ANL-7046

ANL-7046

JUL 8 1965

MASTER

Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

May 1965

PATENT CLEARANCE OBTAINED. RELEASE TO
THE PUBLIC IS APPROVED. PROCEDURES
ARE ON FILE IN THE RESEARCH SECTION.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Reactor Development Program
Highlights of Project Activities for May 1965

EBR-II

The reactor successfully completed its scheduled run (to about 0.8 a/o maximum burnup) and was shut down for fuel inspection as planned. The first fuel irradiation test (G.E. mixed oxide fuel elements) in EBR-II was completed and the irradiation subassembly removed from the reactor. Calibration runs were conducted (using a bare fuel pin in the reactor) and good results obtained from both fuel element rupture detection systems.

The rod-drop and trapezoidal oscillation methods of monitoring the reactor kinetics have been tested and appear to be satisfactory.

FARET

As of the end of May 1965, the A-E Title II design was essentially complete and the ANL design was about 60% complete.

Contract negotiations with Babcock and Wilcox Company for design and fabrication of the reactor vessel were in progress with work estimated to start in June.

An interim status report on FARET was prepared at the request of DRDT.

ZPPR

With release of funds by the AEC in May, the procurement of long lead time items was initiated. A contract for the bed and tables was let to Giddings and Lewis Co., Fond du Lac, Wisconsin. Bids have been received for the matrix tubes and matrix drawers and are being reviewed. Purchase orders and bids for various outdoor items have gone out, and the current expectation is that construction can start on schedule in September 1965.

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60440

REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

May 1965

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

<u>Division</u>	<u>Director</u>
Chemical Engineering	R. C. Vogel
Idaho	M. Novick
Metallurgy	F. G. Foote
Reactor Engineering	L. J. Koch
Reactor Physics	R. Avery
Remote Control	R. C. Goertz

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

Issued June 25, 1965

Operated by The University of Chicago
under
Contract W-31-109-eng-38
with the
U. S. Atomic Energy Commission

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:

November 1964	ANL-6977
December 1964	ANL-6997
January 1965	ANL-7003
February 1965	ANL-7017
March 1965	ANL-7028
April 1965	ANL-7045

TABLE OF CONTENTS

	<u>Page</u>
I. LIQUID-METAL-COOLED REACTORS	1
A. EBR-II	1
1. Operations	1
2. Building Containment	2
3. Systems and Components	3
4. Reactor Physics	4
5. Fuel Cycle Facility	5
B. FARET	6
1. Control Drives	6
2. Fuel Assembly Sodium Flow Test Loop	7
3. Fuel Subassembly	7
4. Argon Enclosure for Fuel-removal Experiments	9
5. Static Test Vessels	9
6. Fuel-handling Machine	9
7. Thermocouple Analysis	10
8. Flowmeters	10
9. Design of Core Support	11
10. FARET Criticals	11
C. General Fast Reactor Physics	14
1. ZPR-6	16
2. ZPR-9	17
3. ZPPR	18
D. General Fast Reactor Fuel Development	19
1. Metallic Fuels	19
2. Jacket Materials	25
3. Carbide Fuel Elements	27
4. Fuel for Zero-power Reactors	29
5. EBR-II Test Fuel Elements	31
6. Corrosion of Fuel-cladding Materials for Sodium-cooled Reactors	31
7. Extrusion Development	32

TABLE OF CONTENTS

	<u>Page</u>
E. General Fast Reactor Fuel Reprocessing Development	33
1. Skull Reclamation Process	33
2. Materials and Equipment Evaluation	33
3. Removal of Nitrogen from Argon	34
4. Halide Slagging Studies	34
II. GENERAL REACTOR TECHNOLOGY	36
A. Experimental Reactor and Nuclear Physics	36
1. Nuclear Constants	36
B. Theoretical Reactor Physics	36
1. Computing Techniques	36
2. Development of Reactor Computing Codes	39
3. Change in Nuclear Temperature to Account for Resolved Levels	40
4. Resonance Interference	41
5. Calculation of Pile-up Distortion	42
6. ZPR-7 Data Analysis	43
C. High-temperature Materials	44
1. Ceramics	44
2. Liquid-Metal Corrosion	48
D. Other Reactor Fuels and Materials Development	51
1. Nondestructive Testing	51
2. Irradiation Testing	52
E. Engineering Development	54
1. Two-phase Flow	54
2. Boiling Liquid Metal Technology	55
3. General Heat Transfer	56
4. ANL-AMU Program	56
F. Chemical Separations	57
1. Fluidization and Volatility Processes	57

TABLE OF CONTENTS

	<u>Page</u>
G. Sodium Coolant Chemistry	61
1. Control of Sodium Oxide Impurity	61
2. Sodium Purification	62
3. Carbon Analysis	62
H. Plutonium Recycle Reactors	63
1. EBWR Facility	63
2. Fuel Development and Fabrication	65
III. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT	67
A. Argonne Advanced Research Reactor (AARR)	67
1. Critical Experiments	67
2. Heat Transfer	73
3. Primary Coolant System	74
4. Fuel and Materials Development	75
5. Studies of Reactor Vessel Stress	76
6. Development of Reactor Components	77
IV. NUCLEAR SAFETY	78
A. Reactor Kinetics	78
1. Fast Reactor Safety	78
B. TREAT	79
1. Operations	79
2. Large TREAT Loop	80
3. Integral Sodium Loop	80
C. Chemical and Associated Energy-transfer Problems in Reactor Safety	81
1. Reactions of Fuel and Cladding Metals with Liquid Sodium	81
V. PUBLICATIONS	83

I. LIQUID-METAL-COOLED REACTORS.

A. EBR-II

1. Operations

At the beginning of this month, the reactor was under sustained operation at a power level of 45 MWt (15 MWe), for a maximum incremental burnup of about 0.15 a/o. On May 4, with a total maximum burnup of 0.64 a/o, the level was decreased in steps while measurements of the power coefficient were made. The shape of the curve of power versus reactivity was essentially the same as the curve at the previous shutdown (on April 11) for 0.50 a/o burnup. Total reactivity change from 45 MWt to zero power was 84 lh.

Fuel handling after this shutdown involved thirteen exchanges between the reactor and the storage basket, with the following experimental subassemblies being loaded: a special stainless steel control rod for use in reactor kinetics measurements; one inner and four outer blanket subassemblies for alpha measurements; a subassembly containing mixed oxides to undergo an experimental irradiation for General Electric Co.; and a subassembly containing U-Pu-Fz alloy for ANL Metallurgy Division.

Two days of zero-power operation followed for the purpose of checking out newly installed reactor-kinetics-monitoring equipment involving rod-drop and trapezoidal techniques (ramp control rod movement). On May 12, reactor power was increased to 45 MWt and the reactor operated to achieve an additional incremental burnup of about 0.15 a/o. This was reached on May 21, with an actual maximum burnup of 0.78 a/o. Rod-drop and trapezoidal tests were made at power at the beginning of the run, and measurements taken to determine power coefficient at the beginning as well as the end of the run.

The most important fuel handlings were the transfer of the G. E. experimental subassembly to the storage basket and then to the Fuel Cycle Facility (FCF), transfer of two core and one inner blanket subassemblies to the basket for later inspection in the FCF, and the loading of one special subassembly containing an unclad pin into the No. 2 control rod location.

The "bare pin" irradiation tests for fission product monitor calibration were then run to calibrate the delayed-neutron monitor and the charged-wire fission gas monitor. The single bare-fuel element was alternately moved from a position below the core to a position inside the core, and the magnitude and the time response of the fission product signals generated through this action were determined with appropriate recording equipment. Three runs at 10 MW were made, one at 50% flow and two at 70% flow.

The results clearly indicated that both monitor systems are capable of detecting the fission product activity associated with a single unclad fuel pin. The results obtained with the Fuel Element Rupture Detection (delayed-neutron) system indicated a very prompt response (over a period of roughly 15 sec) upon the introduction of unclad fuel to the core. Since Na^{24} activity, which constitutes a background effect, was not saturated during the experiment, it is difficult to establish a precise relationship between time, fission product signal, and noise. A rough estimate of the signal-to-noise ratio for saturated Na^{24} activity amounts to approximately 0.5 for the case of a single, completely exposed fuel pin.

As expected, the response time of the charged wire detector was much longer, since its response depends primarily on the decay constants for xenon and krypton isotopes. On the other hand, the results obtained with the charged-wire detector indicated a signal-to-noise ratio of approximately 3, a value significantly greater than for the FERD system.

At month's end, the plant was shut down for scheduled reloading, maintenance, and leak-rate testing of the secondary sodium reactor building penetrations. Total integrated reactor power (thermal) to date is 2013 MWd. Of this total, 547 MWd were accumulated this month.

2. Building Containment

Annual leak-rate tests were performed successfully for the purge exhaust and reactor supply air valves.

3. Systems and Components

a. Control Rod Drives. Control rod drive No. 11, which had been removed because of difficulty with sticking of the jaws, has been examined. Inspection of the sleeve and jaw holder revealed galling marks on the inner diameter of the sleeve and on the outer diameter of the jaw holder. The sleeve was about $1/32$ in. out of round. It appears that interference may have developed between the sleeve and holder which caused binding to occur when the jaws were opened.

b. Reactivity-monitoring Devices. Prior to the 0.64%-0.78% burn-up operation, the special stainless steel control rod was installed in the place of the regular control rod No. 8. Its reactivity value was calibrated by period and against a calibrated control rod No. 3. The worth of the rod was 17 ± 1 lh.

The installation of the control system for the reactivity-monitoring devices was completed and checked out. Data were taken at critical and at power at the beginning of the 0.64% burnup run. Data taken from the power trace obtained during the rod drops were processed to obtain a reactivity plot

versus time, as shown in Figure 1. At least 16% of the reactivity feedback is recovered in less than 0.5 sec. This feedback is probably associated with axial fuel expansion.

Figure 1. Reactivity vs. Time

The trapezoidal monitoring device was used with an 11.6-sec drive time and wait times of 1, 4, 12, and 20 sec at criticality and 40 MW. A comparison between the peak-to-peak amplitudes at criticality and at power for each wait time shows a strong negative feedback component at each frequency (Table I). These two techniques will provide satisfactory monitoring of the reactor during the interim time when no oscillator is available.

Additional rod-drop data were taken during reactor shutdown to investigate the effect of different power levels on the prompt reactivity time response.

Table I. Trapezoidal Results

Drive time of 11.6 sec (corresponds to insertion of 9.4 lh)

Wait Time (sec)	$\Delta N/N$ (Peak to Peak)	
	Zero Power	40 MW
1	0.063	0.056
4	0.078	0.066
12	0.106	0.088
20	0.132	0.100

c. Secondary Cold Trap. The pressure drop across the secondary cold trap crystallizer tank showed a steady increase, indicating progressive oxide plugging or blockage due to particulates of alpha-iron and mill scale. A momentary back flow through the cold trap caused a 50% reduction in pressure drop at month's end. The cold trap was operating at a reduced flow rate (8 gpm) with no increase in pressure drop. The cold trap has been in continuous operation keeping the plugging temperature at about 250°F. The plugging temperature of the primary sodium has remained below 230°F.

4. Reactor Physics

a. Measurements of Power Coefficient. The power coefficient was measured during the two startups and two shutdowns this month, with results given in Table II.

