensile	16	Argon-helium	590	3.6
XA08	AS-1	V-20 w/o Ti	Tube-burst	12	Argon-helium	540	3.1
XA08	AS-2	V-20 w/o Ti	Tube-burst	12	Argon-helium	540	3.1
XA08	AS-3	Hastelloy-X	Tube-burst	12	Argon-helium	540	3.2
XA08	AS-4	Hastelloy-X	Tube-burst	12	Argon-helium	540	3.2
XA08	AS-5	304 SS	Tube-burst	12	Argon-helium	540	3.2
XA08	AS-6	V-20 w/o Ti	Tensile	16	Argon-helium	580	3.4
XA08	AS-7	Hastelloy-X	Tensile	16	Argon-helium	580	3.1
XA08	AS-8	304 SS	Tensile	16	Argon-helium	580	3.1
XA08	AS-12	V-20 w/o Ti	Tensile	16	Argon-helium	580	3.0
XO09	AS-14	V-20 w/o Ti	Tensile	13	Argon-helium	670	2.6
XO09	AS-15	V-20 w/o Ti	Tensile	13	Argon-helium	670	2.5
XO09	AS-27	304 SS	Tensile	13	Argon-helium	670	2.7
		Hastelloy-X					
XO10	AS-16	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.1
XO10	AS-17	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.2
XO10	AS-18	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.0
XO10	AS-19	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.1
XO10	AS-20	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.2
XO10	AS-21	V-20 w/o Ti	Tensile	13	Argon-helium	500	1.4
XO10	AS-22	Hastelloy-X	Tensile	13	Argon-helium	500	1.3
XO10	AS-23	304 SS	Tensile	13	Argon-helium	500	1.4
XO10	AS-24	304 SS	Tensile	13	Argon-helium	500	1.2
XO10	AS-25	304 SS	Tensile	13	Argon-helium	500	1.2
XO10	AS-26	304 SS	Tensile	15	Argon-helium	500	1.2

4. Techniques of Fabrication and Testing

a. Argonne Three-roll Tube Reducer. An improved technique of producing small-diameter, refractory-metal tubing from conditioned extruded tube blanks is to reduce their diameter and wall thickness in the newly developed Argonne three-roll tube reducer. This device has had enough tests on fast-reactor fuel-jacket materials to confirm its general utility as a processing tool.³⁷ The inconsequential amount of friction encountered in the process enables the reducer to effect greater reductions per pass than are possible by other means. Removal of reduced tubing from the mandrel has not been difficult; reeling has never been necessary. Should one wish to eliminate the tube-removal operation, however, a short plug of uniform diameter can be used as successfully as the mandrel.

³⁷Progress Report for February 1966, ANL-7176, pp. 27-29, and Progress Report for September 1966, ANL-7255, p. 29.

Parametric studies are now under way to establish the optimum in design criteria and in operational procedures. Roll design, for example, is critical to both surface finish and the resulting residual stress pattern in the tube wall. Improper roll contour can promote predictable cracking behavior in tubing materials of limited ductility. Operational procedures such as the feed and index rates (tube advance and rotation per reduction cycle) can affect surface quality and the power required to affect the reduction. These parameters, and others, are receiving critical analysis.

b. Determination of Elastic Moduli of High-temperature Materials by Ultrasonics. The first Type 304 stainless steel bar that has been machined for use in the new high-temperature furnace was received, and is being used in the continuing study of the variation of elastic properties of commercially available stainless steels. Considerable time was spent in learning how to mount thermocouples in the furnace. The main feature in the design of the new furnace is that the thermal gradient in the index region of the specimen can be reduced by positioning the specimen within the heat zone properly. Since the specimen, and hence the attached thermocouples, moved up and down, care had to be exercised in the placement of the thermocouples. A run was made on this specimen to about 1000°C, when one of the couples broke. During this run, the temperature difference between the ends of the index region was kept less than 10°C.

At the higher temperatures an attempt was made to correlate the readings of the thermocouples with the readings of an indicating optical pyrometer focused on the surface of the specimen. The pyrometer readings were much too high, indicating that the top heat shields reflected light from the heating element to the specimen surface. Means for correcting this important source of error are now being investigated.

c. Ultrasonic Instrument and Transducer Development. Preliminary measurements of longitudinal velocities in three porous tungsten samples have been made. Two of these samples contain 100 μ particles and have theoretical densities of 73.8 and 97%. The other sample, 83.7% dense, has 400 μ particles. Calculations of the acoustic impedances for these samples reveal that densities below 70% in this particle-size region will be required to obtain an acoustic-impedance match with lead zirconate-titanate elements in ultrasonic transducer probes. In tungsten it is anticipated that better attenuation characteristics to ultrasound will be achieved at densities below 70%.

d. Development of a Neutron-Image Intensification System. Gamma-irradiation studies with an image orthicon television camera tube (RCA Type 5820) have continued (see Progress Report for October 1966, ANL-7267, p. 59). A total cobalt-60 gamma exposure of about 10^7 R yielded a further

decrease in light sensitivity of about an order of magnitude, as compared to the light sensitivity of the tube irradiated to 10^6 R. For exposure to 10^7 R, the tube sensitivity dropped by a factor of about 20 as compared to the unirradiated tube.

At total exposure of 10^8 R, an additional loss factor of 16 as compared to total exposure of 10^7 R, was noted. Total loss in sensitivity is, therefore, about 320, for 10^8 R exposure. At this level, however, a great deal of noise was encountered, indicating that additional effects, other than just loss in light transmission through the darkening glass, are taking place. At this change in electrical response of the tube, the gamma-irradiation test was discontinued.

The image orthicon television system (with a new camera tube) was also used to measure the television response of a europium-activated Li^6I neutron scintillator. The television image of this scintillator could be observed for incident thermal-neutron intensities as low as 10^6 n/cm²-sec. This represents improvement by a factor of about four as compared to the use of a Li^6F , $\text{ZnS}(\text{Ag})$ neutron scintillator.

e. Measurement of Thermal Conductivity of Irradiated Fuel as a Function of Burnup and Temperature. Measurements of thermal conductivity at temperatures to 600°C have been planned for irradiated uranium-5 w/o F's fuel specimens. (Room-temperature measurements for these specimens were reported in Progress Report for April 1966, ANL-7204, pp. 54-59.) In preparation for these measurements, modifications, which will simplify hot-cell work, are being made on the high-temperature facility, and several auxiliary pieces of hardware are being assembled.

C. Engineering Development

1. Master-Slave Manipulator Systems

a. Electric Master-Slave Manipulator, Mark E4A. Continued experience and testing of the Mark E4A shows that the general performance is good. A few modifications are underway to improve further the manipulating performance and reliability.

Electrically actuated brakes for the slave-arm motions are being designed which will permit the manipulator to be locked in any position. Clutches will be provided to prevent damage to the arm which could otherwise be caused by external forces when the brakes are on. The brakes for six manipulator motions are to be controlled by a single switch. A separate, independent switch will be used for the tong squeeze-motion brake, so that objects can be held and moved about without the operator maintaining a squeeze force.

An effort is being made to improve the manipulator stiffness (and thereby improve its sensitivity) in the "Z" motion. Some of the problem appears to be due to mechanical backlash in the bearings of the servo drive unit. High-precision, low-internal-clearance bearings are on order. This problem will be studied further after the new bearings are installed.

We are also considering a change in the synchro gear ratio. The present ratio was chosen to gain a high signal-to-noise ratio. However, with this gear ratio, the positional nulls are close together and it is easy inadvertently to move the master arm out of synchronization with the slave. A higher gear ratio would improve this situation, and it now appears that the synchro signal would be adequate with the higher gear ratio.

A number of other improvements are planned. These include thermal protection devices for the servo motors, a cooling fan for the "X" motion servo drive unit, and shrouds for the exposed mechanical cables on the slave arm.

b. Head-controlled TV for Manipulator Systems, Mark TV2. In this system the camera and monitor are slaved to follow a manipulator operator's head in five motions: two rotational (pan and tilt) and three translational. The operator is free to move his head in the remaining rotational motion, but this movement does not control any motion of the camera or monitor. After donning a simple adjustable headpiece, the operator gets new views simply by moving his head much as he would if he was at the camera position.

The three translational motions incorporated in the Mark TV2 provide some interesting advantages over an earlier two-motion system

(the Mark TV1). One important advantage is the increased depth information that the manipulator operator can gain by moving his head either from side to side (or up and down) to gain new points of view. Since these motions can be made easily, the operator can establish a 3-dimensional mental image. The third translational motion provides another advantage. To get a close look at his work, an operator merely steps forward much as he would if viewing the work directly. The monitor also moves forward and the camera moves closer to the work.

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

a. Boiling Liquid-metal Technology

(i) Niobium-1% Zirconium Loop. Preparations for the resumption of loop operation after completion of modifications are nearly complete. The following items were completed: calibration of dump-system pressure gauge; detector system for the partial-pressure analyzer removed and reactivated; valves for the liquid nitrogen system checked for leaks and reconditioned; thermocouple and data-system readouts checked and recorded; sodium hot-trap system restarted to provide clean charge; instrumentation calibration schedule firmed up. Final system cleanup of the loop externals is underway, and loop operation is scheduled for December 5.

(ii) Electron-bombardment Heater Experiment. This experiment is designed to gain experience with electron-bombardment heating (EBH), 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.

Approximately 500 hr of testing with the 0.030-in.-dia, uncarburized thoriated-tungsten filament and 400 hr with the 0.030-in.-dia tungsten filament have been achieved. Heat fluxes ranging up to 350,000 Btu/hr-ft² have been supplied to the anode during these tests. This experiment is approaching limiting design conditions, and modifications are underway to provide for higher heat fluxes for short periods of time.

A sodium leak occurred at the outside wall of the vessel at a thermocouple well. Examination of the vessel is underway to determine exactly where the leak is and why it has occurred.

These tests and an evaluation of the EBH study by EIMAC Corporation will provide the basis of the final EBH design for use in the 2100°F Nb-1% Zr loop.

b. General Heat Transfer

(i) Heat Transfer in Double-pipe Heat Exchangers

(A) Countercurrent Turbulent Liquid Metal Flow. Experiments with the second mercury-to-mercury concentric tube heat exchanger have been delayed pending reworking of the canned-rotor pump impeller and replacement of the seals in the mechanically sealed pump. Both repairs are in progress. A third test section with a "narrow annular space" is being designed. The primary difficulty with the construction of the narrow annular space test section is the alignment of the inner tube within the outer. A design using neoprene "O" ring seals to eliminate welding of the inner and outer tubes is being considered.

Analytical investigations continue. These included a more detailed study of the validity of the "narrow" annular space approximation used in previous analyses. It has been found that for the purpose of predicting fully developed heat transfer coefficients and overall heat transfer rates, the "narrow" annular space approximation gives sufficiently accurate results for annular radius ratios as small as 0.5.

(B) Nonsymmetrical Geometries. A small heat transfer loop for the purpose of performing experiments with sodium in model heat exchangers with nonsymmetrical cross sections (see Progress Report for September 1966, ANL-7255, p. 56) has been designed. Miscellaneous components have been obtained, including an EM pump, blower, and an air cooler previously used with EBR-II.