Table II. Power Coefficient

Date	Burnup, a/o	Power Mode	Reactivity Change (lh)
4/25	0.5	Increasing	78
5/5	0.64	Decreasing	83
5/12	0.64	Increasing	76
5/21	0.78	Decreasing	78

The results of these measurements continue to present difficulties in absolute analysis, but several trends are emerging: (1) the precise repeatability of reactivity is complicated apparently by slight mechanical movement in the core; (2) the overall coefficient remains fairly constant; (3) the coefficient appears to be largest between 0 and 10 MW; (4) the slope of the power coefficient decreases significantly in the power range from 20 to 30 MW. The accuracy of measurement is still compromised by the lack of instrumentation giving good data for inlet sodium temperature.

5. Fuel Cycle Facility

a. Argon and Air Cell Systems. Failure of the crane-trolley-operating mechanism in the Air Cell necessitated its inspection, so it was moved to a position beneath the roof hatch. Removal of irradiated fuel from the cell made it possible to open the cell roof plugs and examine the trolley drive unit with personnel-exposure levels of less than 150 mR/hr. It was found that a pivot bolt in the clutch engaging lever had become unfastened and fallen out. The bolt was replaced, and the crane was restored to service.

b. Irradiated Fuel Processing

(i) Sodium Removal, Dismantling, and Decanning. A core subassembly (maximum burnup of 0.62 a/o), a control subassembly, and an inner blanket subassembly were received from the reactor for processing. Since the reactor was operating at power when the transfers were made, sodium removed from these assemblies in the interbuilding cask approached an equilibrium Na^{24} activity level for the first time. Personnel radiation exposures in the cleaning operation were very small; the cask provided adequate shielding, and an instantaneous level of only about 100 mR/hr was observed adjacent to the transfer line when the wash solution was dumped.

The subassemblies were dismantled without difficulty. No abnormalities were noted for either the subassemblies or the elements removed from them.

In decanning elements from subassemblies with a maximum burnup of 0.44-0.50 a/o, stiffening of the spiral wire wrap on about one-fifth of the elements was noted. This is possibly an irradiation effect. The use of the master-slave manipulator was required to assist removal of the wire from the element after shearing. Otherwise, decanning proceeded routinely with the use of a heated scrap-separator tube.

(ii) Melt Regining and Oxidation. Four melt-refining runs, and corresponding skull-oxidation runs, were carried out with ingot yields of 93-95%. In three of these runs, irradiated and unirradiated recycle scrap metal was consolidated; the charge to the fourth run was principally irradiated pins of 0.44-0.50 a/o maximum burnup. Following oxidation of the skull from the latter run (after 3 weeks of decay), a bright glow was noted inside the crucible just above the melt line. It appears that the glow is the result of fission products deposited on the surface and is not a thermal effect. The crucible, after dumping the oxides, exhibited an activity level of about 50,000 R/hr at one foot (dumping effected an incomplete separation because the crucible broke around the bottom), and the oxide dumped had an activity of about 40,000 R/hr at one foot.

c. Fuel Fabrication. In the first attempt at injection casting of the ingot obtained from irradiated pins of 0.44-0.50 a/o maximum burnup, a power failure was experienced a few minutes before the scheduled casting time. Since casting could not be accomplished, the charge froze in the crucible. It was subsequently dumped, recharged to an oversize crucible, and cast satisfactorily. Three additional casting runs were made with unirradiated fuel alloy.

Two minor mechanical failures were encountered in operation of the sodium-bond testing machine. A retaining ring, on the piston of an air cylinder, used to move the fuel element through the test coil, was found to be broken when the unit was moved out of the Air Cell for repair. Also, a small spring-actuated plunger on the magazine pivot assembly was limiting piston travel as a result of excessive wear. Both defects were corrected, and the unit was placed in service.

d. Fuel Surveillance. The device for puncturing fuel cans and measuring the pressure and void space above the fuel rod has been placed into service. Representative samples of fuel elements from each irradiated assembly have been subjected to sodium-level measurement (by the eddy current device), pressure and void space measurement, and diameter and length measurement on the pin-processing machine after sodium removal. Visual observation of the pins shows them generally to be in excellent condition. The maximum diameter increase noted was only of the order of one mil. Tendencies for increase in sodium level and decrease in void volume have been noted as burnup has increased.

B. FARET

1. Control Drives

Testing of the bellows seal continues (see Progress Report for January 1965, ANL-7003, pp. 34-37; for April 1965, ANL-7045, p. 9). After 4800 cyclic operations the bellows show no signs of wear or damage.

Fabrication of the control drive gripper and of its test facility is continuing. The control-drive shock absorber and its associated equipment are being installed in a test facility in conjunction with the electromagnetic latching device.

A facility to test electrical components in 150°F dry argon is being fabricated.

The design of the control-drive prototype is about 65% complete. That of the control-drive lifting platform is about 30% complete.

2. Fuel Assembly Sodium Flow Test Loop

The vessel extension piece has been received at ANL. All critical dimensions have been checked and the extension is ready for installation in the loop.

The vessel cover has been completed and can be installed when required.

The vessel internals, which simulate the FARET core-support structure, are in the final stages of completion. The grid plates have been rough machined, and welding of the plenum system is in progress. The Conoseal joint required for the inlet connection was returned to the vendor for rework.

The reflector and mock-up fuel assemblies are in various stages of fabrication. The reflectors are approximately 75% completed and the fuel subassemblies about 85% completed.

The final welding and radiographic inspection of the 2-in. drain lines, including the valves, has been completed. These subassemblies are ready for final field welding into the loop. The heaters, together with the 0.003-in.-thick Rigidized metal thermal barrier, have been installed in the 3-in. overflow line. Application of thermal insulation to the loop continues. All instrumentation is now wired to the patch boards, and the extension of the electrical instrumentation lines will be done after the application of thermal insulation has been finished.

The 800-gpm pump will probably be delivered early in August 1965.

3. Fuel Subassembly

The instrumented core subassembly shown in Figure 2 is typical of those planned for FARET Core I. The noninstrumented subassemblies would have essentially the same general arrangement. Two reference fuel pin sizes have been established. For a reference 61-pin and 37-pin subassembly, the pin OD's are 0.220 and 0.290 in., respectively. Cladding thickness will be in the range of 0.015-0.020 in.

Drawings for two experimental instrument lead cutters and for a test stand on which to operate them are being completed. These units will be used to test the effectiveness of various concepts of shear and clipper-type cutters to cut and/or seal a variety of proposed instrument leads in FARET subassemblies. A surplus General Mills Mod. 300 manipulator will be used to work out spatial and handling problems involved in the operation and maintenance of the cutters. Arrangements are now being made to move the manipulator and support structure from Building 6 to the mock-up area in Building 208. Use of this manipulator for similar tests on other in-cell equipment is also anticipated.

Figure 2. Instrumented Core Subassembly

4. Argon Enclosure for Fuel-removal Experiments

To test the fuel handling mechanisms under simulated reactor conditions, an Argon Enclosure has been designed to be assembled directly above the pressure vessel of the FARET Fuel Assembly Flow Test Loop. This enclosure contains an operational model of the fuel-removal mechanism and is capable of removing a fuel element and transferring it into an adjacent container.

All drawings are complete and have been sent to the shop for procurement and erection. This facility is essentially a simple steel box set above the pressure vessel on H-beam columns. Inside this box are rails to accommodate a fuel-removal machine. Adjacent to the pressure vessel and penetrating this box is an 8-in. pipe to contain a fuel element, thus simulating the fuel-removal process in the reactor.

5. Static Test Vessels

The final drawings have been sent to the shop for procurement. Orders for the three small vessels have been placed with Stearn's Rogers of Denver; probably the order for the large vessel (see Progress Report for March 1965, ANL-7028, Figure 2, p. 10) will be placed with this company. Inquiries have been made for building the required pit in Building 206.

6. Fuel-handling Machine

Design of a gas-cooling and cleaning unit for the cooling loop of the fuel-handling machine has been completed, and shop fabrication of a prototype unit has been started.

The principal feature is that decay heat from the fuel, transferred to argon in the fuel-handling machine, is rejected in the cooling loop by direct contact with NaK through which the gas is bubbled. The NaK is kept at a low temperature (about 45°F) by a simple, immersed cooling coil through which refrigerant ammonia is circulated.

At the same time the gas is cooled, sodium vapors are condensed from the gas by the liquid NaK. The degree of condensation of sodium and therefore the purification of argon obtainable in the loop is a function of temperature. This method will provide a convenient method for changing the filters merely by replacing the NaK at some convenient time when the sodium concentration reaches a predetermined limit. It is estimated that the NaK will be replaced when the sodium content has reached about 50%.

7. Thermocouple Analysis

Tungsten-rhenium alloy, bare wire, thermocouple calibrations are being made in an effort to extend, to at least 2800°C, the temperature-emf table currently published and adopted by the wire manufacturers. Measured values treated by a least-squares fit to the data taken to date for the W-3% Re/W-25% Re thermocouple combination are listed in Table III along with the wire manufacturers' current average values.

Table III. Measured Emf-Temperature Relationship
W-3% Re vs. W-25% Re Thermocouples

Temperature (°C)	Emf (mV), Measured	Emf (mV), Manufacturers
1000	18.439	18.242
1100	20.313	20.232
1200	22.212	22.192
1300	24.113	24.083
1400	25.994	25.956
1500	27.836	27.780
1600	29.623	29.551
1700	31.340	31.269
1800	32.975	32.922
1900	34.519	34.504
2000	35.966	36.007
2100	37.312	37.406
2200	38.554	38.677
2300	39.693	39.777
2400	40.732	40.678
2500	41.677	-
2600	42.536	-
2700	43.319	-
2800	44.040	-

8. Flowmeters

a. Electromagnetic Flowmeter. The magnets for the electromagnetic (EM) flowmeter (see Progress Report for November 1964, ANL-6977) have been fabricated and tested. The magnetic flux before temperature aging was measured and agrees very well with the calculated value (about 5000 G). The measured flux in the air gap (about 410 G average), however, is about 10 times higher than originally calculated. The high flux is attributed to the use of a fringe factor much too high in the original calculation. The centerline axial flux distribution in the air gap is shown graphically in

Figure 3a and the radial flux distribution 1 in. below the top of the magnet and, from pole piece to pole piece, in Figure 3b. The higher flux in the meter increases the meter sensitivity considerably and is an unexpected advantage. The meter sensitivity is now expected to be about 5.0 mV/ft-sec.

Figure 3
Flux Distributions in Electro-magnetic Flowmeter

After temperature aging the magnets at 650°C, a permanent reduction of about 20% in the flux was noted. Further aging up to about 700°C is planned. The EM flowmeter is about 75% assembled and should be completed and ready for testing within the next month.

b. Thermal Flowmeter. The thermal flowmeter is being fabricated by an outside vendor and should soon be available.

c. Turbine Flowmeter. The high-temperature wire for the turbine flowmeter coil has finally arrived. A preliminary coil has been fabricated and tested at room temperature on the modified flowmeter. Further high-temperature tests will first be made on this coil design before a sodium loop test will be performed on the completed flowmeter.

9. Design of Core Support

The theoretical strength and deflection analysis of the FARET two-plate core-support structure has been completed. The algebraic equations have been independently checked. The problem was programmed for the CDC-3600 computer. This program is not as yet perfected for final numerical calculations.