(ii) Heat Transfer in Liquid-metal-cooled Reactor Channels. Investigations of the effect of axial heat conduction within the liquid metal in heated channels for the case with long unheated inlet and outlet ducts have been nearly completed. The study has been extended to include the generalized symmetrical geometry represented by the annular space heated at both sides with flux distributions arbitrary functions of axial position. A simple temperature-correction factor has been found for this general case that offers a convenient basis for estimating corrections to temperatures predicted by analyses which neglect axial heat conduction. This factor is defined by

$$\theta = \left(\frac{1+R}{4} \right) (Pe)^2 \frac{(t - t^*) k}{(Rq_1 + q_2) D},$$

where

R = annulus radius ratio ($0 < R < 1$);

Pe = Peclet number;

- t = fluid temperature at any radial and axial position;
- t^* = fluid temperature as above, but predicted by analyses which neglect axial heat conduction;
- k = fluid thermal conductivity;
- q_1 = local heat flux density into fluid from inner wall of annular space;
- q_2 = local heat flux density into fluid from outer wall of annular space;
- D = hydraulic equivalent diameter of duct.

The factor θ is dimensionless, and for sufficiently large distances from the duct inlet and outlet (typically, a few duct diameters is "sufficiently large,") is equal to unity.

Related studies for the uniform flux case at the University of Illinois³⁸ and by students of the AMU-ANL Summer Engineering Practice School³⁹ have been published recently. A paper, describing the results of the more detailed investigation, is being prepared.

c. Effect of Interfacial Transport Processes on Film Condensation. Construction of the loop was completed and preliminary tests of the system were made. At high flow rates of water (above 2 ft/sec in the injection inlet), flow distribution in the entrance region of the film became highly three-dimensional. The source of this disturbance was traced to the injector itself. The necessary modifications are now being made.

d. The Pressure Recovery of a Two-phase One-component Flow through a Diffuser. A high efficiency of converting kinetic energy in a one-component two-phase fluid to pressure energy is of primary importance to the development of an magnetohydrodynamic generator. This investigation will try to optimize the diffuser efficiency by studying the effect of diffuser angle, area ratio, and cooling the diffuser walls for steam-water flow. The program will deal with subsonic and supersonic, adiabatic and nonadiabatic flow through diffusers.

3. Instrumentation and Control

a. Neutron-parameter Determinations: The number of pulses produced by a given fission chamber per unit neutron flux can be theoretically predicted from the equation

³⁸Burchell, W. E., and Jones, B. G., The Influence of Axial Conduction on Heat Transfer to Liquid Metals in Reactor Coolant Channels, Trans. Ans., 1966 Winter Annual Meeting, 9, 568 (1966).

³⁹Krolkowski, T. S., Ginsberg, T., Moy, H. C., and Yadigaroglu, G. C., The Significance of Axial Heat Conduction in Liquid Metal Cooled Heated Ducts, AMU-ANL Summer Engineering Practice School Report, Sept. 2, 1966.

$$N_n = \frac{N_a \sigma_f W E}{1000 A \text{ sec-nv}} \text{ pulses} \quad (1)$$

where

N_n = number of pulses produced per unit flux;

N_a = Avogadro number (6.02×10^{23} atoms/g-atom);

σ_f = fission cross section in barns;

W = weight in milligrams of fissionable material;

E = the counting efficiency;

A = atomic weight of fissionable isotope.

For the fission chamber considered in these experiments,

$$W = 1720 \text{ mg } U^{235}$$

$$E = 0.59,^{40}$$

and therefore

$$N_n = k = 1.43 \text{ pps/nv.}$$

This number was experimentally checked by extrapolating the integral bias response curve to neutron pulses to zero discriminator voltage. By this technique a counting sensitivity of 1.55 pps/nv was obtained when all the pulses are counted. This is in good agreement with the theoretical value.

The mean charge per neutron pulse was determined in two different ways. First, the mean neutron current (I_{DCn}) was measured at a known neutron flux and the charge determined by

$$\bar{q}_n = \frac{I_{DCn}}{N_n} = \frac{1.85 \times 10^{-13}}{1.43} = 1.29 \times 10^{-13} \frac{\text{coulomb}}{\text{pulse}}.$$

The second method consisted of photographing the neutron pulses displayed on an oscilloscope and by using an equation that describes the output pulse shape determining the average charge necessary to reproduce the average pulse shape. By this method, the following values were determined for the two different cables used in the experiment:

⁴⁰ Abson, W., Salmon, P. G., and Pyrah, S., The Design, Performance, and Use of Fission Counters, Proc. IEEE, 105, Part B, 349-56 (July 1958).

$$\overline{q_n} = 1.45 \times 10^{-13} \frac{\text{coulomb}}{\text{pulse}} \text{ from the short cable;}$$

$$\overline{q_n} = 1.50 \times 10^{-13} \frac{\text{coulomb}}{\text{pulse}} \text{ from the long cable.}$$

This is in good agreement with the charge measured by the first technique and the value of 1.5×10^{-13} coulomb reported in the literature. A charge $\overline{q_n}$ of 1.4×10^{-13} coulomb per pulse will be the accepted value.

The mean square charge per neutron pulse was experimentally determined by measuring the voltage E_{rmsn} at a given neutron flux level and then calculating the charge from

$$\overline{q_n^2} = \frac{2C_0(E_{\text{rmsn}})^2}{N_n R_0 K_1},$$

where

C_0 is the cable capacitance;

E_{rmsn} is the root-mean-square voltage;

N_n is the number of neutron pulses;

R_0 is the input circuit resistance;

K_1 is given by

$$\frac{2\tau_0}{\tau_e} \left[1 - \frac{\tau_0}{\tau_e} \left(1 - e^{-\tau_e/\tau_0} \right) \right];$$

τ_0 is $R_0 C_0$;

τ_e is the electron collection time of the detector.

The following values were determined from measurements taken with two different cable lengths, and hence different capacitances:

$$\overline{q_n^2} = 6.37 \times 10^{-27} \text{ coulomb}^2 \text{ per pulse from the short cable;}$$

$$\overline{q_n^2} = 6.59 \times 10^{-27} \text{ coulomb}^2 \text{ per pulse from the long cable.}$$

A value of 6.5×10^{-27} coulomb² per pulse will be the accepted value.

b. EBR-II Test Results. The wide-range neutron-monitoring equipment (see Progress Report for March 1966, ANL-7193) has been installed at the EBR-II. The detector has been placed in the "J-2" instrument thimble.

Approximately 55 ft of RG-114-U cable were needed between the detector and the preamplifier. By use of the results obtained from previous measurements, predictions of the system response were made prior to reactor operation. These predictions are compared to the measured response in Fig. 15. The measurements were taken after a prolonged shutdown and

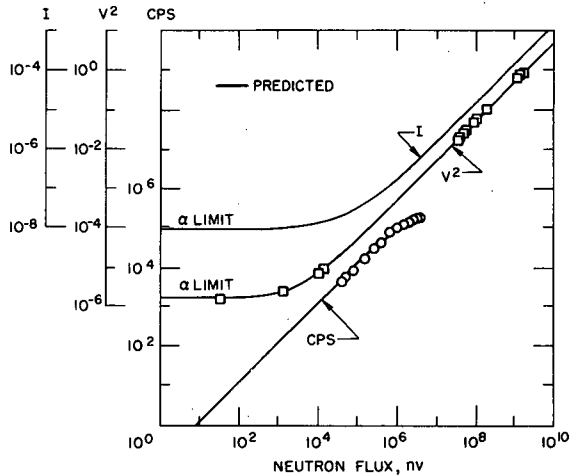


Fig. 15. System Response as a Function of Neutron Flux

represent the system responses in the absence of significant residual gamma activity. The measured responses are in excellent agreement with the predictions and show that the performance of the mean-square-voltage system can indeed be calculated if system parameters are adequately defined.

A portion of a typical reactor startup is shown in Fig. 16. The reactor is kept on an essentially constant period of ~ 60 sec until a power of about 2 MW is reached. This can be seen to occur at 590 sec, when the flux levels out quite rapidly. The counting system begins to saturate at approximately 2×10^5 cps, for which a counting loss of 20% was predicted. The linear overlap between the counting-system output and the true mean-square-voltage signal can be seen to be almost two decades, whereas the average magnitude-squared voltage response yields very little, if any, overlap. This again is proof that an average magnitude-squared signal cannot be used with a single detector system when linear overlap is a requirement.

c. A.C. Flux Penetration in a Conducting Cylinder.

The variation of a.c. field intensity and total flux in a cylindrical conductor with an applied axial magnetizing force is a problem that is common to several instrumentation problems: measurement of void fraction in liquid metals by electromagnetic coupling, measurement of liquid-metal level by electromagnetic coupling, and transmission of data from a fuel assembly by electromagnetic coupling.

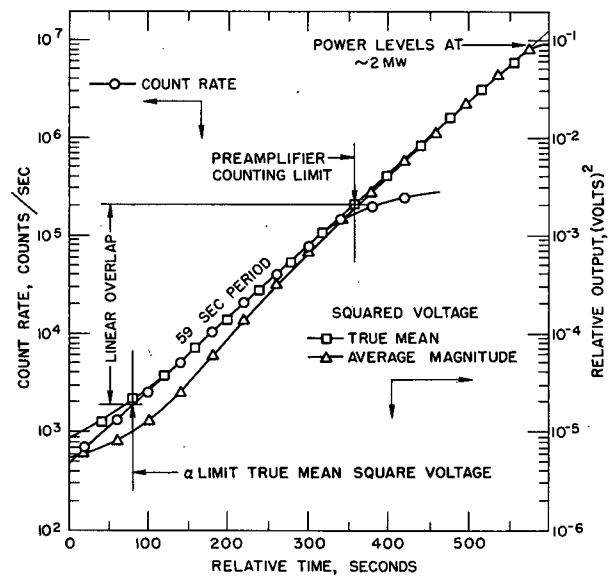


Fig. 16. Measurements at Reactor Startup

A step-by-step solution of the flux-penetration problem was programmed for our 160A computer. These computer data are in a form which provides direct calculation of the field variation and total flux in the cylinder. A comparison with analytic solutions obtained using Bessel functions shows the step-by-step solution to be sufficiently accurate for practically all engineering work.

The tabulated data were used to calculate curves of the attenuation and phase characteristics for sodium at 150°C. The very high rates of attenuation, even at the 60-Hertz power-line frequency, made it quite evident that any coil system providing coupling between small coils on the fuel subassemblies and large coils near the vessel wall is impractical. However, coupling between closely spaced coils, one on the fuel subassembly and the other placed around it, is workable (see Table XVIII).

TABLE XVIII

Frequency, Hertz	60	1,000	10,000
Thickness of sodium for 6-db attenuation and 42° phase shift, in.	0.65	0.16	0.05

On the basis of convenient clearance spaces and tolerable signal loss, the coupling system in a sodium medium appears workable for signal carrier frequencies up to 10 kc.

d. Condensing Injector Studies. Preliminary data have been obtained for condensing injector performance with steam and water. Analysis of the data indicates that effects of relative velocity of vapor to liquid, liquid subcooling, and liquid jet breakup are significant in reducing condensing injector performance.

Models for predicting condensing injector performance are being prepared for numerical computation. Results will be compared with the data obtained thus far and additional Freon-11 data to determine the applicability of any numerical results. In addition, further experimental data is to be taken to substantiate the trends thus far established and to investigate the effects of inlet vapor quality and superheat. Comparison of Freon-11 and water data can be made.

D. Chemistry and Chemical Separations

1. Fluoride Volatility Processes

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

(i) Fluorination of UO_2 - PuO_2 -Fission Product Pellets. Development studies are being performed in a 2-in.-dia fluid-bed reactor to determine optimum conditions for fluorinating UO_2 - PuO_2 pellets containing fission products. It had been observed in previous runs (see, for example, Progress Report for October 1966, ANL-7267, p. 66) that fine particles containing plutonium collected in the disengaging and filter sections of the fluid-bed reactor, and were not returned to the fluid bed for fluorination during the run. Recovery of this material had been achieved only by rapping and brushing of the interior surface of the reactor. The plutonium content of these fines amounted to as much as 15% of the plutonium charged to the reactor.

An experiment has been performed to determine whether plutonium could be recovered from the fines by recycle-fluorination using 90 v/o fluorine. The fines (18 g, containing 1.2 g plutonium), recovered from the reactor following runs Purse-10, -11, and -12 (see ANL-7267, p. 66), were fluorinated in a boat reactor using 90 v/o fluorine for 10 hr at 300°C, for 5 hr while the temperature was increased from 300 to 550°C, and for 3 hr at 550°C. By this treatment, the plutonium concentration in the fines was reduced from 6.7 to 0.2 w/o. This residue amounts to 0.4% of the plutonium charged to the reactor in the three runs. It appears that the extent of plutonium removal from the fines is similar to that achieved during fluorination of plutonium-bearing oxide fuel in a fluid bed of alumina and therefore, if the fines were returned to the bed prior to fluorination with 90 v/o fluorine, high plutonium removal from the fines would be expected. Attempts are being made to determine whether the principal source of plutonium in the fines is the result of elutriation or PuF_6 decomposition.

(ii) Fluorination of Complex Plutonium Fluorides. Plutonium fluorides form complex compounds with alkali metal fluorides. A series of laboratory-scale experiments have been performed to investigate the stability of several alkali metal-plutonium fluoride complexes toward fluorination with fluorine at elevated temperatures. A complex salt, $7\text{NaF} \cdot 6\text{PuF}_4$, prepared by adding excess NaF to a solution of plutonium (IV) nitrate, was contacted with 100 v/o fluorine for 2 hr at 250°C, 2 hr at 400°C, 1 hr at 450°C, and 1 hr at 550°C. The samples were weighed after each hour of exposure. It was observed that $7\text{NaF} \cdot 6\text{PuF}_4$ is not stable in a fluorine atmosphere at temperatures above 250°C and that a fraction of the plutonium is fluorinated to PuF_6 . The solid product after reaction with fluorine was identified by X-ray diffraction as $\beta_3\text{-}2\text{NaF} \cdot \text{PuF}_4$. This material appears to be stable in

fluorine up to 550°C. On the basis of these observations, it appears that efficient recovery of plutonium from a sodium fluoride complex by fluoride volatility techniques would be difficult.

An experiment was performed to identify the complex formed by reaction of PuF_6 with NaF at 100°C. The product was identified by X-ray diffraction as $3\text{NaF} \cdot \text{PuF}_4$. On the basis of previous observations which indicated that the stability of the complex fluorides towards fluorination increased as the mole ratio of sodium to plutonium increased, it appears that the 3:1 complex would be very stable in a fluorine atmosphere, and removal and recovery of plutonium from the complex would be difficult.

The stability of a cesium fluoride-plutonium fluoride complex, tentatively identified as Cs_2PuF_6 , toward fluorine at 400 to 500°C was evaluated in an experiment. No significant reaction between fluorine and Cs_2PuF_6 was observed during exposure of the complex salt to fluorine for 3 hr at 400°C and 1 hr at 500°C.

(iii) Instrument for Plutonium Analysis. An experimental program is under way to develop techniques for remote plutonium analysis based on the counting of neutrons derived from (α, n) -type reactions. Current studies involve the use of a water-moderated He^3 detector for estimating the quantity of plutonium in solid samples. In the application of this instrument to the analysis of plutonium in process vessels and lines, the instrument response may be markedly affected by radiation from plutonium adjacent to the source being measured. Because of shielding requirements to attenuate neutrons from undesired directions, it is difficult to achieve a large degree of angular discrimination with a portable detector weighing less than 10 lb. Therefore, the possibility of achieving some degree of discrimination by rearrangement of the detector shielding is being investigated.

Preliminary tests with a 0.064-in.-thick cadmium sleeve around the He^3 tube indicated that efficient shielding may be obtained with cadmium. However, such counts are still partly obscured by scattered neutrons from the environment. The addition of a paraffin moderator does not appreciably alter the number of scattered neutrons counted. A cadmium-shielded detector, designed to minimize counting of scattered neutrons, is being constructed.

(iv) Neptunium Fluoride Chemistry. Laboratory-scale studies are being carried out to determine the behavior of neptunium when oxidized nuclear fuel is reacted with bromine pentafluoride. It has been determined that the reaction between NpF_4 and BrF_5 at 300 to 400°C produces volatile NpF_6 (see Progress Report for October 1966, ANL-7267, p. 67). Recent experiments have shown that NpF_6 reacts readily with bromine at 30°C to

produce solid NpF_4 . In this reaction, the bromine is fluorinated to BrF_3 , but not to BrF_5 , even in the presence of excess NpF_6 .

Since UF_6 does not react with bromine under these conditions, this reaction affords a means for separating uranium from neptunium. A series of experiments are being performed to evaluate this separation technique.

A series of experiments is being performed to define the nature of the reaction between neptunium dioxide and fluorine or bromine pentafluoride. Particular emphasis is placed on identifying the intermediate compound formed in the reactions. Two experiments were performed in which NpO_2 was partially reacted with fluorine at 350°C in one case, and partially reacted with BrF_5 at 300°C in the second case. In both cases a green layer was observed over a dark-brown core which closely resembled the original NpO_2 . X-ray analyses of the green layer indicated the presence of NpF_4 . No evidence of NpO_2F_2 was found. The fluorination of neptunium dioxide, therefore, appears to be of the same nature as that of plutonium dioxide in which plutonium tetrafluoride is formed as an intermediate and not of uranium dioxide in which uranyl fluoride is formed as an intermediate.

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

(i) Process Development Studies for Uranium Dioxide Fuels.

Engineering-scale studies are being performed in a 3-in.-dia fluid-bed reactor facility to determine the effects of important process variables on the fluorination of UO_2 fuels by bromine pentafluoride. The fluid-bed reactor used in previous experiments in the facility (see Progress Reports for August 1966, ANL-7249, pp. 59-60, and September 1966, ANL-7255, pp. 58-59) was cooled by natural air convection. A new reactor, equipped with an air-water cooling system, has been installed in the facility. Additional modifications of the facility have included the installation of a device for sampling the off-gas from the reactor. Future modifications which must be made, prior to resumption of experimental studies, include the installation of a BrF_5 -metering system and an electrical power supply for the new reactor.

(ii) Fluid-bed Studies with Irradiated Fuels. A series of experiments are under way in a $1\frac{1}{2}$ -in.-dia fluid-bed reactor to determine the distribution of fission products during the processing of highly irradiated UO_2 fuel by the oxygen-bromine pentafluoride-fluorine process. An experiment (BRF-2) was completed using a processing sequence consisting of fuel oxidation at 450°C , uranium volatilization with BrF_5 at 300°C , and plutonium volatilization with fluorine at 300 to 550°C . Although complete analytical results are not available, some preliminary observations on the behavior of certain fission product species during the process can

be made. The major gamma-active contaminant found in the uranium product was ruthenium-106. The plutonium product contained ruthenium-106 and niobium-95. About 13% of the krypton-85 was released during the pellet oxidation step; the remainder of the krypton activity volatilized during the fluorination of uranium with BrF_5 .

Analytical results obtained from the previous run (BRF-1, see Progress Report for September 1966, ANL-7255, p. 60) showed that about 99% of the plutonium present in the irradiated pellet fuel was removed from the fluid bed. The final plutonium concentration in the bed was 0.005 w/o.

A trapping system to investigate the decontamination of PuF_6 from high-boiling fission product fluorides by sublimation of the PuF_6 has been built and installed in a hot cell. Several experiments are planned to evaluate the facility and to develop sampling procedures prior to incorporating the system into tests with irradiated fuels.

(iii) Basic Studies on the Fluidization of Fine Particles. In the oxidation of uranium dioxide pellet fuel, fine particles of U_3O_8 are produced. Since the fluidization of fine particles is frequently made difficult by the tendency of the particles to aggregate or by channeling of the gas through the bed, a development program has been established to analyze the properties of a fluidized bed of fine particles in order to predict the behavior of the particles during fluidization. Initial studies are directed toward describing the behavior of a fluid bed by analysis of the mechanical vibrations generated by the bed. Electronic equipment for recording the vibrations has been installed and is being tested.

IV. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. General

The architect-engineer, Burns and Roe, Inc., is continuing a minimal effort on the AARR facility design. Activity is centered around the development of specifications for Construction Package 1, primary pumps, and primary heat exchangers. Review of the Title I report is still in progress.

The redirection of the AARR project effort to employ the HFIR core has resulted in numerous changes to the preliminary safety analysis report (PSAR). In order to handle these changes in an expeditious manner, it was decided to divide questions from AEC-CH, RDT, and DRL into the categories of "facility" or "core" type questions. All AEC facility-type questions were answered in a supplement to the PSAR, "Supplement 1," issued on November 9, 1966.

The core-type questions will be answered by updating Section IV, Design Description, of the PSAR with new material relating to the HFIR core and will include responses to those questions on the core. This material will be presented in "Supplement 2" to the PSAR and is under preparation.

2. Fuel and Core Development

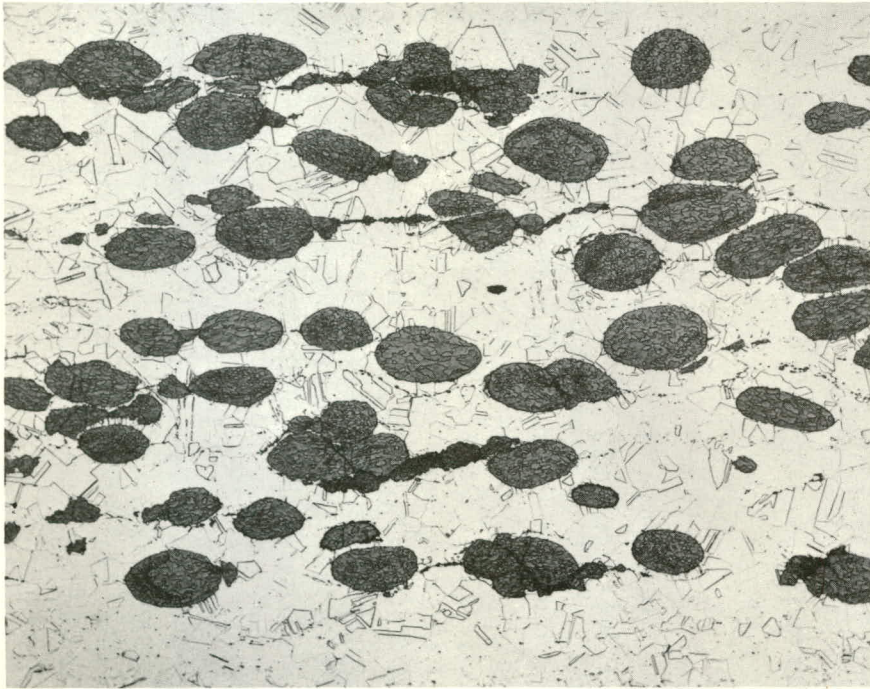
a. Thermal Conductivity in AARR Fuel Plates. The preliminary data reported in Progress Report for October 1966, ANL-7267, p. 70, were in error due to incompleteness at the time of reporting.