10. FARET Criticals

Experiments continued with Assembly 46, the series of small, Pu-U-fueled cores providing data for the design of FARET. Measurements

of the neutron lifetime in the 68-liter, carbide-simulation core, core 46B (see Progress Report for February 1965, ANL-7017, pp. 19-21) were completed in April. This core was replaced with an oxide-fuel simulation core, core 46C, and lifetime measurements with the pulsed neutron generator were repeated. Experiments with the oxide core were in progress at the end of May.

Neutron-lifetime determinations were carried out in core 46B by both a pulsed-neutron technique and the conventional Rossi-alpha technique. Various neutron detectors, placed in the core and in the reflector, were used to examine the pulse-decay characteristics of the reactor at subcritical reactivities ranging from 50 to 1200 lh (about $2\frac{1}{2}\%$ $\Delta k/k$). Variations of results with different detectors and reactor locations suggest that spectral equilibration must be considered in the interpretation of the data. Further analysis of these experiments is under way.

Construction of core 46C was done with the neutron generator equipment in place. After attaining a critical loading with this core and calibration of control rods, the alpha measurements with pulsed neutrons were repeated. Subcritical reactivities for these experiments were attained by withdrawing core-loaded control rods and also by removal of some of the reflector.

The composition for the $\text{PuO}_2\text{-UO}_2$ fuel simulation core is given in Table IV. This simulation was achieved with plates of Pu, U, U_3O_8 , and with some graphite representing part of the oxygen. Figure 4 shows the interface view of Half No. 1 of the reactor with the critical configuration of the core (after removing the neutron generator tube), Half No. 2 being

Table IV. Composition of ZPR-3 Assembly 46C

Material	Composition	
	g/cc	10^{22} atoms/cc
$\text{Pu}^{239+241}$	0.424	0.107
$\text{Pu}^{240+242}$	0.020	0.005
U^{235}	1.305	0.334
U^{238}	1.356	0.343
$\text{U}^{234+236}$	0.019	0.005
O	0.230	0.866
C	0.100	0.502
Na	0.322	0.843
Fe	1.019	1.099
Cr	0.255	0.295
Ni	0.141	0.145

Figure 4. Interface View of ZPR-3 Assembly 46C

symmetrical. The length of this oxide core is 20 in., which may be compared with 24 in. for the carbide core, but the radial and axial reflector compositions and outer extensions are the same as for core 46B. From an evaluation of the available excess reactivity of the core configuration of Figure 4 and measurements of fuel worth at the core edge, the just-critical volume for core 46C was calculated to be 86.9 liters. This is equivalent to about fifty-seven 20-in. core subassemblies in FARET.

Experiments are nearing completion with mock-ups of B_4C control rods in the reflector at positions relative to the core as proposed for FARET. The mock-up rods simulate a FARET active-rod channel loading of about 55 v/o B_4C , 25 v/o sodium, and 20 v/o steel. Two series of rod-worth studies are involved, one with an enrichment of about 31% B^{10} , and the other with approximately 70% B^{10} in B.

Four mock-up rods with 20-in.-long, 31%-enriched active sections were first constructed to simulate four adjacent rods in the "up" position in FARET, with the bottom of the active rod at the top of the core. The reactivity loss incurred by the introduction of these mock-up rods indicates that twelve control rods of similar composition and situation in FARET would require an additional $2\frac{1}{4}$ fuel subassemblies at the core edge for compensation. The active section of one of the mock-up rods was then moved inward in steps a total of 20 in.; the active length was then aligned with the axial length of the core. The total reactivity lost from this simulated rod motion was 235 lh, or about 0.47% $\Delta k/k$. Simulated insertions of the other three mock-up rods were also done in stages to evaluate their worths and determine what interactions or shadowing effects exist between adjacent rods. The results of this work and the studies with the 70% enriched B_4C mock-up rods are being analyzed.

C. General Fast Reactor Physics

1. ZPR-6

Assembly 4Z went critical during the first week of May 1965. This assembly is a zoned reactor having four radial regions: central, buffer, driver, and reflector. The purpose of the zoned assembly is to test, in conjunction with the reference core (full size), the extent to which physics parameters of the reference assembly may be predicted on the basis of measurements performed on the zoned system. The fundamental advantage of zoned cores is that they require a smaller fuel inventory.

a. Composition and Critical Mass. Assembly No. 4Z consists of four regions: (1) the central region extends from the center of the core to 41.43 cm in the radial direction and has the same composition as the reference core; (2) the buffer region extends from 41.43 to 46.96 cm; (3) the driver region extends from 46.96 to 53.88 cm; and (4) the blanket is 30 cm of depleted uranium. Assembly No. 4Z is cylindrical in geometry and the axial length is 122 cm. The axial blanket is 30 cm of depleted uranium. Table V lists the concentrations of all the materials in both Assembly No. 4Z and the reference core.

Table V. Volume Fractions in Assembly 4Z and the Reference Core

Material	Volume Fractions in Reference Assembly ^(a)		Volume Fractions in Assembly No. 4Z ^(b)			Volume Fractions in Blanket (Common to Both Assemblies)
	Core Region	Critical Mass	Region			
			Central ^(c)	Buffer ^(d)	Driver ^(e)	
U ²³⁵	0.0319	1525 kg	0.0319	0.00095	0.1428	0.00169
U ²³⁸	0.2213		0.2213	0.4539	0.1119	0.8319
Na	0.4208		0.4208	0.00	0.4208	-
C	0.1539		0.1539	0.3601	0.1539	-
SS	0.1483		0.1483	0.0708	0.1483	0.0708

(a) The volume of the reference core is 2554 liters.

(b) The volume of the zoned core is 1100 liters.

(c) Central region extends from 0 to 41.43 cm.

(d) Buffer region extends from 41.43 to 46.96 cm.

(e) Driver region extends from 46.96 to 53.88 cm.

Table VI shows a comparison of the calculated and measured critical mass of Assembly No. 4Z. The measured value does not include any corrections for heterogeneity effects.

Table VI. Comparison of Calculated and Measured Critical Mass (kg)
of Assembly No. 4Z (ZPR-6)

	Code and Cross Section Set			
	MAIM-6 (Set 224--22 Groups)	RE-122 (Set 635--16 Groups)	RE-122 (Set 149--4 Groups)	SNARG-S4 (Set 224--22 Groups)
Calculated*	1082	1103	1152	1074
Measured	1101			

*Calculations were done in cylindrical geometry.

b. Spectral Matching. The fluxes and adjoints in the zoned and reference assemblies are compared at the center of each core in Table VII. The fluxes and adjoints are normalized to $100 \text{ neutrons cm}^{-2} \text{ sec}^{-1}$ at the center. Although the values reported in Table VII were calculated for spherical geometry, a similar agreement between the fluxes and adjoints was obtained in cylindrical geometry. Table VIII shows the comparison between the fluxes and adjoints in the two assemblies at 30 cm from the center of each core. The last column of Table VII and of Table VIII is an indication of the "goodness" of spectral matching.

c. Fatman Effect. The fatman effect was calculated for the reference core by means of a one-dimensional diffusion code. The fatman consisted of a slab of Benelex placed adjacent to the face of one-half of the reactor. Two thicknesses for the slab, 20 cm and 50 cm, were used. The calculated k_{eff} and the measured k_{eff} of Assembly No. 4Z are shown in Figure 5. Although the calculation and measurement of k_{eff} are associated with two different assemblies, the k_{eff} of the bare half of both assemblies was the same. The uncertainty in the measurement is perhaps 5%. The discrepancy between measurement and calculation is perhaps due to the fact that the half was far subcritical.

d. Measurement of the Spectrum and Doppler Effect. The neutron spectrum, at various locations in Assembly No. 4Z, was measured with a proton recoil counter. The data are being analyzed.

Experiments for measuring Doppler coefficients are now underway.

Table VII. Comparison of Fluxes and Adjoints in the Reference and Zoned Cores[†]

Group/ E_L (keV)	ϕ_{Center}		ϕ_{Center}^*		$\phi\phi_{\text{Ref.}}^* - \phi\phi_{4Z}^*$
	Ref. Assembly	Assembly No. 4Z	Ref. Assembly	Assembly No. 4Z	
1/3680	1.462	1.475	5.411	5.425	-0.091
2/2230	2.837	2.835	5.130	5.137	-0.009
3/1350	4.774	4.763	4.676	4.678	+0.042
4/821	7.301	7.282	4.054	4.051	+0.099
5/498	10.479	10.459	4.029	4.024	+0.133
6/302	12.995	12.981	4.070	4.061	+0.174
7/183	13.197	13.193	4.064	4.052	+0.175
8/111	11.445	11.448	4.046	4.034	+0.205
9/67.4	9.823	9.832	4.034	4.024	+0.062
10/40.9	7.801	7.814	4.037	4.029	+0.010
11/24.8	5.952	5.965	4.063	4.059	-0.029
12/15.0	4.742	4.756	4.137	4.135	-0.048
13/9.11	2.977	2.986	4.252	4.254	-0.044
14/4.31	2.330	2.336	4.445	4.448	-0.034
15/2.61	0.251	0.251	4.621	4.625	-0.001
16/2.03	0.518	0.519	4.637	4.641	-0.007
17/1.23	0.488	0.489	4.630	4.634	-0.007
18/0.961	0.256	0.256	4.635	4.640	-0.001
19/0.583	0.243	0.243	5.025	5.029	-0.001
20/0.275	0.082	0.082	5.284	5.289	-0.001
21/0.101	0.0255	0.0255	5.393	5.397	-0.000
22/0.029	0.0016	0.0016	5.323	5.327	-0.000

[†]Calculations were done in spherical geometry (MAIM-6, cross-section set 224).

Table VIII. Comparison of Fluxes and Adjoints at 30 cm from the Center of the Reference and Zoned Cores†

Group	$\phi_{30 \text{ cm}}$		$\phi_{30 \text{ cm}}^*$		$\phi\phi_{\text{Ref.}}^* - \phi\phi_{4Z}^*$
	Ref. Assembly	Assembly No. 4Z	Ref. Assembly	Assembly No. 4Z	
1	1.476	1.470	5.411	5.53	-0.142
2	2.838	2.800	5.130	5.219	-0.054
3	4.775	4.681	4.676	4.737	+0.154
4	7.362	7.242	4.054	4.077	+0.320
5	10.478	10.490	4.028	4.048	-0.254
6	12.993	13.042	4.069	4.089	+0.540
7	13.194	13.240	4.064	4.068	-0.240
8	11.443	11.480	4.046	4.033	-0.001
9	9.822	9.874	4.035	4.008	+0.057
10	7.801	7.850	4.037	4.000	+0.093
11	5.953	5.985	4.064	4.019	+0.139
12	4.744	4.741	4.137	4.086	+0.254
13	2.979	2.963	4.252	4.203	+0.214
14	2.332	2.319	4.445	4.409	+0.142
15	0.251	0.250	4.621	4.600	+0.010
16	0.518	0.515	4.637	4.611	+0.027
17	0.489	0.481	4.630	4.607	+0.048
18	0.257	0.249	4.636	4.616	+0.042
19	0.244	0.232	5.025	5.010	+0.064
20	0.0819	0.0775	5.284	5.280	+0.024
21	0.0256	0.0243	5.392	5.400	+0.007
22	0.00165	0.00158	5.323	5.340	+0.003

†Calculations were done in spherical geometry (MAIM-6, cross-section set 224).

Figure 5

Dependence of k_{eff} of One-half of the Assembly on the Thickness of Benelex Placed Adjacent to the Midsection of the Core

e. Control Rod Calibration. The control rods were calibrated by the period method. The worths of the safety rods were determined by the rod-drop method in relation to a calibrated rod. The total shutdown margin was found to be 2.6% $\Delta k/k$.