Plate BMI-57 that exhibited thermal properties that decreased with increasing temperatures was a purposely defective plate fabricated with a very porous low-crust-strength oxide. Figure 17 shows cross sections of plate BMI-57 and of plate BMI-50, using a commercially produced high-quality UO_2 . In both cases the bond area between clad and matrix shows grain growth across the boundary. It should be emphasized that there is complete bonding of clad and matrix and no evidence of non-bond as reported earlier.

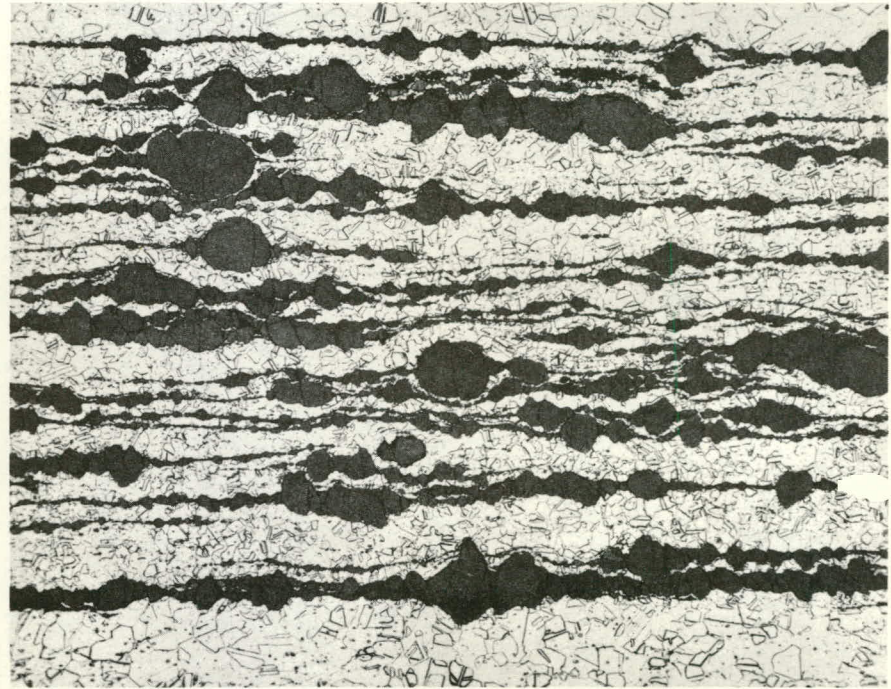
The adverse effect of severe stringering, as for plate BMI-50, is shown in Fig. 18, containing plots of thermal conductivity K and thermal diffusivity α .

b. Termination of Development of SS- UO_2 Cermet Core

(i) Thermal Stress Analyses of Fuel Plate. The following contains an outline of tasks specified for the thermal stress analysis



BMI-50 Plate



BMI-57 Plate

Fig. 17. Cross Sections of Fuel-plate Materials Used in Determining Effect of Dispersion Quality on Thermal Conductivity and Thermal Diffusivity in 37 w/o UO_2 -SS-347 AARR Fuel Plates

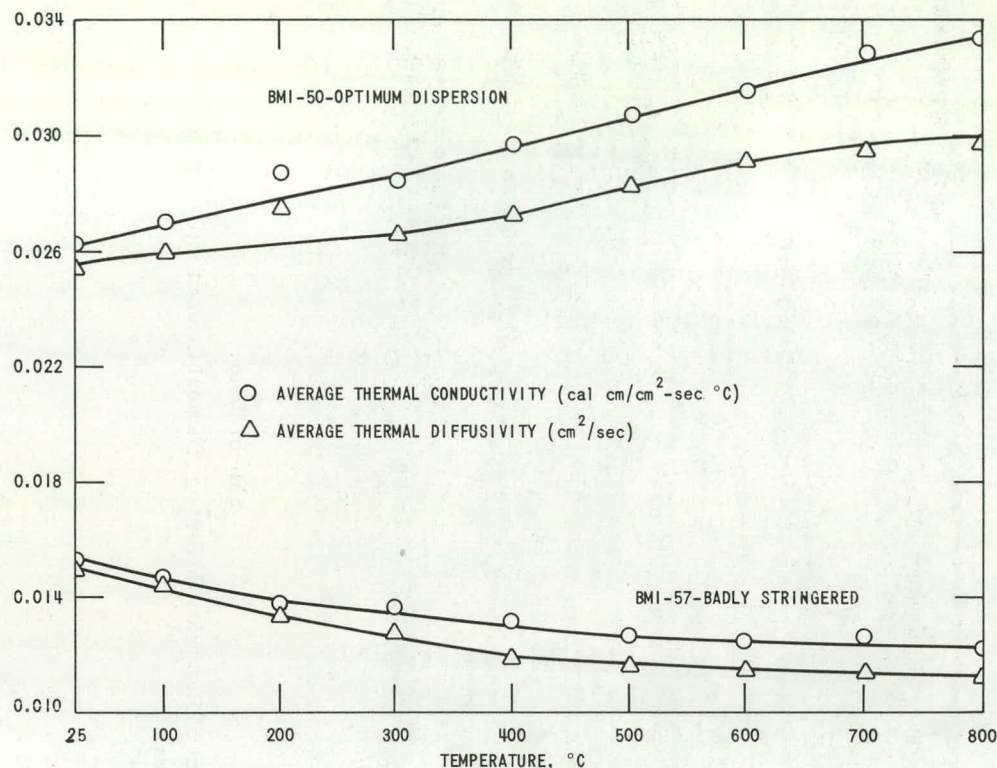


Fig. 18. Variation in Thermal Conductivity and Thermal Diffusivity in 37 w/o UO₂-SS-347 Cermet Plates having Different Dispersion Quality

of fuel plates and their status at the time of termination of activity. All activities were directly related to flat-plate fuel elements and it is not anticipated that the results will have any direct application to the HFIR core concept.

Thermal stresses in the fuel plates due to temperature variations through the plate thickness were computed based on the assumption of a free plate. Computations gave stresses of the order of the yield stress of the plate material. Refined calculations that would take into consideration the restraint due to mounting the plate in the element, the element bowing, etc., would almost certainly have given higher stress levels. A meaningful elastic-plastic analysis of the fuel element can only be performed by a code of the type used by NUS in their early study of the AARR fuel elements (PLASTIC-SASS). At the time of termination of activity, a review of this code was underway, and the tentative conclusion was that, in its present form, it is not capable of treating accurately the extreme stress conditions likely in the AARR plates.

A study of the transient thermal stresses in heat-generating flat plates caused by sudden changes in coolant temperature was being completed at the time of termination. Preliminary numerical results indicated that the portion of the stress field caused by internal heat generation was of minor importance, at least for preliminary design

computations; existing solutions relating to the quenching of flat plates would be sufficient. Internal heat generation produces a slight increase in the peak stress magnitudes and increases the length of time at which the plates are at a given stress level; however, both of these effects are relatively minor for the range of parameters relating to conditions in AARR.

An additional thermal-stress problem that received considerable attention concerned the stresses caused by abrupt axial changes in internal rates of heat generation. This problem related to cylindrical fuel pins and was not directly applicable to the AARR fuel elements; however, the qualitative results of this study should also hold for flat plates.

The results for the cylindrical case indicated that abrupt axial changes in heat-generation rate do not produce significantly higher stress levels than those computed on the basis of a uniform heat generation of a magnitude equal to the maximum occurring level. The one exception would be in the shear stresses which occur in the region of the change in generation rate. These shear stresses, however, are quite localized and are of a smaller magnitude than the other stress components.

(ii) Fuel-assembly Analyses. The mathematical and experimental modeling of interacting beam-type structures simulating fuel assemblies mounted to a support grid were studied. The initial models consisted of beams cantilevered from a support beam or clamping fixture. The objective of the study was to provide answers to the questions:

- a) How many modes are required for an adequate dynamic analysis?
- b) How many elements are required to display the characteristic interaction aspects of a core bundle?

A consideration of second-order effects,⁴¹ removed by the linearization process in similar analyses of previous investigations, led to an additional stability criterion in the form of an upper bound on the amplitude of deflections for stable oscillations, or neutral stability, about the undeflected position of the fuel plate. A result of this study is the definition of new critical flow velocities for reactor parallel-plate assemblies, in which anticipated fuel-plate deflection is a parameter.

Thermally induced deflection of fuel plates, one of the many mechanisms leading to channel closure, was studied both analytically and experimentally. Theoretical curves were plotted describing the lateral deflection of the AARR fuel plate, with and without center spacers, for various uniform temperature increases in the plate only. Experimental

⁴¹Wambsganss, M. W., Second-order Effects as Related to Critical Flow Velocities and Reactor Parallel-plate Fuel Assemblies, ANL-7261.

tests were conducted to gain an understanding of the deformation of a flat fuel plate resulting from differential heating within a fuel subassembly. A plate specimen with a restrained length corresponding to the width of the AARR fuel plate was tested. Results showed fair agreement with theory. Discrepancies can be attributed, in part, to the fact that the analytical expression assumes perfectly flat plates.

The AARR control blades are intimately related to the fuel assemblies in that the control-blade channel is formed by two adjacent fuel assemblies. The following analyses related to the control blades were performed:

- a) stability analysis of a flat blade subjected to a suddenly applied longitudinal force (as during SCRAM);
- b) fluid dynamic modeling of the hydraulic loading exerted on the blade;
- c) mathematical modeling of the piston-type damper.

(iii) Fabrication Techniques for FTS-4D Dummy Fuel Element.

The purpose of this effort was to fabricate a dummy fuel element. This element had one outside plate having a greater coefficient of expansion (α) than the balance of the element, which would simulate, at a test temperature, a fuel plate operating as a hot plate in the fuel assembly.

In order for the test to be somewhat meaningful the room-temperature or base stress of the dissimilar metal plates should be as stress-free as possible. Since both brazing and electron beam (EB) welding require elevated temperatures, the attainment of a stress-free condition is difficult if not impossible. The use of epoxy adhesives to attain this joint was investigated and may prove to be an acceptable method of assembly.

At the time of cutoff, only development of various methods of fabrication of test plates for the assembly and of several assembly techniques had been evaluated. They are discussed below.

Fabrication of Plates:

Machining. The first approach was to machine half-spacer wires integral with the simulated plates. In these plates the center half-height spacer wires were left after milling the plates on both sides. The surface finish on these plates left something to be desired, and the method was time consuming. Several test plates for EB welding were fabricated by this method.

Electrical Discharge Grinding. As an alternate to the above method the use of electrical-discharge grinding was considered as a stock removal method. Several test plates were made by this method. The surface finish on the plate was ~160-175 micro-inch, with good dimensional control and a fairly sharp corner between the rib base and plate surface. Additional effort would be required to extrapolate this method to full-size fuel plates.

Chemical Milling. Ten attempts were made by the Chemical Micro Milling Co. to mill chemically $\sim 1\frac{1}{2} \times 4$ -in. areas to a depth of 0.020 in. in 14 gauge, Type 304 stainless steel test plates. After milling 0.020 x 0.040-in. ribs remained in the center. No test plate was milled to exact size. Normal procedures do not permit milling to a depth of 0.020-in. in stainless steel. Therefore the process evolved followed a 2-step procedure:

Step 1: Using normal procedures, etch until the photo-resist breakdown is imminent. Stop etching.

Step 2: Neutralize, and recoat the land surfaces with an asphaltum resist and re-etch.

During the development it was determined that the normal drafting compensations built into the master were not adequate for a one-side (not through) etch 0.020 in. deep. A second master must be drawn to etch to specification dimensions.

Inspection of five of the Chem-milled samples indicated a generous corner radius and a surface finish on the etched surface of the plates of 200-250 micro-inches. This is considerably rougher than the 125-micro-inch specified on the drawings. The widths of the rib lands (0.040 in. nominal) were from 0.053 to 0.075 in. The depth (0.020 in. nominal) varied from -0.010 to +0.002 in. The rib off-set did not meet drawing-dimension register, and the corner radii varied from 0.010 to 0.020 in. The width of the nominal 1.320-in. cavity had a variation from -0.010 to -0.033 in.

The above method is not considered of sufficient merit to warrant further research and development for this application, though if it were the "only" way, the process could be developed.

Assembly:

Electro Beam Welding. Several test plates were machined of Types 430 and 304 stainless steel. Attempts to weld these were, in general, unsuccessful. The first attempt lacked weld penetration. Upon revising the power setting and rewelding, a full penetration weld was obtained; however, the weld shrinkage resulted in a "toe-in" condition.

of $\sim 1/32$ in. of the plate legs on the open side. A third attempt with a lower power setting and welding from both sides resulted in a satisfactory weld, but the weld shrinkage again resulted in a toe-in of the side legs of ~ 0.020 in. Because of the narrow (0.040 in.) channel gap and tight tolerance (± 0.001 in.) required, this method does not appear promising as an assembly procedure.

Bonding. Consideration was given to the use of epoxy as a bonding medium for attachment. Several epoxy mixtures were formulated and tested according to the given specifications to withstand a temperature of 400°F in a water environment. Pieces of 0.109-in.-thick strips of Types 430 and 304 stainless steel were furnished for the evaluation.

Proposed Bonding Method. The process proposed is as follows:

Clean the stainless steel by sandblasting with an S. S. white air abrasive unit; then wipe the areas to be bonded with acetone. Bond pieces using the following formulation:

100 g EP201 (Union Carbide resin);

28 g PMDA (DuPonts Pyromellitic dianhydride).

The PMDA should be ground and sifted through a 280 mesh screen before using. Mix components well. PMDA will not go into solution, but will remain as a powder until heated; therefore it is important that it be well dispersed in the mixture.

Coat pieces to be bonded and clamp together. Place clamped assembly into an oven and raise the temperature 15°F per hour to 400°F. Hold for 20 hr and slowly cool the assembly to room temperature. Unclamp the assembly.

When the test assembly was heated to 428°F in an oven, the sample was bowed $\sim 3/32$ in. When the temperature was raised beyond this point, the bond broke, i.e., the pieces came apart.

The above test was in a dry oven. No information on the moisture resistance of the bond material was developed because of the termination of this phase of the AARR project effort.

(iv) Fabrication of Subassembly. In preparation for the fabrication of the element to methods developed above, Types 304 and 430 stainless steel sheet and strip stock material were purchased. From the above stock, 43 dummy fuel plates were rolled to 0.040-in. thickness. Thirty-three of these plates were machined to rough width and length, preparatory to spot welding of the spacer wires. A micatta fixture and copper electrodes were made for spot welding the spacer wires to the plates. All activity has been terminated.

c. Fabrication of Eu_2O_3 and Hafnium Control and Safety Rods for Test Purposes. Two types of control rods were proposed for the AARR core. The inner rods were to have a poison core composed of TiO_2 -stabilized Eu_2O_3 dispersed in stainless steel and clad with 347 stainless steel. This rod would have a Type 347 stainless steel follower. The outer rods were to have a poison section composed of hafnium attached to a Zircaloy-2 follower.

The design of rods for insertion in the hydraulic loop is different from the prototype rod design because of dimensions and limitations of the loop test section. Also the design had to account for modification of the Eu_2O_3 poison section for the critical experiments.

Two sheets of hafnium (0.200 in. thick x 5.5 in. wide x 22 in. long) and one sheet of Zircaloy-2 (0.200 in. thick x 6.5 in. wide x 55 in. long) were on hand for fabrication of the test control rods. Physical properties and chemical analysis of the above plates are available.

The upper handling fitting has been finish-machined except for drilling the screw holes and was ready for chrome-plating. The follower material was rolled to 0.200-in. thickness and has been finish-machined except for drilling of the screw holes. The poison section was machined preparatory to electron-beam welding on an extension piece to fill out the width.

Test pieces of hafnium have been electron-beam welded and parameters for an acceptable weld have been established. The two hafnium pieces had been machined preparatory to electron-beam welding.

All further effort on the fabrication of the test control rods has been terminated.

d. Support-grid Analyses. The slots in the prototype support grid, which accommodates the control blades, were investigated for changes in width upon load application to the grid plate. Extrapolated results indicate that the slot could be expected to narrow down by less than 0.005 in. with a 125-psi pressure drop across the core.

Treatment of the combined hydraulic and thermal-stress problem as a problem in optimization was considered.

The dynamic response (natural frequencies and mode shapes) of hexagonal and circular plates, supported at three points in a manner similar to the way in which the AARR support grid is supported, was studied. The tests were conducted for the vibration exciter. The objectives of the tests were to relate characteristics of hexagonal and circular shapes, and from the mode shapes to obtain insight into the type of deflection patterns that might be expected with the AARR grid plate.

3. Reactor Physics

a. Experiments. The program of reactor physics experiments for the AARR design has been terminated. Results of some of the most recent critical experiments on SS-U²³⁵ loadings are reported. Some additional data will be published in future monthly reports after there has been an opportunity to review and to analyze completed experiments.

The core was reloaded to simulate the physics design reference graded-fuel loading described earlier (see Progress Report for December 1965, ANL-7132, p. 66). Grading of fuel in the vicinity of the inner and outer radial core boundaries is required, in a practical reactor design, to reduce the peaks in power density which result from the steep gradients of thermal-neutron flux near the edges of a heavily absorbing core with highly moderating reflectors. In the Criticality Facility, this was achieved by the use of special, fuel foils of thin uranium metal. The first eight composite fuel plates, at the interface between internal thermal column (ITC) and core, increased in foil thickness from plate to plate. Starting at this boundary, the ratios of foil thickness to the thickness (4.4 mils) of a "standard" fuel foil were: 0.22, 0.28, 0.37, 0.47, 0.60, 0.75, 0.91, and 1.0. The total thickness of this graded-fuel region is 0.56 in. Starting at the outer radial core boundary, the relative thicknesses of the first 4 fuel foils were: 0.42, 0.57, 0.78, and 1.0.

The ungraded core loading which was revised to include fuel grading has been described previously (see Progress Report for October 1966, ANL-7267, p. 72). The ITC is only slightly larger than the design reference ITC. As a result of the fuel grading, the total fuel content was reduced from 63.7 to 61.5 kg of 93%-enriched uranium.

To compensate for the reactivity loss due to fuel grading, a total of 258 boron stainless steel poison strips were removed. The final loading included: 1089 poison strips, with a total mass of ~572 g natural boron; 1086 standard fuel foils (4.4 mils thick); and 87 thin fuel foils. From preliminary data of control-blade calibrations it is estimated that the net available excess reactivity of this system was $4\frac{1}{4}\%$. Criticality was achieved with three alternate in-core control blades fully inserted, and a fourth in-core blade partially inserted.

The fuel grading was introduced only in fuel assemblies with plates parallel to only one face of the core. It may be seen in Fig. 19 that there are 3 corner fuel assemblies at the ITC-core interface, and 3 at the beryllium-core interface, with ungraded fuel.

Figure 19 is a plan view showing the radial locations of U²³⁵-Al detector foils with which radial traverses of U²³⁵-fission activity were obtained. The traverse in the direction of the fuel grading appears to have a more gradual slope than the traverse into an ungraded, corner fuel assembly near the ITC (see Fig. 20). The traverses give the relative

fission activity per atom of U^{235} , and it therefore appears that the reduced neutron blackness of the graded-fuel zone is reflected in a less steep gradient of thermal and near-thermal neutron flux. This result certainly is in qualitative agreement with theory, and, indeed, it agrees with physical intuition. A radial traverse of U^{235} -fission activity for the corresponding ungraded fuel loading of 1173 standard foils and 1347 poison strips was included in ANL-7267 (Fig. 16, p. 72). Note that the peak fluxes in the ITC and in the beryllium per unit reactor power are little affected by the fuel grading, as predicted by earlier theoretical analysis (see Progress Report for March 1966, ANL-7193, p. 84).

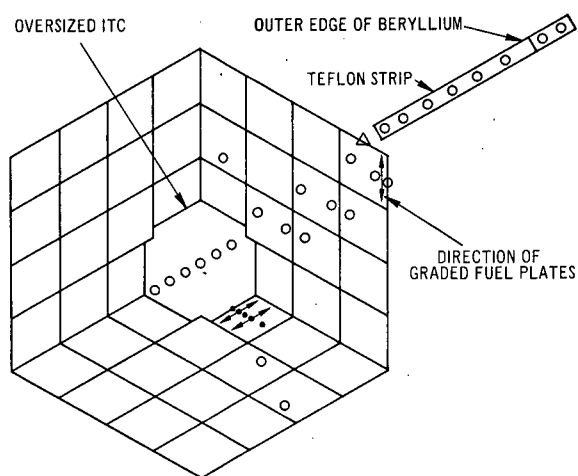


Fig. 19. Radial Location of Bare Foils for Radial U^{235} -fission Activation Traverse (Aluminum ITC Shroud)

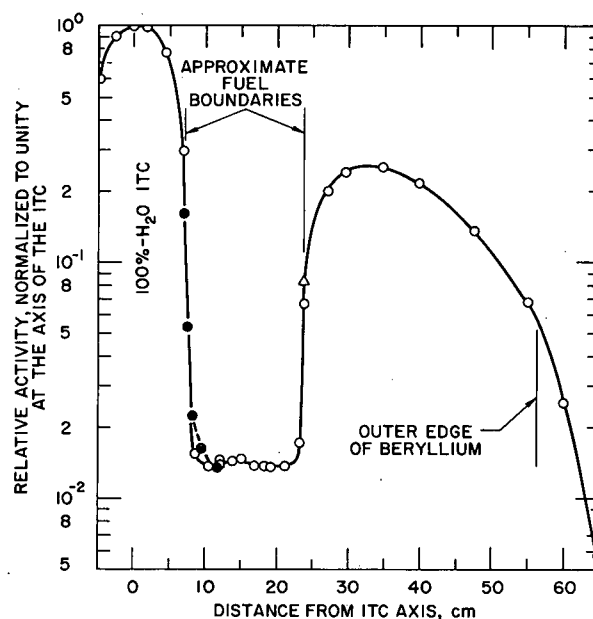


Fig. 20. Radial Activation Traverse of U^{235} -fission of Bare Foils for the 1173/1089 Graded Loading and the Oversized ITC (Aluminum ITC Shroud)

b. Reactor Physics Analysis. With the use of only the reactivity-feedback effects of thermal expansion of the HFIR-type fuel plates, the response of the AARR to various reactivity insertions has been studied. In the October 1966 Monthly Progress Report, ANL-7267, p. 72 two models of fuel-plate expansion were discussed. The preferred model (A) is based upon a full volumetric expansion of insulated fuel plates, with a constant total volume of the reactor fuel zone. Model B assumes that unrestrained axial expansion occurs for that one-third of the volumetric expansion, but that expansions in thickness and in width occur in a fuel zone of constant area.

Preliminary numerical results have been obtained for Model A with an analog computing machine. Calculations were made for feedback Model A, and for total ramp reactivity insertions of from \$1 to \$7 in time

periods of 30, 10, and 0 (step) msec. Some of the results are summarized in Table XIX. All calculations are based upon an initial power level of 100 MW.