2. ZPR-9

Reactivity measurements were begun on Assembly No. 6 (see Progress Report for April 1965, ANL-7045, pp. 17-19) by homogeneously adding hydrogen in the aluminum reflector. Strips of polyethylene were used as a source of hydrogen. Twenty-mil strips were placed on the bottom and 5-mil strips at the side of each drawer. Results of these measurements are listed in Table IX. Small reactivity adjustments were made with a carefully calibrated control rod while large changes were made by removing fuel from the central core drawers.

Table IX. Reactivity Effect due to
Polyethylene in the Reflector

Polyethylene Weight, kg	Total Number of Drawers Loaded	Reactivity Added, Ih
0.148	8	1.82
0.206	10	2.73
0.593	32	16.64
1.168	66	25.50
1.755	100	35.84
4.031	202	102.70
5.900	324	151.43
7.936	434	193.06

The axial fuel-expansion coefficient for Assembly No. 6 was calculated for slab geometry (16-group diffusion code) to be -1.65 Ih/half-cm expansion of the fuel at the center of the assembly of a single drawer into an aluminum reflector and -1.25 Ih for the same expansion into the Al_2O_3 axial reflector. Measurements confirming these calculations were made preparatory to reloading the reflector with Al_2O_3 .

Triangular-shaped fuel wedges were separated in such a way as to make a 12-in.-long by 2-in.-high fuel column 0.2 in. longer and something slightly less than 2 in. high in order to simulate fuel expansion in a reactor incident. The change in reactivity associated with this geometry was measured by two means:

(a) In the least reactive state, the reactor was put on a very long period (about 400 sec). The more reactive condition then shortened the period. Period data were taken with the RIDL analyzer in 400 channels, each 1/2 sec wide. These data were then fitted by the least-squares method with the DDP-24 for the two periods. The periods were then used in the in-hour equation to solve for the excess reactivities.

(b) The critical rod positions were determined for "expanded" and "normal" states, and reactivities were determined from previously determined rod calibrations.

Results of the measurements are given below.

Fuel Expansion into	Ih for Center Drawer		
	Calc	Method A	Method B
Al	-1.65	-1.25	-1.24
1 drawer Al ₂ O ₃	-	-1.28	-1.33
9 drawers Al ₂ O ₃	-1.25	-1.18	-1.29

In the second measurement above, only the central drawer had an Al₂O₃ axial reflector, while in the third measurement the central 9 drawers had Al₂O₃ axial reflectors. It is estimated that the experimental errors are between ± 0.05 and 0.1 Ih.

3. ZPPR

The architect-engineer has completed 80% of the Title II effort. Final details of control and instrumentation items of the facility were agreed upon with the architect-engineer.

The release of funds by the AEC has allowed the project to proceed with the procurement of long-lead-time items.

The status of the long-lead-time reactor components is as follows:

1. Bed and Tables: A purchase contract was mailed to Giddings and Lewis Co., Fond du Lac, Wisconsin.

2. Matrix Tubes: Bids have been received. Representatives of the Laboratory have visited the apparent low bidder, and are evaluating his capabilities and qualifications.

3. Matrix Drawers: Bids have been received. Representatives of the Laboratory have visited the apparent low bidder, and are evaluating his capabilities and qualifications.

4. Fuel-rod Drives: The purchase order for the modified fuel-rod drives was placed. The prototype, which includes the modifications of the gearing readout and motors, was tested satisfactorily.

5. Poison Rod Drives: The final drawings of the poison rod drives are being prepared for the bid package.

The outside electrical and steam system package went out for bids May 7, 1965, and were due back May 27, 1965. A purchase order was issued for the outdoor substation to be installed under this bid package May 14, 1965. A purchase order for the steel beam posts for the ZPPR foundation was placed May 19, 1965. These items should allow the construction of the ZPPR cell to start on schedule in September 1965.

Six sections of the specifications for the main ZPPR facility package have been received from the architect-engineer and are being reviewed. The remaining 42 sections are presently being drafted by Mason & Hanger.

An informative meeting with the Project Roller Coaster personnel indicated that the data obtained from their experiments substantiate the design and containment features of the ZPPR reactor cell.

D. General Fast Reactor Fuel Development

1. Metallic Fuels

a. Improved Uranium-Plutonium-based Metal Fuels. Emphasis in fast reactor metal fuel investigations has turned to the development of high-uranium-plutonium-based metal fuels that are compatible with inexpensive cladding or jacketing materials that are, themselves, resistant to corrosion in sodium. The higher melting temperatures (compared with the relatively low-melting U-Pu-Fz alloys) should permit higher fuel operating temperatures and higher burnups, as well as safer fuels in the event of an excursion. The alloying elements that presently show the most promise are titanium and zirconium; they raise substantially the solidus temperatures of the binary uranium-plutonium alloys and, from physics considerations, are the least undesirable of the elements studied. Therefore, the pertinent properties of U-Pu-Ti and U-Pu-Zr alloys are being investigated.

(i) Compatibility. The possibility that an inexpensive commercially available iron- or nickel-based alloy can be used as a cladding or jacketing material has greatly increased the interest in uranium-plutonium-based metal fuels.

We have recently found that Type 304 stainless steel is compatible with U-18.5 w/o Pu-14.1 w/o Zr alloy up to 800°C. This is contrary to all of our previous experience with U-Pu-Fz and U-Pu-Ti alloys, which showed melting of reaction products and drastic penetration into the cladding and into the fuel in the range from 650 to 675°C (see Figure 6).

Figure 6. Compatibility of Type 304 Stainless Steel with Uranium-Plutonium-based Fuels after 7 Days at Temperature. Compositions given in w/o.

Figure 6a shows the reaction zones after seven days at 650°C of Type 304 stainless steel with U-15 w/o Pu-10 w/o Fz, U-18.5 w/o Pu-14.1 w/o Zr, and U-22.1 w/o Pu-11.8 w/o Ti alloys. The reaction layer for the zirconium alloy is so thin (approximately 1 μ) that it is hardly discernible, whereas melting and thick reaction layers formed on both the cladding and the fuel side of the other couples. Figure 6b shows the influence of increasing temperature on the U-18.5 w/o Pu-14.1 w/o Zr stainless steel couple. The reaction layer remains thin and melting does not occur up to 800°C. At 850°C gross melting did occur. The transformation to the gamma phase is complete just above 650°C, which accounts for the change in the fuel microstructure.

Microprobe analysis has shown that the very thin reaction layer in the U-18.5 w/o Pu-14.1 w/o Zr stainless steel couple contains about 50 w/o Zr, about 15 w/o Fe, and lower amounts of uranium, plutonium, and chromium.

(ii) Phase Relationships and Properties. Earlier emphasis on U-Pu-Ti alloys resulted in our doing more work on the ternary U-Pu-Ti phase diagram than on the U-Pu-Zr system. The U-Ti alloys are different from the U-Zr alloys in that they are characterized by the presence of a compound U_2Ti in the composition range of interest. The compound exists over only a narrow homogeneity range very near to the stoichiometric composition. It is formed from the γ -U solid solution at 898°C.¹ It has been reported² that plutonium will replace about half of the uranium in the U_2Ti compound. The introduction of plutonium will cause a slight expansion of the U_2Ti lattice. The lattice constants of U-16.7 a/o Pu-33.3 a/o Ti, homogenized for one week at 1050°C and water quenched, were found to be $a_0 = 4.843 \pm 0.001 \text{ \AA}$ ³ compared with $a_0 = 4.828 \text{ \AA}$ and $c_0 = 2.847 \text{ \AA}$ for the binary U_2Ti .¹

Water quenching of the γ -U solid solution did not prevent the compound formation. When the same alloy was subsequently heat treated for 3 days at 775°C and water quenched, the parameter a_0 was reduced by 0.23% to $4.832 \pm 0.0002 \text{ \AA}$, while the parameter c_0 was expanded by 0.42% to $2.8586 \pm 0.0002 \text{ \AA}$, contrary to the observations in Ref. 2.

These lattice changes are reflected in the dilatometric curves. For example, an alloy with the nominal composition U-10.7 a/o Pu-35.6 a/o Ti expands by no less than 1% during the transformation of the $(U, Pu)_2Ti$ compound to a γ -U solid solution. This large expansion during transformation is observed on all alloys of sufficiently high titanium content in which the compound $(U, Pu)_2Ti$ predominates. This applies particularly to

¹Knapp, A. G., The System Uranium-Titanium, J. Inst. Metals **83**, 497-504 (1954-55).

²Quarterly Status Report Advanced Reactor Technology for Period Ending January 31, 1965, LA-3244-MS, pp. 31-32.

³Reactor Development Program Progress Report, ANL-7017 (Feb 1965), p. 29. (The a_0 value is in error. It should read 4.843 $\text{\AA}$. The values for c_0 and the c/a ratio are correct.

alloys with 33.3 a/o Ti and more in which the compound is in equilibrium with α -Ti or β -Ti depending upon temperature. In alloys with less than 33.3 a/o Ti the phases of the uranium-plutonium system increasingly determine the expansion characteristics of the alloys as the titanium content decreases (see Figure 7).

Figure 7
Thermal-expansion Curves for U-Ti,
U-Pu-Ti, and U-Pu-Zr Alloys

The temperatures above which the ternary alloys consist only of the (γ -U, β -Ti) solid solution are listed in Table X.

Table X. Completed Temperature of the $(U, Pu)_2Ti \rightarrow (\gamma-U, \beta-Ti)$
Solid-solution Transformation

Nominal Composition of Alloy (a/o)			Transition Temperature (°C)
U	Pu	Ti	
79	0	21	878
75	10	15	809
72	18	10	755*
69	25	6	673
65	35	0	700
47	0	53	890
53.7	10.7	35.6	848
56	14	30	800
63	25	12	732
68	32	0	710
45	15	40	816

*Interpolated value.

The thermal expansion properties of three alloys: the binary U_2Ti compound, a ternary U-Pu-Ti alloy, and a quaternary U-Pu-Ti-Zr alloy, are shown in Table XI. The results of measurements of other U-Pu-Ti and U-Pu-Zr alloys were also reported in Ref. 3.

Table XI. Thermal Expansion of Three Uranium Alloys

Alloy Composition (a/o) (w/o)	U-33 Ti U-9.1 Ti	U-12.9 Pu-17.1 Ti-15.8 Zr U-16.9 Pu-4.5 Ti-7.9 Zr	U-10 Pu-15 Ti U-11.4 Pu-3.4 Ti
Expansion Coef. $\times 10^6/^{\circ}C$ (25 $^{\circ}C$ to 1st transformation)	20.1	17.4	19.2
Transformation Range, $^{\circ}C$	860-897	562-769	566-809
Expansion, $\Delta L/L \times 10^3$	10.6	9.0	12.0
Expansion Coef. $\times 10^6/^{\circ}C$ (from transformation temp to 960 $^{\circ}C$)	18.2	18.4	22.3 to 855 $^{\circ}C$ only.

Finally, enough experimental evidence was obtained to make reasonable estimates of the solidus temperatures of the U-Pu-Ti alloys. We see from Figure 8 that a titanium content of at least 20 a/o and no more than 10 to 15 a/o plutonium will result in alloys with reasonably high solidus temperatures. The combination, however, of both a low titanium content and a high plutonium content should be avoided.