TABLE XIX. Summary of Reactor Responses to Reactivity Transients,
Assuming Only Fuel-plate-expansion Feedback of Reactivity
(Model A: Fixed Reactor Core Volume)

Total Reactivity Added (\$)	4			5			6			7		
Total Time for Ramp Reactivity Insertion, msec	0	10	30	0	10	30	0	10	30	0	10	30
Maximum Percentage Expansion of Average Fuel Plate	19	19	16	25	24	19	29	28	21	34	33	32
Total Incremental Energy Release, MW-sec	76	76	65	97	95	77	115	113	83	135	132	90
Ratio of Peak Power to Initial Power of 100 MW	128	126	68	225	210	90	335	315	113	370	370	140

As may be seen from Table XIX, even for so rapid a reactivity insertion as a 10-msec ramp, for insertions of \$4-7 the total incremental nuclear energy release to the time of reactor shutdown is almost the same for the step and for the 10-msec ramp. For a step reactivity insertion of \$7, the total incremental energy release to the time of shutdown is only 135 MW-sec for Model A.

An anticipated, and realized, result of the analysis is that the nuclear excursion is terminated before the major portion of the fuel-plate expansion occurs. It is expected that the occurrence of a rapid massive metal-water reaction would require fuel-plate temperatures in the vicinity of the boiling point of aluminum, roughly 3600°C. For the hypothetical case of a reactivity step of \$7, the peak averaged fuel plate temperature during the transient is 1700°C. With such a rapid transient, the temperature at the cladding surface of the assumed insulated plate would rise much less rapidly than the temperature at the center of the plate. Thus, even in the "hot" channels it is doubtful that there would be much, if any, metal-water reaction during the reactivity transient. Any such metal-water reaction would proceed relatively slowly.

4. Thermal Analysis of Beam Tube

The design of the beam-tube adapter has been revised to eliminate the extension piece at the shield face. The calculations for the through-tube deflection (see Progress Report for September 1966, ANL-7255, p. 76) have been extended to consider this change. Results are shown in Table XX

(which is a continuation of Table XX, ANL-7255, p. 78). A comparison with the earlier results shows that the effect is to produce slightly more deflection of the through-tube for "case I," in which adapter is fixed and supported near its outer end. The deflection is reduced when the adapter is cantilevered from the vessel ("case II") and for the more complex "case III" (see ANL-7255). These deflections can be easily accommodated in the clearance between the through tube and the reflector. All of these results are based on 0.050-in. vertical movement of the through tube nozzle due to thermal growth of the vessel after the adapters have been set in place.

TABLE XX. Summary of Calculations for Through-tube Deflection

Case	Vessel Condition	Deflection ^a at Tube Supports (in.)		Rotation ^a at Tube Supports (10 ⁻⁴ Radian)		Deflection ^a (in.) from Centerline at Positions		
		Floating End	Fixed End	Floating End	Fixed End	A	B	C
I	Cold	-0.001	-0.0003	-0.20	-0.37	-0002	+0002	+0004
	Hot	-0.026	-0.0023	+7.39	-3.60	+0142	+0148	+0129
II	b	-0.033	-0.0023	+9.45	-3.60	+0154	+0157	+0131
III	Cold	-0.033	-0.0023	+9.45	-3.60	+0154	+0157	+0131
	Hot	-0.058	-0.0043	+17.03	-6.77	+0291	+0294	+0246

^aDeflections (measured from nozzle centerline) are positive upwards; clockwise rotations are negative.

^bLoading independent of temperature.

5. Analysis of Flow Coastdown

In the event of a total loss of pumping power, the primary-system flow will coast down rapidly to that supplied by the Shutdown Emergency Cooling System, which is composed of a d.c. series, battery-operated, "pony" motor mounted on the shafts of each main pump-motor. Studies show that the important parameters determining the rate of flow coastdown are the rated motor torque and speed, and the total "shaft" rotary inertia. Water inertia normally has a very small effect upon coastdown rate. The rate of coastdown is not affected by the pony motors during the initial stage of the transient, i.e., up to about 2.5 sec. Thereafter, however, the difference becomes great because the flow goes to zero without pony motors.

An analog model was derived and used to solve a set of four nonlinear differential and algebraic equations describing the coastdown dynamics. An additional equation was used when coastdown dynamics included pony motors. The model corresponds to the primary-system layout, loop losses, pump-motor sizes, and rated flow (19,000 gpm) as verified by discussions with Burns and Roe. Each pony motor in the model was sized to yield 10% of rated flow (1900 gpm) when operating singly. When both pony motors are operating the total flow has been calculated to be about 2500 gpm. Pump speed and flow following simultaneous tripout of two main pump-motors were calculated for the following three cases:

- (a) tripout with two pony motors energized;
- (b) tripout with one pony motor energized;
- (c) tripout with no pony motor energized;

Results for Case (a) show that flow is down to 50% of rated in 1.8 sec, to 20% in 7.0 sec, and that the final value of 13% flow (2 pony motors) is reached at about 20 sec. The next step planned in this study is to determine the thermal response of the core to flow coastdown under a variety of conditions with the analog computer.

The model discussed above was also used to make an analytical versus experimental comparison of coastdown for the General Purpose Hydraulics Loop (GPHL). Pump speed and flow response to motor tripout were recorded starting from a series of 4 initial steady-state flow conditions. The measured data were highly reproducible and showed, for example, that flow was down to 50% of initial value in 1.35 sec when starting from 1000 gpm. By means of the pump and motor data, obtained earlier from the manufacturer, and the calculated loop parameters, the analog model showed a 50% flow reduction in 1.41 sec. This calculated versus measured time differential of about 5% is consistent for all the runs (1200, 1000, 800, and 600 gpm), indicating that the manufacturer's estimate of the total rotary inertia was apparently about 5% too high. This analysis again showed that the water inertia had little effect upon the coastdown dynamics.

6. Safety

a. Vessel

(i) Calculations of Vessel Stress. Detailed calculation of the stresses has been completed for the most recent vessel configuration, in which the vessel is made of Inconel-600, and the main shell wall is $3\frac{1}{2}$ in. thick. The stresses were computed by the computer code Seal-Shell-2.⁴² The output stresses are:

- (a) the combination of all primary and secondary (mechanical stresses only) stresses;
- (b) the combination of all primary and secondary (mechanical and thermal stresses) stresses;
- (c) the combination of all primary, secondary (mechanical and thermal stresses) and peak (thermal stresses only) stresses.

The stress profiles are in the process of preparation.

⁴²Friedrich, C. M., Seal-Shell-2, A Computer Program for the Stress Analysis of a Thick Shell of Revolution with Axisymmetric Pressures, Temperatures, and Distributed Loads, WAPD-TM-398 (Dec 1963).

(ii) Vessel Containment. The potential for vessel containment has been estimated. In the analysis, the following assumptions were made:

(a) The stress and strain are related by the power-law strain hardening equation

$$\sigma = 132,000 \epsilon^{.5}.$$

(b) The deformation of the vessel for a given radial strain is obtained from the equation

$$v = 1.44 V_0 \frac{(R-R_i)}{R_i}$$

in which

v = incremental volume;

V_0 = undeformed volume;

R = deformed radius;

R_i = undeformed radius.

For a minimum tensile strength of 90,500 psi and a minimum ultimate strain of 0.45 in., the strain energy of the Inconel vessel at the ultimate strain is about 380 MW-sec and that for 1/3 of the ultimate strain is about 79 MW-sec.

The use of 1/3 the ultimate strain has been recommended for the safe design of reactor pressure vessel to accommodate "TNT equivalent detonations."⁴³ But, in nuclear excursions, the reactions will be much slower than that for TNT detonation. Furthermore, in the AARR vessel, the belt section of the shell, the nozzles, and piping are adequately reinforced. Premature failure at these locations is not to be expected. It is believed that the vessel will undergo a gross ductile deformation before rupture, and the marginal strain will be much greater than 1/3 the ultimate strain.

b. Secondary Containment. Calculations were completed for the damage that might occur to the AARR containment from outside missiles. The results showed that containment will not be lost. Although the formulas were used for outside missiles, they are equally applicable to missiles generated internally. Hence, they should be useful in any reactor hazard study that involves the effect of missiles on containment.

⁴³Wise, Jr., W. R., and Proctor, J. F., Explosion Containment Laws for Nuclear Reactor Vessels, NOLTR-63-140 (Aug 1965).

V. NUCLEAR SAFETY

A. Coolant Dynamics

1. Superheat Experiments

The system has been filled with sodium and liquid superheat tests have begun. Test runs to date involve a pressure range from 4 to 7 psia, heat fluxes ranging to approximately 2×10^4 Btu/hr-ft², liquid superheat required to initiate boiling ranging from 70 to 170°F, steady-state superheats ranging from 20 to 40°F, and transient pressure pulses ranging up to 60 psia. Temperatures of liquid sodium ranging up to approximately 1525°F have been obtained.

Figure 21 shows the results of a typical test run. The lower curve shows the temperature-time history, including the natural-convection region, the initiation of boiling, and steady-state boiling region. The upper curve shows the pressure-time history of the system, and shows a relatively small pressure pulse (from 2 to 10 psia) at the initiation of boiling and larger pressure pulses (ranging up to 60 psia) during steady-state boiling.

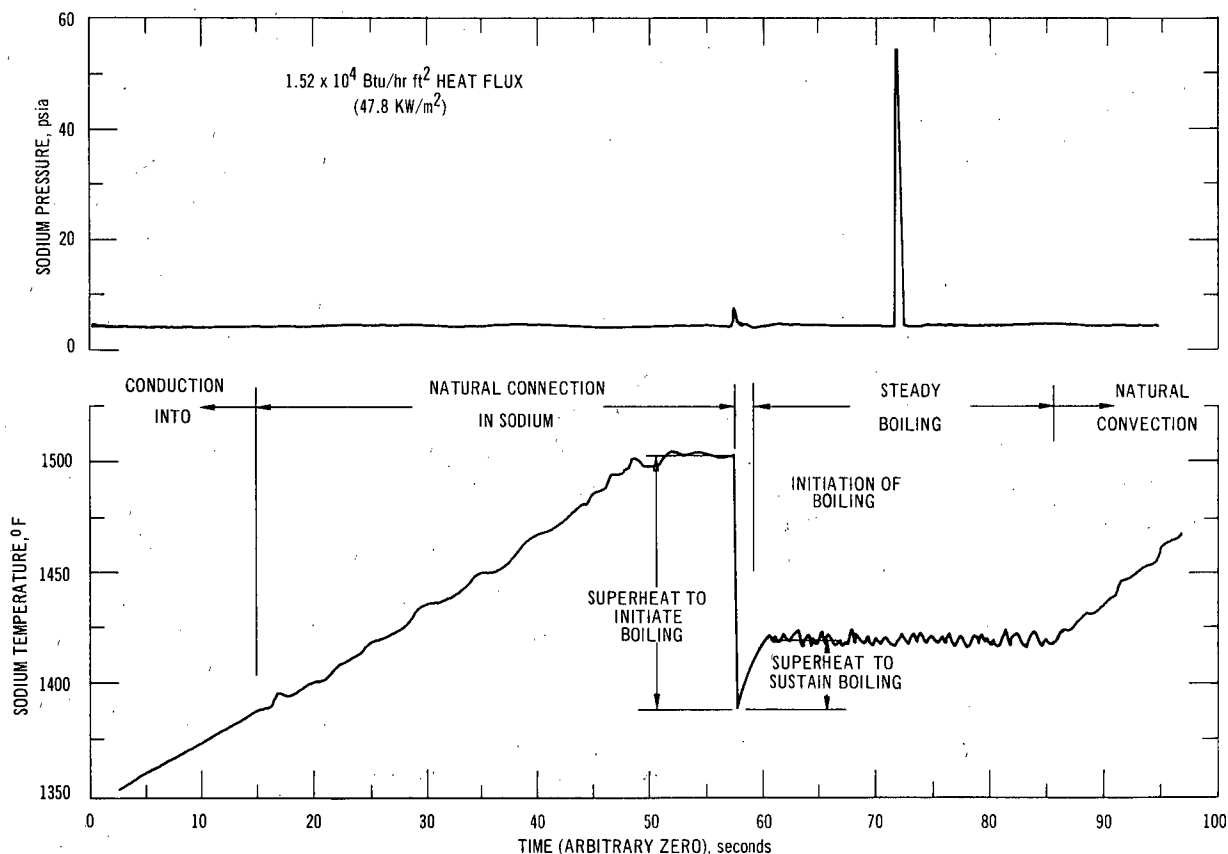


Fig. 21. Temperature and Pressure Transients during Typical Run

An additional heater is being installed in the system to permit extension of the heat-flux range beyond 2×10^4 Btu/hr-ft².

2. Sodium Expulsion Experiment

Fabrication of the facility is continuing. An order for the programmed electron-beam heater has been placed.

3. Studies of Critical Flow

The construction of equipment to study critical flow of different fluids is continuing. The major portion of the sodium loop has been completed. The first test section is being installed in the loop, and shakedown tests of the instrumentation for measuring pressures, temperatures, and flowrate have been completed.

B. Fuel Meltdown Studies with TREAT

1. Post-transient Inspection of Mark-I Integral Sodium Loop No. 3

Transient experiments 1049 and 1050 on the second 7-pin cluster of EBR-II pins (see Progress Report for September 1966, ANL-7255, p. 83) were run in the TREAT facility using Mark-I integral sodium loop No. 3. Transient oscillograph records for run 1050 revealed that pressures over 15 atm were reached and that high temperature rises occurred in the bulk sodium of the loop (approx. 1260°C by the inlet thermocouple, and 880°C by the outlet thermocouple). The above results, in conjunction with the failure of the EM pump to operate during the process of removing the 7-pin fuel cluster after transient experiments, prompted the removal of the loop from its containment can for inspection.

The sealed container can showed no evidence of failure. However, upon removing the loop from its secondary containment housing, it was observed that the loop proper had been breached. Analyses of samples of the sodium from the breached loop showed active loose contamination.

Inspection of the electromagnetic pump tube and base-pedestal regions showed the presence of sodium. The deposits of sodium above the breached area as well as on the inner wall of the loop containment can indicated a forceful ejection of sodium, apparently of a short duration, because only small amounts of sodium (approx. one cc) were found in the sprayed areas. Most of the sodium had escaped from the loop. The base pedestal served as a good heat sink for the sodium and the pedestal silastic rubber, O-ring seal remained intact.

After removal of the EM pump coils and the associated transformer, the pump flow tube was mechanically cleaned of sodium, exposing a crack in the upper round-to-flat flow-tube transition. The jagged crack extended about equally into both faces of the transition, occurring across the edge of the accommodating conic relief in the abutting coil face. Swelling of the flow tube, which was principally along the greater width (the coils and pole pieces provided restraint normal to the flattened sides), amounted to about 3 mm.

A probing inspection of the opening in the pump tube did not show any signs of fuel-steel alloying. Samples for contamination analysis did not show alpha activity above 200 dpm.

The diaphragm to the loop safety tank was intact, but showed signs of some pressure-forming. Rupture pressure of the diaphragm is 30 atm.

Inspection of the sample remains from both clusters is underway.

C. Materials Behavior, Equation of State, and Energy Transfer

1. Transient Heat-transfer Studies in Liquid Sodium

Experimental studies of the transient heat transfer from small hot particles moving through liquid coolant continue. Knowledge of the rate and quantity of energy transfer that occurs between the fuel particles and the coolant is necessary in order to analyze the consequences of a reactor incident in which hot fuel materials may be dispersed into a liquid coolant (water or sodium). This study was initiated because neither theoretical nor experimental information was available to allow calculation of the heat transfer that occurs in such a situation.

Experimental studies of the heat transfer between 1/4-in.-dia meter nickel spheres and water were carried out by the swinging-arm method (see Progress Report for July 1966, ANL-7245, p. 66). Similar experiments have recently been performed with 1/2-in.-dia meter tantalum spheres in liquid sodium.

a. Experimental Method. The basic apparatus for the heat-flux studies in liquid sodium consists of a swinging-arm (or pendulum) with a 1/2-in.-dia meter tantalum sphere attached to the end of the arm. Tantalum was selected as the sphere material because of its high melting point and the ease with which it can be machined. The sphere is supported by a tantalum support tube attached to the shaft of a printed-circuit d.c. motor. The sphere is heated inductively by a 3-turn coil. The angular velocity of the sphere assembly is measured with a photocell timing unit at the rear of the motor. The sodium is contained in a stainless steel tank which is located directly below the sphere assembly. The entire assembly is housed in an argon-atmosphere glovebox.

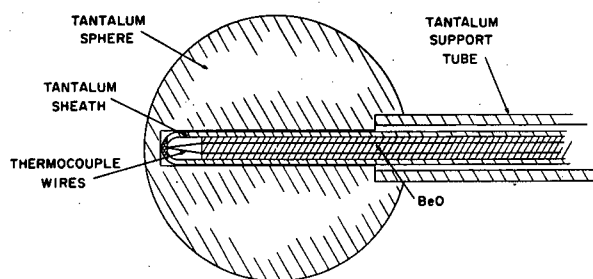


Fig. 22. Cross Section of Tantalum Sphere

A cross section of the tantalum sphere is shown in Fig. 22. A W-5% Re/W-26% Re thermocouple is welded into the sphere at a point just under the leading surface.

In an experiment, the heated sphere is moved from the furnace through the sodium at a preset velocity, backward to a holding position at low velocity, and again into the furnace position. A total of 87 experimental runs have been made. Sodium temperatures of 572 and 842°F and sphere velocities of 6.0 and 10.0 ft/sec were used. Initial sphere temperatures ranged up to 3600°F (1980°C).

The data collected for each experiment consist of a temperature-versus-time measurement at the radial position corresponding to the location of the thermocouple junction in Fig. 22. A typical temperature-time trace is shown in Fig. 23. From such measurements for each experimental run, the instantaneous temperature of the sphere surface and the heat flux were calculated using the method of Stolz.⁴⁴

b. Experimental Results. Values of instantaneous heat flux and corresponding values of surface temperature were calculated for each experimental run. Results for a sphere velocity of 10.0 ft/sec and a sodium temperature of 572°F are given in Fig. 24. Heat-transfer rates up to 36×10^6 Btu/(hr)(sq ft) were calculated from the data. Experimental results could be correlated by the following empirical expression:

$$q/A = C(T_w - T_b)^{0.88}, \quad (1)$$

where

q/A = heat flux, Btu/(hr)(sq ft);

T_w = surface temperature of sphere, °F;

T_b = sodium bulk temperature, °F;

$C = 2.98 \times 10^4$ [Btu/(hr)(sq ft)(°F)^{0.88}] for velocity = 10 ft/sec, $T_b = 572^\circ\text{F}$;

2.23×10^4 [Btu/(hr)(sq ft)(°F)^{0.88}] for velocity = 6 ft/sec, $T_b = 572^\circ\text{F}$;

2.34×10^4 [Btu/(hr)(sq ft)(°F)^{0.88}] for velocity = 10 ft/sec, $T_b = 842^\circ\text{F}$.

⁴⁴ Stolz, G., Jr., Numerical Solutions to an Inverse Problem of Heat Conduction for Simple Shapes, Trans. ASME, Series C, 82, 20 (1960).

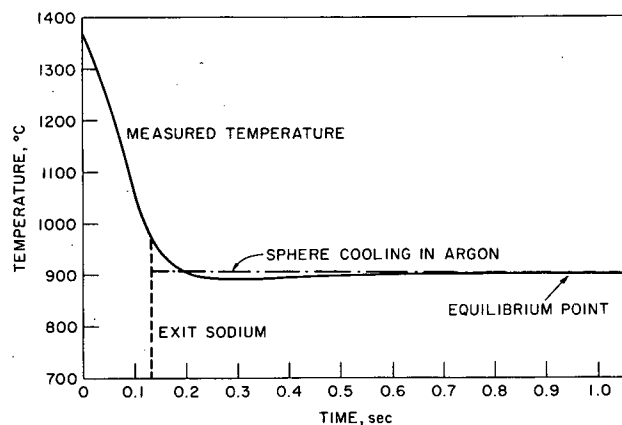


Fig. 23. Temperature-Time Trace for 1/2-in. Tantalum Sphere Moving through Sodium

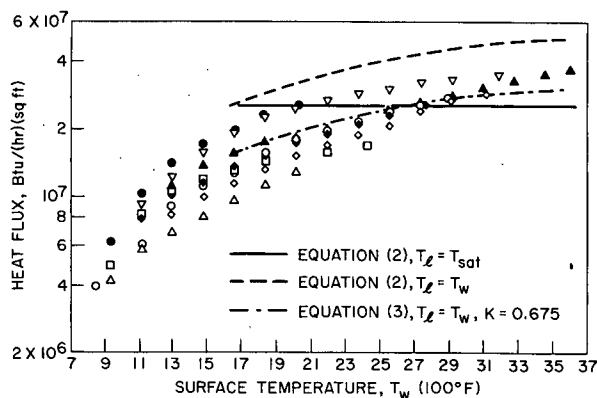


Fig. 24. Heat Flux vs. Surface Temperature for 1/2-in.-dia Sphere Moving through Liquid Sodium. ($U_{\infty} = 10.0$ ft/sec; $T_{Na} = 572^{\circ}\text{F}$)

A check on the method of calculating instantaneous heat flux was employed in which the total heat loss from the sphere during the period of immersion in the sodium was calculated. The total heat loss computed from a summation of instantaneous values was compared with the total heat loss from the sphere determined from the difference in sphere temperature just before immersion and that after emergence from the sodium. A correction was included for the small heat loss rate to the argon environment after emergence of the sphere. Values of total heat loss computed by the two methods generally agreed within 10%.

c. Theoretical Studies. Although theoretical studies of film boiling as related to heat transfer are being made, a simplified theory based upon potential flow has been used to study the experimental results. The assumption has been made that surrounding the hot sphere there is a vapor film, which is thin in comparison with the radius of the sphere. Liquid velocity at the liquid-vapor interface was assumed to be unaffected by vapor drag and was therefore described by the laws of potential flow. It was also assumed that the sphere has a uniform surface temperature. (This assumption may be removed in a more refined treatment.) It was shown that thermal radiation could be neglected.

The development of the equations was based on an energy balance for a small element of vapor adjacent to the sphere surface. It was found that the heat used to form vapor in this element was negligible in comparison to the heat transferred from the element of vapor into the bulk sodium. The development of the equation describing the heat transferred from the liquid-vapor interface into the bulk sodium followed the general treatment outlined by Sideman.⁴⁵ The resulting equation is

$$q/A = 1.13 \left[\frac{U_{\infty} \rho_{\ell} (C_p)_{\ell} k_{\ell}}{D} \right]^{1/2} (T_{\ell} - T_b), \quad (2)$$

⁴⁵ Sideman, S., The Equivalence of Potential Flow and Penetration Theory, Ind. Eng. Chem. **58**(2), 55 (1966).

where

- U_{∞} = sphere velocity;
- ρ_{ℓ} = sodium density (liquid);
- $(C_p)_{\ell}$ = specific heat of sodium (liquid);
- k_{ℓ} = thermal conductivity of sodium (liquid);
- D = sphere diameter;
- T_{ℓ} = temperature of sodium at interface;
- T_b = temperature of bulk sodium.