Figure 8. Estimates of Solidus Temperature of U-Pu-Ti Alloys

A comparison of the solidus temperatures of the U-Pu-Ti alloys with those of the U-Pu-Zr alloys is made in Figure 9.

Figure 9. Solidus Temperatures of U-Pu-Ti and U-Pu-Zr Alloys

Compressive properties of U-15 w/o Pu-10 w/o Ti (see Progress Report for March 1965, ANL-7028, p. 22) have been supplemented with tensile tests at elevated temperatures. The ratio of compressive to tensile strength varies from 3:1 to 1:1, depending on whether the material is brittle or ductile in tension. Tensile ductility was appreciable only at temperatures greater than 750°C. (There was appreciable ductility in compression at temperatures as low as 600°C.)

U-15 w/o Pu-10 w/o Ti and U-18.5 w/o Pu-14.1 w/o Zr alloys have similar creep properties at 700°C (see Table XII). At 600°C the latter has the greater creep strength. The results are reported as the time required to reach 1% creep strain, including "first" and "second" stage creep strains. Since the same specimens were used for several temperatures and loads, the prior creep affected the results of the latter tests, but the trends are significant nevertheless. In both alloys the creep behavior was found to obey the following law reasonably well up to 1% strain:

$$\epsilon = at^n$$

where

ϵ = strain

a = a constant for a particular test

t = time

n = a constant dependent on stress and structure.

Table XII. Minimum Time to Attain 1% Creep Strain in U-Pu-X Alloys

Load (kg mm ⁻²)	U-15 w/o Pu-10 w/o Ti			U-18.5 w/o Pu-14.1 w/o Zr		
	600°C	650°C	700°C	600°C	675°C	700°C
0.28	-	210	64	-	94	70
0.42	170	35	30	2.97×10^5	27	32
0.56	120	30	17	2.95×10^5	13	12

The thermal conductivity of U-15 w/o Pu-8 w/o Ti alloy has been measured in the comparative apparatus from 200 to 800°C. The values obtained run about 10% less than for Inconel.

2. Jacket Materials

a. Vanadium Alloys

(i) Consolidation and Extrusion. Vanadium-20 w/o titanium (TV-20) alloy has a generally favorable combination of properties for use as a jacketing material for uranium-plutonium-fizzium fuel alloys, as well as for many other fast-reactor fuels, in case it is necessary to select these fuel alloys for use in fast breeder reactor fuel elements. Additional 10-cm-diameter TV-20 arc-cast ingots (AM37 and AM41) have been recently consolidated at ANL. The ingots were machined, canned and extruded to bar stock. A total of 12 billets for re-extrusion were obtained from the two ingots. Five of the billets obtained from arc-cast ingot AM37 have been extruded to tube-blanks. After dejacketing, the tubes were given one light swage pass over a hard mandrel and vacuum annealed. The present tube-blank size is 2.03 cm OD x 1.52 cm ID (0.80 in. OD x 0.60 in. ID) by about 100 cm (40 in.) in length. One tube will be fabricated to 0.76 cm OD x 0.05 cm wall (0.297 in. OD x 0.020 in. wall) tubing at ANL; the remaining four tubes will be processed by a commercial vendor to tubing of various sizes.

A total of six billets for tube-blank extrusion and one solid billet for sheet-bar extrusion were realized from arc-cast ingot AM41.

(ii) Reclamation of Solid V-20 w/o Ti (TV-20) Scrap Material. Some of the largest scrap pieces generated during processing of TV-20 materials (ingot croppings and extrusion tailings) have been reprocessed into drip rods and electron-beam (EB) remelted into 5.8-cm-diameter ingots. No abnormalities occurred during melting, although the ingots contained porosity, which has been typical of EB-melted ingots. Analyses for titanium and interstitial elements are being made to determine if contamination or preferential loss of titanium occurred. One section of this reclaimed material has been extruded to rod stock and cold rolled to a 0.79-cm diameter. The material behaved in a normal fashion during fabrication.

(iii) Mechanical Properties of Vanadium Alloys. Strain-time curves for V-Ti and V-Ti-Cr alloys are shown in Figure 10 for tensile creep tests in a vacuum of $<1 \times 10^{-6}$ mm Hg at 650°C and 15 kg mm⁻² stress. The sheet specimens were fabricated from small arc-cast buttons with the exception of the V-20 w/o Ti, which was fabricated from a large EB-melted ingot.

Figure 10. Creep of Annealed V-Ti and V-Ti-Cr Sheet
Alloys at 15 kg mm^{-2} and 650°C

From these data it appears that the 900°C anneal results in greater resistance to creep than a 700°C anneal. This is consistent with creep data previously reported (see Progress Report for September 1964, ANL-6944, p. 25) for recrystallized versus stress-relieved V-20 w/o Ti. The addition of 10 w/o chromium relative to 5 w/o also improves the creep resistance. The addition of 30 w/o Ti relative to 20 w/o may lower the creep resistance. This may be a result of testing material fabricated from a small arc-cast button in contrast to a large ingot. Future testing on V-Ti-Cr alloys with lower titanium content should resolve this question.

A comparison of the creep behavior of V-20 w/o Ti sheet and tube material at 650°C and 15 kg mm^{-2} stress is shown in Figure 11. Both specimens were made from material produced in a full-size melt and both were given a one-hour recrystallization anneal at 900°C before testing. The uniaxially stressed sheet specimen was tested in vacuum ($<1 \times 10^{-6} \text{ mm Hg}$) whereas the biaxially stressed tube specimen (internally pressurized with argon) was tested in an argon atmosphere ($40 \mu \text{ Hg}$). The difference in strain behavior is typical of the difference to be found between uniaxially and biaxially stressed material. Likewise, the total strain at rupture can be expected to be less in the tubular specimen than in the sheet specimen. Similar behavior has been reported for uniaxially and biaxially stressed Zircaloy-2,⁴ leading to the conclusion that creep data obtained for sheet material tends to be conservative when applied to tubes.

⁴Aungst, R. C., Stress Rupture Tests of Zircaloy-2 Pressure Tubes, BNWL-8 (Jan 1965).

Figure 11

Creep of Annealed V-20 w/o Ti Alloy at 650°C

(iv) Corrosion of Vanadium-Titanium Alloys. Alloys of vanadium containing from 10 to 90 w/o Ti have been exposed for one week to sodium (50 ppm oxygen) at 650°C. The average total metal penetration decreased with increasing titanium content, resulting in a minimum penetration at about 70-80 w/o Ti. However, starting at about 40 w/o, stringers of a second phase appeared with the same etching characteristics as the oxygen-hardened zone near the surface. These stringers extend much further into the metal than the average depth of the hardened surface zone and would represent a potential cracking hazard in many practical applications. Apparently, the region of practical titanium content is 0-40 w/o of titanium.

3. Carbide Fuel Elements

a. Fabrication of UC-PuC Irradiation Specimens. For fast breeder reactors vibratorily compacted uranium-plutonium mixed-carbide fuels appear to combine refractory characteristics suitable for use in the temperature range from 650 to 1100°C, to have thermal conductivity superior to oxide fuels of the same relative density, and to provide a reasonably simple means of remote-control fabrication. Programs to develop means of producing PuC and (U-Pu)C fuels, for the fabrication of carbide fuel-element specimens, and for testing them have therefore been under way for some time.

A project of loading fifteen mixed-carbide irradiation fuel rods by vibratory compaction has been nearly completed. To date twelve tubes have been loaded, ten decontaminated, and ten welded. All of the specimens have a bulk density in excess of 80% of theoretical. Radiographs indicate homogeneous density distribution with only the top 2.54 cm of the 35.56 cm of fuel column giving evidence of porosity, and that only at the extreme edge next to the cladding. Autoradiographs made of fuel pins containing the physical mixture UC (80 w/o coarse) + PuC (20 w/o fine) indicated minor inhomogeneity of plutonium in the first nine samples tested. This uneven distribution possibly related to a procedure for loading the coarse fraction in two or four stages. In subsequent UC + PuC loads the coarse-fraction UC

was loaded in one continuous pour from a shallow trough; autoradiographs showed improved homogeneity of the fine-fraction PuC.

One advantage of the one-pour coarse fraction loading is that the size limits of the two coarse fractions can be expanded from 12/14 and 30/35 to 12/20 and 30/50 mesh, respectively. Another advantage is that immediately after loading the fuel-column height exceeds the desired 35.56 cm by only 0.635 to 1.27 cm (compared with 5.08 cm to 8.89 cm before). Very little vibration time is required to bring the column to length, resulting in less self-attrition of the coarse fractions, and therefore larger and more uniform porosity to accept the subsequent fine-grained PuC infiltration. Autoradiographs of recently completed fuel pins show lesser activity in the top third of the fuel column, indicating incomplete filling of the pores between the coarse UC grains by the fine PuC. A test showed that 86% density could be achieved by continued loading of fine PuC.

Two operations in the above fabrication scheme that require the most man-hours of continuous attention are: (1) material crushing-sieving-weighing and (2) decontamination. Improvements in crushing technique and efficiency are imminent; a custom-designed jaw crusher has been built and crushing parameters are now being determined. At present grinding raw carbide stock gives a usable coarse fraction yield of only 30%, causing too much nonusable material, which must be recycled via time-consuming and tungsten-contaminating arc remelting. Selection of the optimum parameters for the jaw crusher should increase the original crushing yield twofold. A miniature continuous sieve has been designed and most parts for it received; this should replace tedious, repetitious hand-sieving operations. Time spent in weighing can be reduced only when weighings can be done in the same glovebox with other operations; at present time-consuming material transfers must be made to and from the weighing box.

Decontamination is still very time-consuming. The greatest delay occurs in the transfer (bagging out) of swab contamination samples to the monitoring box, and in the time spent in carefully handling the swab samples in the monitoring box to prevent contamination therein. Facilities to monitor swab specimens adjacent to the fuel pin decontamination stations are sorely needed.

During the past two months the high-temperature resistance furnace has been rebuilt completely, with new support plates, heater coil, radiant heat shields, and wall and floor insulation. A series of four out-gassing runs have been made. To date one carbide reaction run is in progress: $\text{UC}_2 + \text{Pu} \rightarrow 2(\text{U, Pu})\text{C}$. Results from this run are not yet available.

4. Fuel for Zero-power Reactors

a. Evaluation of ZPR-3 Elements for SEFOR Critical Experiments.

Methods for the fabrication, manufacture, and quality evaluation of critical test fuels for ZPR-3 are being developed (see Progress Report for March 1965, ANL-7028, p. 30). Present work is concerned with SEFOR test fuels.

The contract for manufacture of SEFOR critical test elements was awarded to Nuclear Materials and Equipment Corporation. The contractor has submitted core plates, components, and assembled elements for preproduction evaluation by ANL. He has also submitted copies of his manufacturing and quality control procedures.

In general, it has been possible to accept the quality of the core plates, jacket components, and assemblies subject to the following major comments:

- (1) The contractor should eliminate the use of BeO as a crucible wash. This resulted in beryllium contamination of the core plates and a 30% to 50% increase in neutron emission.
- (2) The analyses of the core plates should be brought within the specified limits.
- (3) Massive copper contamination of the welds from the welding chills should be eliminated.
- (4) Cleaning of all components before assembly should be improved.

The contractor has been able to revise his procedures to produce acceptable plates. The process was qualified on this basis and approved for production of fuel plates. Minor deviations from drawing dimensions, weld quality, and composition were noted during the course of the qualification examination. These were brought to the attention of the General Electric Corporation, which is supervising in-plant quality inspection.

During the reporting period the twin-differential bridge calorimeter has been placed in operation and several SEFOR samples have been checked. Sample power as indicated by the calorimeter is within 3% of calculated isotopic values. The instrument precision, i.e., ability to reproduce readings, needs to be improved. This appears to be a matter of operating technique and it appears that assay may be possible with somewhat greater precision.

Earlier problems of noise signals have been reduced by eliminating the bridge compensation circuit. Some method of compensating the slight differences in bridge resistance is necessary to operate in the automatic mode.

b. Properties of Zero-power Uranium-Plutonium-based Fuel Alloys.

We are now developing fuel alloys for ZPPR and for ZPR-6 critical experiments for future fast power breeder reactors. Flexibility and cost considerations have pointed up the need for alloys containing higher plutonium concentrations than the U-20 w/o Pu-2.5 w/o Mo SEFOR fuel alloy. Plutonium concentrations between 25 and 50 w/o Pu are of interest. Also, although molybdenum was found to satisfy most of the requirements of an alloying addition, there is room for improvement from a physics standpoint.

Air-corrosion and metallographic studies indicate that titanium, an element that is favored from physics considerations, may be a satisfactory alloying addition. Preliminary air-corrosion and expansion characteristics of cast U-Pu-Ti alloys were previously reported (see Progress Report for February 1965, ANL-7017, pp. 29 and 35). Chemical analyses of these alloys (of nominal composition U-25 a/o Pu-6 a/o Ti and U-25 a/o Pu-12 a/o Ti) have shown that they contain 5 and 10 a/o Ti rather than the given amounts.

The air-corrosion tests of these alloys have been continued. The 5 a/o Ti alloy began powdering very slightly by the ninth day. Although it did not show any weight gain until after seven weeks, it was about 10% powdered after five weeks and 50% powdered after ten weeks, at which time it had gained 2.0% in weight. After 13 weeks it had gained 4.9% in weight and was about 80% powdered.

The 10 a/o Ti alloy, subject to an identical air-corrosion test, has shown only a very slight surface powdering and a 0.2% weight gain after ten weeks. After 13 weeks it has gained 0.3% in weight and has shown no perceptible increase in powdering.

The encouraging air-corrosion test results for U-19 a/o Pu-3 a/o Ti (see ANL-7017, p. 35) apparently are counteracted by the increase of plutonium content to 25 a/o, even though the titanium content was raised to 5 a/o. We are now making U-25 a/o Pu alloys with titanium contents between 5 and 12 a/o in order to determine the lowest amount of titanium that will give increased corrosion resistance and will eliminate pyrophoric tendencies for this amount of plutonium.

Because estimates for ZPPR fuel requirements go as high as 3000 kg of contained plutonium, we are investigating rolling as potentially more economical than casting for fabricating the ZPPR alloys. Alloy castings with nominal compositions of U-25 a/o Pu-12 a/o Ti and of

U-24 a/o Pu-6 a/o Mo have been rolled after being preheated to 650°C. Both showed considerable plasticity at this temperature, but the rolling of both produced edge tears. The small sizes of the specimens made it difficult to maintain the desired rolling temperature, since the rolls can be heated only to a maximum of 180°C. We are presently making larger castings for further rolling studies with emphasis on eliminating edge cracking. Expansion characteristics in and across the direction of rolling will be measured to determine the degree of anisotropy developed as the result of rolling.

5. EBR-II Test Fuel Elements

A total of 80 EBR-II test fuel rods, 16 EBR-II-type TREAT test specimens, and three CP-5 test specimens are presently in fabrication. The 80 EBR-II rods are of four compositions: U-10 w/o Pu-10 w/o Fz, U-15 w/o Pu-10 w/o Fz, U-15 w/o Pu-10 w/o Zr, and U-15 w/o Pu-10 w/o Ti. These compositions are being jacketed in TV-20 jackets with varying sodium annuli and void volumes, and will be irradiated in the MTR and in the EBR-II. Four specimens of each alloy are being fabricated for transient tests. The three CP-5 specimens, all U-15 w/o Pu-10 w/o Zr, will be jacketed in stainless steel to examine compatibility of this alloy with the jackets under irradiation.

One problem in making specimens for test in these three reactors is that each requires a different degree of U^{235} enrichment, and those for EBR-II may require additional adjustment of enrichment to achieve a desired experimental fuel temperature. In order that results may be compared, it was necessary to make several melts of each composition with the following enrichments: 93 w/o U^{235} , 15.5 w/o U^{235} , 7.7 w/o U^{235} , and depleted uranium; and to maintain all other fabrication parameters as nearly constant as possible. With the variation in alloy and isotopic composition, a total of 22 melts were required to make the specimens.

All 99 required fuel pins have been cast, machined to length, and inspected. Forty of the EBR-II test rods have been assembled, sodium bonded, welded, leak detected, and inspected. Measurement of the sodium required caused some difficulty. Very careful volumetric measurements were required of each tube and fuel specimen. Of the 40 specimens loaded, 38 now contain sodium at the specified level. Bond-inspection eddy-current traces are being evaluated for these rods.

The remaining fuel rods are in varying stages of assembly.

6. Corrosion of Fuel-cladding Materials for Sodium-cooled Reactors

Analysis of sodium that has been hot-gettered with zirconium at 650°C has consistently given values of 40 to 50 ppm by weight of oxygen.

(The samples are withdrawn at 650°C in stainless steel sampling tubes.) The oxygen content of the hot-gettered sodium should be extremely low (<5 ppm).

A possible cause of the discrepancy between the expected and experimental values is the reduction of the oxide film invariably present on stainless steel by pure sodium at 650°C. This problem has not been encountered when the oxygen content of the sodium is actually in the 50-ppm range. In such cases equilibrium solubility data and analytical results are in good agreement.

The problem is being attacked in a variety of ways within the framework of the above hypothesis. Sodium was filled into the system at 175°C through a 5- μ filter. A sodium sample was withdrawn in the standard stainless steel tube at 150°C. The equilibrium oxygen solubilities at 175°C and 150°C are 14 ppm and 7 ppm, respectively. (It is not definitely known that enough time elapsed between filling at 175°C and sampling at 150°C to permit attainment of equilibrium at 150°C.) Analysis of the sample yielded 12 ppm.

The temperature of the system was then raised to 650°C. A sodium sample was taken in the standard welded sample tube of Type 304 stainless steel. In order to exaggerate any effect of a surface oxide film the weld temper film at the base of the tube was not removed before sampling. Duplicate analytical samples gave results of 95 and 104 ppm oxygen.

Unalloyed nickel sample tubes have been fabricated and will be used in an attempt to eliminate the problem apparently caused by the surface oxide film on stainless steel. An alternative method is to employ means of circulating an appreciable quantity of sodium through the tube before sample collection. The oxygen pickup from the walls of the tube is insignificant when related to the system volume.

7. Extrusion Development

Because of the growing demand for fuel alloy in the form of bar stock, an investigation was initiated to determine the feasibility of producing U-Pu fuel alloy bar stock by extrusion. The extrusion press may have two advantages over the presently existing rolling equipment: (1) fewer heating operations are required, and (2) closer control of side spread may be maintained. A set of dies, consisting of a die block, a backup block, and a bolster, has been designed and constructed for extruding bars 6.4 x 51 mm in cross section.

Small resistance-heated extrusion tools have been designed for use in a vertical 250-ton press so as to produce a single fuel pin by indirect extrusion. The extrusion container and ram have been constructed. The base, the die, and the shroud are partially completed.

E. General Fast Reactor Fuel Reprocessing Development

1. Skull Reclamation Process

The skull reclamation process, being developed as an auxiliary process in the EBR-II fuel cycle to recover and purify residual uranium left in melt-refining crucibles, is being studied on a pilot-plant scale of 1.5 kg of uranium. The pilot-plant demonstrations are expected to be completed soon; the development of full-scale, remotely operated equipment is under way.

In the most recent experiments, emphasis has been placed on the development of techniques which would reduce uranium losses in the waste streams. Uranium losses due to entrainment in the metal supernatant during its removal following precipitation of the uranium as a zinc-uranium intermetallic compound have been of particular concern (see Progress Report for April 1965, ANL-7045, p. 22); the losses have now been reduced to a low level (0.6 to 1.9%) by techniques which produce a well-settled bed and decrease the rate of supernatant metal transfer, and by the use of a strainer-weir attached to the inlet end of the transfer line.

When skulls from highly irradiated, short-cooled fuels are processed, fission product heating may be sufficiently great that it may not be possible to freeze the flux prior to removal of a zinc solution containing noble metal fission products. Therefore, removal of the zinc from beneath a molten salt was attempted. More than 90% of the zinc was removed from the molten salt at a transfer temperature of 750°C. The operation went so well that it will probably become the regular procedure, since considerable time is saved by eliminating the freezing of the flux.

2. Materials and Equipment Evaluation

a. Corrosion Resistance of Iron-Chromium Alloys. One of the processes currently under development for advanced reactor fuels utilizes cadmium and halide salts. Magnesium and zinc are added to the cadmium during the process. It has been shown that Type 405 stainless steel possesses adequate resistance to corrosion by the process solutions to merit consideration as a material of construction for this process (see Progress Report for April 1965, ANL-7045, p. 23); however, less expensive and more easily fabricable metals are being sought. Accordingly, three iron-chromium alloys: (1) Type 430 stainless steel, (2) Type 1-JR steel (Type 410 stainless with 3% aluminum addition), and (3) Type 7-Mo (Type 329 stainless steel) were tested for corrosion resistance in a run conducted at 650°C for 24 hr in an 8l w/o Cd-1l w/o Zn-6 w/o Mg-2 w/o U/chloride salt system. Although slight attack was evident on all samples tested (maximum attack, about 1 mil), the corrosion rate was sufficiently low to warrant additional testing of all three materials.

3. Removal of Nitrogen from Argon

Experimental work on the removal of nitrogen from argon by gettering the nitrogen on hot titanium sponge has been completed. Although this method has general utility for purifying argon for such applications as glovebox atmospheres, the major incentive for its development has been the possible need to remove nitrogen from the atmosphere in the Argon Cell of the EBR-II Fuel Cycle Facility.

Analytical results from a purification run conducted at 800°C with 5000 ppm nitrogen in argon indicate that for reaction times up to about 25 hr the reaction rate can be approximated by a parabolic rate equation which yields an Arrhenius activation energy of 27.4 kcal/mole. However, the experimental data are best fit by an asymptotic equation of the empirical form.

$$\frac{C^* - C}{C} = K_1 p^{-K_2} t^{-K_3},$$

where

C^* = asymptotic value of conversion of titanium to titanium mononitride, percent

C = conversion of titanium to the mononitride at time t , percent

p = partial pressure of nitrogen in the argon-nitrogen mixture, mm Hg.

This equation is typical of a reaction mechanism in which the reaction rate is controlled by alternate cracking and healing of the reacting film.

4. Halide Slagging Studies

Melt refining is being used currently to reprocess fuel discharged from the first core loading of EBR-II. Although development work on the melt-refining process has been completed, a modification of melt refining that involves the use of a chloride flux (halide slagging) has been investigated further. In the halide slagging process, the lanthanons and other electropositive fission products would be extracted from molten fuel alloy into an overlying CaCl_2 flux containing a small amount of MgCl_2 which serves as an oxidant. Plutonium would then be extracted from the molten alloy by a second CaCl_2 flux containing UCl_3 as the oxidant.