Calculations of heat fluxes based upon Eq. (2) are compared with experimental results in Fig. 24. If it is assumed that the sodium boils at the saturation temperature, 1620°F (883°C) at 1 atm pressure, without superheating, then $T_{\ell} = T_{\text{sat}}$ and the straight line in Fig. 24 is obtained. It is apparent from this figure that the experimental data show an increasing heat flux with increasing surface temperature. If it is assumed that the liquid-vapor interface temperature, T_{ℓ} , is the temperature of the surface of the sphere, T_w , i.e., complete superheating occurs, then the upper curve in Fig. 24 is obtained. A good fit to the data is obtained by using a constant fraction of the value calculated from Eq. (2) while substituting $T_{\ell} = T_w$ as follows:

$$q/A = K \left[\frac{U_{\infty} \rho_{\ell} (C_p)_{\ell} k_{\ell}}{D} \right]^{1/2} (T_w - T_b). \quad (3)$$

The heat fluxes calculated by using a value of 0.675 for K are represented by the lower curve in Fig. 24.

The thickness of the vapor film required to transfer these large quantities of heat is extremely small. For example, the thickness of the vapor film at the 90-degree position for a 1/2-in.-dia meter sphere at 3472°F moving at 10.0 ft/sec was calculated to be 7.65×10^{-6} in. Although the sphere surfaces were relatively smooth, it is believed that the magnitude of the roughness of the surface greatly exceeded this value. It is possible that protuberances on the sphere surface could extend through the vapor film into the liquid sodium. This would cause the film to become unstable and possibly to collapse completely. This collapse would bring liquid sodium into contact with the sphere surface which is at a high temperature. The sodium would then tend to assume the temperature of the surface of the sphere and could become highly superheated.

d. Conclusions. The agreement between experimental data and the theory that was developed by assuming superheating of liquid sodium

indicates that this theory is adequate to describe the actual physical phenomenon. The physically unrealistic vapor-film thicknesses that were calculated also tend to support the theory of superheating.

The results of this study indicate that large quantities of energy can be transferred from extremely hot spherical particles to highly sub-cooled liquid sodium without vapor formation.

Both experimental and theoretical investigations of heat transfer from moving spheres into a coolant are continuing.

2. Studies of High-temperature Physical Properties

a. Measuring Circuit for Liquid Level. A necessary requirement for the analysis of severe accidents involving reactor core materials is a knowledge of their high-temperature physical properties. Measurement of the thermal expansion of molten uranium carbide and material compatibility studies have been undertaken in the temperature range from 2300 to 3000°C. Figure 25 shows a schematic diagram of a modified GE high-temperature furnace used in these investigations. The furnace is pressurized with helium to slightly over one atmosphere.

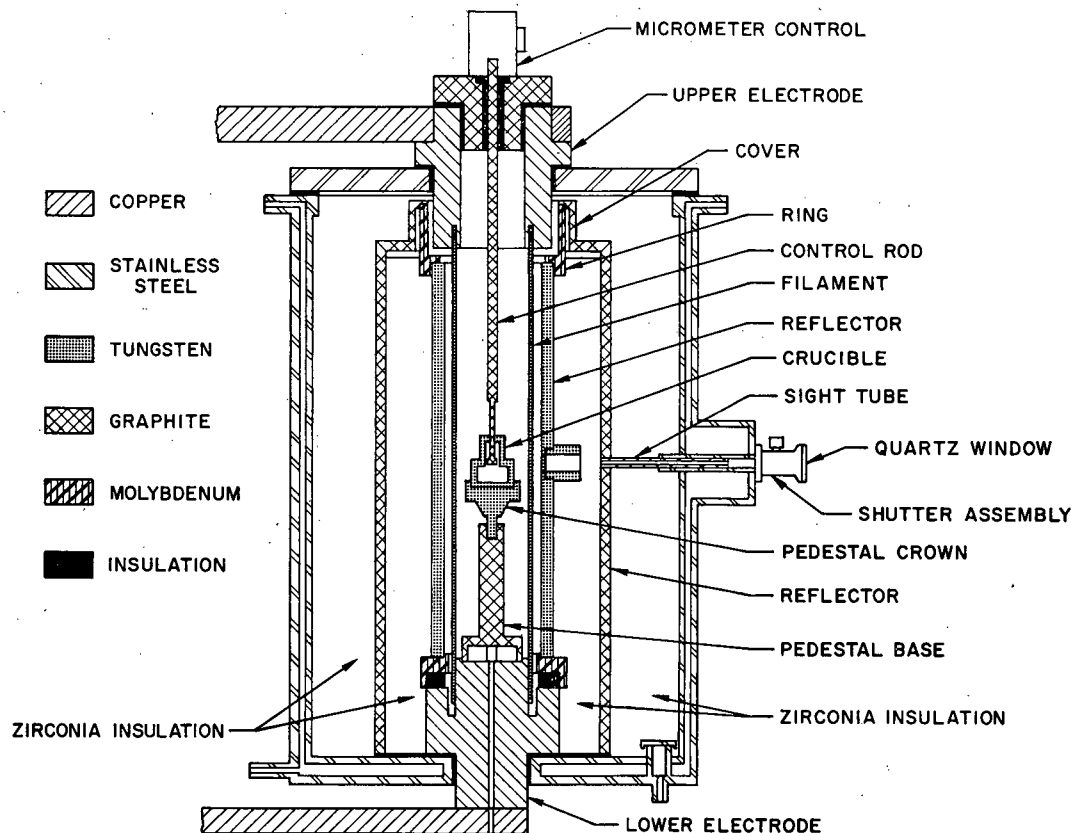


Fig. 25. Modified High-temperature Furnace with Sight Tube and Crucible

The details of the crucible assembly including the pedestal are shown in Fig. 26. A micrometer controls the position of the end plate which is attached to a tungsten rod (control rod). The plate can be traversed over the distance between the crucible cover and the bottom of the empty crucible. The determination of the liquid level is however made by means of high-frequency difference measurements.

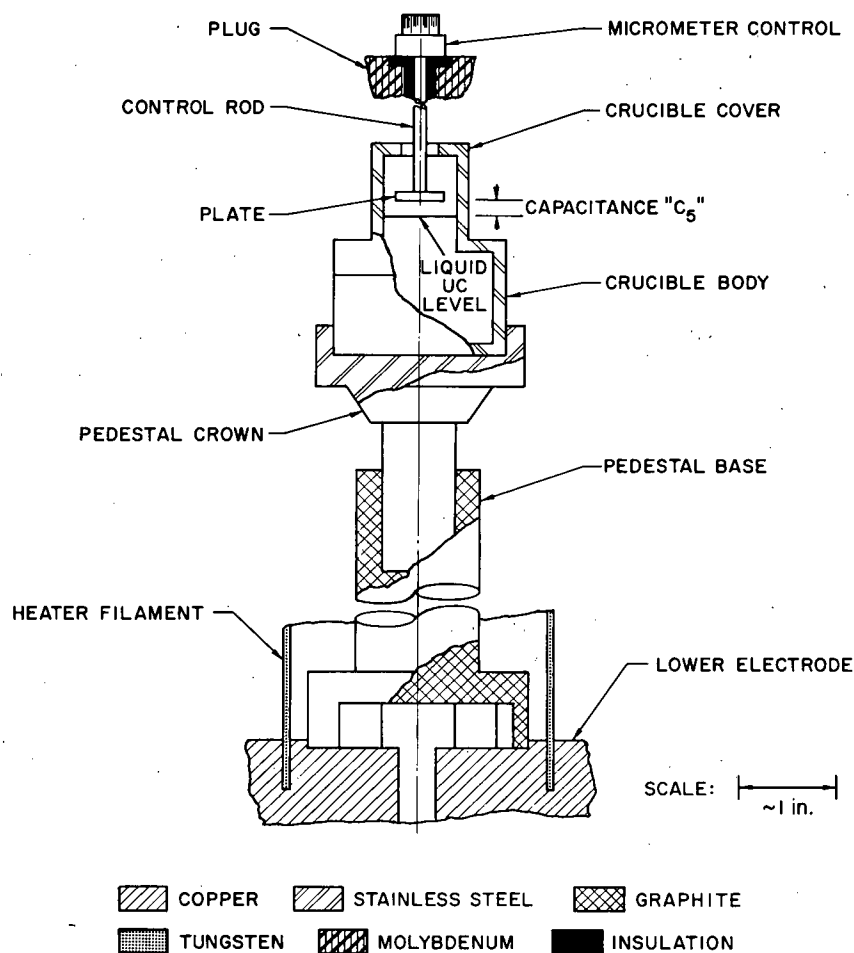


Fig. 26. Pedestal and Crucible with Molten UC

In addition to the furnace setup, two crystal controlled oscillators and a readout system are required.

The variation of the distance between the surface of the molten uranium carbide and the bottom of the plate produces a change in capacitance, producing a change in the frequency of the first oscillator. The change in oscillator frequency by this method is limited from 300-500 cps for a crystal frequency of about 2 Mc. If an inductance is installed either in series or in parallel with the crystal, the change in oscillator frequency can be increased to several thousand cps.

The output of the first oscillator is then mixed with the constant output of the second oscillator to produce a beat frequency. The beat frequency is then analyzed by means of an audio oscillator, oscilloscope, or an electronic recorder to determine the height of the liquid in the crucible. It is believed possible to measure liquid levels to a precision of 10^{-5} in. or less by this method.

b. Furnace Operation. Experiments indicate that graphite pedestals are only useful to about 2700°C . Above this temperature, evaporation of graphite appears to attack the tungsten filament, forming a lower-melting eutectic.

Uranium carbide melts at about 2450°C . This material was heated up to 2700°C in a tantalum carbide crucible and was cooled very slowly. After the run, the crucible was examined destructively. Pieces of the crucible showed that the uranium carbide adhered to the tantalum carbide of the crucible, but the materials remained separated, as far as could be observed.

D. TREAT

1. Operations

Two samples of prototype fast reactor ceramic-type fuel were irradiated for the General Electric Company. A metal-water reaction sample (CEN-224T) was irradiated in a transparent capsule to obtain photographic data on the disintegration of a Zircaloy-clad, uranium dioxide fuel pin in water. Previous metal-water reaction experiments with Zircaloy-clad fuel pins in transparent capsules had been limited to those that did not cause gross melting of the cladding.

E. Plutonium Volatility Safety

1. Chemistry of Tellurium Fluorides

Studies of the reactions of volatile tellurium fluorides are under way to determine the conditions under which tellurium is fixed on solid materials and to investigate the kinetics of the reaction. Equipment is being installed for studies using gas streams in which the concentration of tellurium hexafluoride simulates closely the concentration expected in the off-gas from fluoride volatility processing of reactor fuel materials. In these studies, the TeF_6 gas stream will be spiked with $\text{Te}^{125\text{m}}$ and radiochemical techniques will be used to follow tellurium sorption on the solid materials. In addition, gas-chromatographic methods are being developed for detection of trace quantities of TeF_6 which pass through the sorption beds.

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