To provide supporting chemical data for the plutonium-extraction step, equilibrium constants have been determined for the reaction

The variables investigated were temperature (1150 and 1200°C), initial plutonium concentration in the charge (0.2 to 4.2 w/o), time (15 to 90 min), and UCl_3 :Pu ratio (~1 to 18). Average experimental values of the equilibrium constant were approximately 200 at 1150°C and 1200°C. Changes in initial plutonium concentration and UCl_3 :Pu ratio had no effect on equilibrium, which was attained in 15-30 min. This value is sufficiently high so that satisfactory separation of uranium from plutonium should be achievable in one equilibrium stage. The free energy changes for the reaction are -15.2 kcal/mole at 1150°C and -15.1 kcal/mole at 1200°C (assuming all activity coefficients to be unity).

Material balances ranged from 97.1 to 101% for plutonium and from 99.2 to 99.8% for uranium. In addition to the salt and metal phases, the material balances also include a thin layer of gray material formed between the crucible and the melt; this thin layer contained up to 11 w/o plutonium and 1 w/o uranium. It was assumed that this material did not affect the equilibrium between the salt and metal phases. Blank experiments in which the CaCl_2 flux contained no UCl_3 indicated that about 7 w/o of the plutonium had been separated from the liquid uranium. It is suspected that this effect might have resulted from reactions of plutonium with impurities in the uranium charge or crucible to form compounds that are insoluble in the liquid metal phase. The rate of reaction of the molten alloy with the beryllia crucibles used in these experiments (~150 $\mu\text{g}/\text{cm}^2$ in 45 min) was roughly greater by an order of magnitude in the present experiments with a flux present.

The halide slagging process has been developed on a laboratory scale to a point where it can be evaluated, along with other processes, for application to specific fuel-recovery problems. Possible uses include (1) removal of rare earth and alkaline earth elements from a metallic uranium-plutonium alloy and (2) separation of metallic plutonium and uranium. However, no further development of the process is planned until a specific need arises.

II. GENERAL REACTOR TECHNOLOGY

A. Experimental Reactor and Nuclear Physics

1. Nuclear Constants

a. Radiative Capture. Measurements of neutron cross sections of substances important to the fast reactor program are continuing. One of the nuclear reactions which absorbs neutrons unproductively in a reactor is radiative capture. The cross sections for radiative capture as a function of neutron energy are needed in order to make proper use of materials proposed for structural and control use in reactors. Further, the study of neutron capture allows one to test theories of nuclear reactions, and to derive information on the spacings and widths of nuclear energy levels.

The range of neutron energy of interest in fast reactors lies between about 0.1 keV and several MeV. In the present program, radiative capture cross sections of rhenium have been measured as a function of neutron energy between 4 keV and 2.6 MeV, covering most of the fast reactor region.

The basic experimental technique used in the capture work is the activation method, wherein the nucleus to be studied is exposed to a monoenergetic neutron beam, and the radioactive species produced thereby are counted with either a beta counter or a gamma-ray analyzer. The counting rate of the activation product is then converted to a capture cross section by suitable calibrations which yield the absolute neutron flux and the absolute disintegration rate of the capture product. The monoenergetic neutron beam is produced by the reaction ${}^7\text{Li}(p, n){}^7\text{Be}$ or $\text{T}(p, n){}^3\text{He}$ when lithium or tritium targets are bombarded by protons from the Van de Graaff accelerator. The neutron flux is measured with a fission chamber, which counts fissions in a thin deposit of uranium.

The data, consisting of the cross sections for the production of the 90-hr state of ${}^{186}\text{Re}$, and the 17-hr and 18.7-min states of ${}^{188}\text{Re}$, are shown in Figure 12. Earlier values of other investigators^{5,6} are also shown.

B. Theoretical Reactor Physics

1. Computing Techniques

The current trend toward larger computer memories has again brought the use of stored tables into prominence as an efficient method of computing functions. This is so even for functions of a complex variable, such as the (modified) complex error function $w(z)$, which appears in the Doppler-broadened resonance line shape function.⁷

⁵Hughes, D. J., Garth, R. C., and Levin, J. S., Phys. Rev. 91, 1423 (1953).

⁶Macklin, R. L., Lazar, N. H., and Lyon, W. S., Phys. Rev. 107, 504 (1957).

⁷O'Shea, D. M., and Thacher, H. C., Jr., Computation of Resonance Line Shape Functions, Trans. Am. Nucl. Soc. 6, 36 (June 1963).

Figure 12. Radiative Capture in Rhenium

In applications where speed rather than memory space is of paramount importance, it is clearly advantageous to perform as much of the processing as possible in advance, and to use finite-difference interpolation techniques with prestored differences, rather than the currently more popular Lagrangian methods. If, as in the case of $w(z)$, the function is analytic, it is important to take advantage of this fact.

Let the function to be interpolated be denoted by $w(z) \equiv u(x+iy) + iv(x+iy)$, and let it be tabulated at the points z_0 , and $z_0 \pm h \pm ih$. If we introduce the usual difference operators

$$\mu\delta_x f_0 = [f(z_0 + h) - f(z_0 - h)]/2; \quad (1)$$

$$\mu\delta_y f_0 = [f(z_0 + ih) - f(z_0 - ih)]/2; \quad (2)$$

$$\delta_{xx}^2 f_0 = f(z_0 + h) - 2f(z_0) + f(z_0 - h); \quad (3)$$

$$\delta_{yy}^2 f_0 = f(z_0 + ih) - 2f(z_0) + f(z_0 - ih); \quad (4)$$

then the following interpolation formula, which has not been found in the literature, appears to be highly efficient.

$$\begin{aligned} w(z_0 + ph + iqh) = & u_0 + iv_0 + \left[\frac{(\mu\delta_x u_0 + \mu\delta_y v_0) + i(\mu\delta_x v_0 - \mu\delta_y u_0)}{2} \right] \{p + iq\} \\ & + \left[\frac{(\delta_{xx}^2 u_0 - \delta_{yy}^2 u_0) + i(\delta_{xx}^2 v_0 - \delta_{yy}^2 v_0)}{4} \right] \{(p+q)(p-q) + i(2pq)\} \\ & + \left[\frac{(\mu\delta_x u_0 - \mu\delta_y v_0) + i(\mu\delta_x v_0 + \mu\delta_y u_0)}{2} \right] \{p(p^2 - 3q^2) + iq(3p^2 - q^2)\} \\ & + \left[\frac{(\delta_{xx}^2 u_0 + \delta_{yy}^2 u_0) + i(\delta_{xx}^2 v_0 + \delta_{yy}^2 v_0)}{4} \right] \{(p^4 - 6p^2q^2 + q^4) + i4pq(p^2 - q^2)\} + O(h^5). \end{aligned} \quad (5)$$

In application, the quantities in square brackets (or as many as required to give the required accuracy) would be stored along with the basic table. Only the quantities in braces would require evaluation for each new evaluation of z . The number of arithmetic operations required for the various numbers of terms to evaluate both u and v and either u or v are as follows:

Order	u or v			u and v			Maximum Relative Error*	
	A**	M**	E**	A**	M**	E**	u	v
1	2	2	0	4	4	0	0.407(-2)	0.839(-2)
2	6	6	1	10	10	1	0.249(-3)	0.621(-3)
3	10	12	1	16	18	1	0.696(-5)	0.333(-4)
4	13	17	2	21	25	2	0.223(-5)	0.661(-5)

*Tested on complex error function with $h = 1/8$, $z_0 = 0.5 + 0.5i$, $-1/2 < p$, $q < 1/2$, 20×20 grid; power of 10 in parentheses.

**A = number of additions.

**M = number of multiplications.

**E = number of exponent modifications (multiplications by 2).

From these results it appears likely that the third-order formula will give adequate accuracy for the complex error function at a very considerable saving in computation, not only compared with the computation from continued fractions, but also with a six-point bivariate interpolation.

2. Development of Reactor Computing Codes

An automated and consistent reactor computation system is being developed. Such a system will include a cross-section-preparation code, reactivity codes, a depletion code, etc. The effort is currently concerned with implementing that portion of the system which will generate multi-group cross sections. The code concerned is designated as MC².⁸

The MC² program calculates multigroup constants from a basic evaluated nuclear data file (ENDF). Given the user's input specifications, the program will form group cross sections for a large number of fine groups. These cross sections are then averaged over either a prespecified or a calculated weighting spectrum to produce the coarse group constants to be used in the neutronics portion of the computation system.

Doppler-broadened cross sections are calculated over the resolved resonance region, and the intermediate representation of Goldstein and Cohen⁹ is utilized in regard to the NR and NRIA approximations. Interference between resonance and potential scattering is included as is the interference due to overlapping resonances.

In the unresolved region, chi-squared distributions are used in the usual way to perform statistical averaging of the level parameters.

A modified Bell approximation for the Dancoff factor will be used with an energy-dependent scattering cross section to calculate effective escape cross sections for use with the equivalence principle.

A fast table lookup and interpolation routine will be included to facilitate obtaining the Doppler-broadened line shapes. This device will permit unresolved resonance calculations for all fine groups in the unresolved region. The present method, in which the continued-fraction expansions for determining the line shapes is used, is very time-consuming and only permits the calculation of cross sections for about five energy points in the unresolved region.

Cross sections which vary smoothly over energy are represented in the MC² library by the coordinates of end points of linear segments taken from graphs of $\ln E$ versus $\ln \sigma$, $\ln E$ versus σ , or E versus σ . Fine group cross sections are then obtained by interpolation over the fine group limits.

⁸Reactor Physics Division Annual Report - July 1, 1963 to June 30, 1964, ANL-7010 (Jan 1965).

⁹Goldstein, R., and Cohen, E. R., Theory of Resonance Absorption of Neutrons, Nucl. Sci. Eng. **13**, 132 (1962).

Inelastic scattering is computed for discrete levels when available; otherwise a statistical treatment involving energy-dependent average nuclear temperatures is used.

The elastic-scattering treatment of ELMOE will be used for the light elements. Additions to the program will be made to allow hydrogen scattering.

As in ELMOE, the weighting spectrum used in collapsing the fine group cross sections will be obtained from either a P1 approximation, or from a consistent P1 or B1 approximation.

The first version of MC² will prepare input to the XLIBIT cross-section library code.

Future developments with MC² will include: reading of smooth cross-section data at run time; automated updating of the MC² library from an ENDF such as that from Brookhaven; direct integration of the slowing-down integral equations without using NR and NRIA approximations in the resonance region; coupling of MC² with a thermalization code such as THERMOS; use of higher Pn and Bn approximations to generate the weighting spectrum.

3. Change in Nuclear Temperature to Account for Resolved Levels

The density of nuclear energy levels in the statistical region of inelastic neutron scattering is usually described by a continuous function of energy E . Sums over the states of the nucleus are replaced by integrals over the density function $r(E)$. The effective nuclear temperature τ at excitation energy u is the implicit solution of

$$\int_0^{\infty} E r(E) e^{-E/\tau} dE / \int_0^{\infty} r(E) e^{-E/\tau} dE = u.$$

To find the effect on τ of replacing the sums by the integrals, assume one level (E_1) is known and that u is high enough that a statistical fit is good at energies above E_1 . Finally, suppose

$$r(E) = c(\exp BE)/\sqrt{E},$$

where B and c are empirical constants. In the discrete plus continuum model, replace $r(E)$ by

$$N_1 \delta(E - E_1) + E(E - E_1) r(E),$$

where $\delta(x)$ is the Dirac delta-function and

$E(x) = 1$ for $x \geq 0$, and 0 for $x < 0$;

replace τ by τ' .

With use of typical heavy-atom values: $B = 1$, $E_1 = 0.05$, $\tau = 0.5$, we find $\tau' = 0.82\tau$. This is a sizeable change which will be further investigated by optical model methods.

4. Resonance Interference

A program has been written for the CDC-3600 to compute resonance absorption of neutrons by mixtures. An energy range of interest is divided into many narrow intervals of equal lethargy width, each width so narrow that neutrons will suffer only one collision in the interval. Consequently, the collision rate in the interval is identical with the slowing-down source.

The slowing-down source for the k th interval has the form

$$S_k = \sum_{j=1}^J \sum_{\ell=1}^{L_j} P_{\ell j} \Sigma_{s_{k-\ell,j}} \phi_{k-\ell}, \quad (1)$$

where

$P_{\ell j} \equiv$ probability that a neutron is scattered down ℓ groups in a scattering collision with material j ;

$L_j \equiv$ maximum number of groups through which a neutron can be downscattered in a collision with material j ;

$\Sigma_{s_{k-\ell,j}} \equiv$ scattering cross section of material j in group $k-\ell$;

$\phi_{k-\ell} \equiv$ neutron flux in group $k-\ell$.

Equation (1) would be a time-consuming expression to use because a neutron can be scattered through so many groups. Since $P_{\ell} = e^{-\Delta u} P_{\ell-1}$, where Δu is the width of an interval, Eq. (1) can be reduced to the simpler form

$$S_k = \sum_{j=1}^J P_{1j} \Sigma_{s_{k-1,j}} \phi_{k-1} + e^{-\Delta u} \left[S_{k-1} - \sum_{j=1}^J P_{L_j} \Sigma_{s_{k-L_j,j}} \phi_{k-L_j-1} \right]. \quad (2)$$

The program stores information for 2500 intervals. The only approximation made in using Eq. (2) is that for materials with $L_j > 2500$, the asymptotic flux is used for ϕ_{k-L_j-1} .

5. Calculation of Pile-up Distortion

In many experiments the existence of a high background of undesirable radiation may be detected when the detection of relatively rare events, or other types of radiation, is actually desired. In such circumstances, reasonably accurate values of the pile-up count rate and distortion may be required.

By convention, the distribution of the maxima in response amplitude (or the distribution of peak values) is called the pulse-height distribution of the detector.

A distinction must be made between the two different types of pulse-height distributions to be considered: those which contain pile-up distortion and those which do not. These distributions will be designated as actual and ideal pulse-height distributions, respectively. An actual pulse-height distribution is one that arises from the output of a detector and therefore inherently contains pile-up distortion. In contrast, an ideal pulse-height distribution cannot occur in practice. It may be regarded as that pulse-height distribution which is obtained from a detection system in the limit of vanishing count rate.

The actual pulse-height distribution $D(\nu)$ can be expressed in terms of the ideal pulse-height distribution $P(\nu)$ by an integral representation of the form

$$D(\nu) = \int_0^{\infty} B(\nu') P(\nu - \nu') d\nu'. \quad (1)$$

The function $B(\nu')$ in Eq. (1) depends upon the ideal pulse-height distribution, the pulse shape, and the counting rate.

A special case has been studied in which the ideal pulse-height distribution is exponential:

$$P(\nu) = e^{-\nu} \quad (2)$$

and the pulse shape is square

$$\begin{aligned} F(t, \tau) &= 1, & 0 \leq t \leq \tau; \\ F(t, \tau) &= 0, & t < 0, t > \tau. \end{aligned} \quad (3)$$

(The parameter τ is customarily called the resolving time of the detector.) For this special case, the integral representation of $D(\nu)$ given in Eq. (1) reduces to

$$D(\nu) = \frac{2}{\pi} \int_0^{\infty} \left[\frac{\cos(\nu\xi) + \sin(\nu\xi) - e^{-\nu}}{1 + \xi^2} \right] \exp\left(\frac{-\alpha\xi^2}{1 + \xi^2}\right) \cos\left(\frac{\alpha\xi}{1 + \xi^2}\right) d\xi, \quad (4)$$

where

$$\alpha = N\tau; \quad (5)$$

N = average counting rate.

Computer programs have been developed to evaluate this special case. Figure 13 displays $D(\nu)$ for $\alpha = 1.0, 0.5$, and 0.10 in comparison with $P(\nu) = e^{-\nu}$. It is interesting to note that the general condition

$$\lim_{\alpha \rightarrow \infty} D(\nu) = P(\nu)$$

is satisfied by the numerical results obtained for this special case.

Figure 13
Comparison of the Actual Pulse-height
Distribution $D(\nu)$ for $\alpha = 1.0, 0.5$, and
 0.10 in the Region $0 \leq \nu \leq 4$ with the
Ideal Pulse-height Distribution
 $P(\nu) = e^{-\nu}$

6. ZPR-7 Data Analysis

The cross sections from the GAM-I output and the thermal cross sections obtained previously from the THERMOS code were used to calculate the critical bucklings for all BORAX-V and Hi-C uniform lattices measured in the ZPR-7 critical assembly. Pl and B1 approximations

were used in running the GAM-I problems, which were based upon three fast groups and the Hanford cross sections for U^{235} and U^{238} . Reactivities obtained from these bucklings were generally less than one percent above the experimental values obtained when using the P1 approximation in solving GAM-I problems. This represents a considerable improvement over the results obtained when using the U^{235} and U^{238} cross sections obtained from the original GAM-I library.¹⁰ Most of the improvement is attributable to the changes in U^{238} cross sections. A similar result was found in analyzing the Yankee critical experiments.¹¹

C. High-temperature Materials

1. Ceramics

a. UC-PuC Bodies. Precipitated uranium carbide normally is composed of UC plus various amounts of free carbon. Compensation for the free carbon with an addition of UH_3 should result in a material that can be reaction-sintered to a near stoichiometric UC. After thorough ball-milling of the hydride-and-carbide mix, four different heats were run in vacuum. Specimens were heated with intermediate soaks at 1100, 1200, and 1300°C followed by a final heat at 1550°C. A fourth set of specimens were rapidly heated to 1650°C after decomposition of the hydrides. The specimens heated to 1650°C had the best density, only 80% of theoretical. The lack of success in reaction sintering to a higher density may be associated with the quality of the hydride, which was old and probably contained oxide. Fresh hydride will be prepared and the experiment will be tried again.

b. Properties of (Th, U) Phosphides. Studies of the system UP-UN have continued. Approximately 0.6 w/o UN entered the UP lattice (see Progress Report for March 1965, ANL-7028, p. 44). A small amount of UP (about 0.3 w/o) entered the UN lattice, producing a slight dilation. The near absence of solid solubility in the system is due to the large size differential between the radii of the two nonmetal atoms, 1.57 Å for nitrogen versus 1.92 Å for phosphorus. This corresponds to a difference of -18.4% when UP is the solvent compound and +22.6% when UN is the host compound.

The phase diagram of the system UP-UN is relatively straightforward. Solid solubility is very restricted and no intermediate compound forms. A eutectic occurs at a composition of about 40 w/o UP-60 w/o UN and a temperature of $2280 \pm 40^\circ\text{C}$.

Tungsten dissolves readily in molten UP. The melting point of UP sealed in a tungsten crucible was found to be $2610 \pm 20^\circ\text{C}$. About 40 w/o of tungsten metal dissolved in the UP, only to be almost completely rejected

¹⁰ Reactor Physics Division Annual Report--July 1, 1963 to June 30, 1964, ANL-7010, (Jan 1965) p. 45.

¹¹ Moon, D. P., Consistent Heavy-atom Cross-section Evaluation for Plutonium Recycle Calculations, ANL-6972 (Dec 1964).

on cooling. Some diffuseness in the X-ray photograph of UP suggests slight residual solubility of the metal. No intermediate compound was found in the system W-UP.

c. Mechanical Properties of Uranium Compounds. A study of the mechanical properties of uranium sulfide and phosphide was started. Preliminary results are given in Table XIII.

Table XIII. Uranium Sulfide and Phosphide Tested in Vacuum at Room Temperature

Number of Samples	Density (g/cc)		Rupture Stress (kg/cm ²)		Mean Load Rate (g/sec)	Mean Strain Rate (mm/sec x 10 ⁴)	Total Deflection (mm)
	Mean	Standard Deviation	Mean	Standard Deviation			
Sulfide							
4	10.15	0.98	445.5	73.7	317	0.12	0.14
3	10.06	0.01	516.2	53.43	299	0.11	0.13
Phosphide							
2	9.7	-	1099	46.64	222	0.08	0.20

The sulfide examined was coarse grained, contained UOS in the boundaries, and was not particularly strong. It broke with a coarse cleavage fracture at room temperature, but showed similar ductility to UO₂ under the same conditions.

The phosphide was fine grained, and strongly resembled UO₂ in appearance. The strength was high in good samples, but a number broke unexpectedly after yielding. All samples showed a yield point and considerable ductility compared with UO₂ or US. This may not be so evident when the deflection data are presented in millimeters.

Further work is proceeding to improve the structure of the sulfide and to make consistently good samples of phosphide.

d. Thermal Stability. Work continued on the study of the oxidation and evaporation behavior of plutonium ceramics. The construction of glovebox apparatus for these studies is in progress.

Additional data were obtained on the oxidation of uranium monosulfide. Figure 14 shows a cross section of the product layer on a US pellet reacted for one hour at 700°C in an atmosphere of helium containing about 0.1% oxygen. The specimen was polished with 1- μ diamond paste and etched with a sulfuric acid-peroxide solution. Four distinct product layers are visible. For the sake of discussion, these will be called numbers 1 through 4, starting with the outer layer.

Figure 14. Cross Section of the Product Layer on a US Pellet
Oxidized for One Hour at 700°C in Helium
Containing 0.1% of Oxygen (130X)

The outer layer was identified as UO_2 by X-ray diffraction. Although layer number 2 appears distinctly different, it has not yet been identified. The black appearance of layer number 3 is a result of severe erosion by the etchant. The composition of this layer was determined to be αUS_2 , which is known to be soluble in dilute acids.¹² The composition of the inner layer, 4, is still unknown.

The presence of US_2 in the product layer is consistent with the previously reported observation that a weight gain occurred during oxidation (see Progress Report for April 1965, ANL-7045, pp. 21-29). The data obtained thus far strongly indicate that the mechanism for US oxidation is similar to those established for UP and UN when higher phosphides and nitrides are formed during oxidation.¹³

Figure 15 shows an unetched section of the product layer formed on a US specimen reacted for one hour at 700°C in helium saturated with water vapor. Although this specimen also gained weight during oxidation, the only product species identified so far is UO_2 .

¹²Dell, R. M., and Allbutt, M., The Nitrides and Sulphides of Uranium, Thorium and Plutonium: A Review of Present Knowledge, AERE-R-4253 (1963).

¹³Baskin, Y., Oxidation Behavior of Uranium Monophosphide, J. Am. Cer. Soc. 48(3), 153-156 (1965).

Figure 15. Cross Section of the Product Layer on a US Pellet
Reacted for One Hour at 700°C in Helium
Saturated with Water Vapor (130X)

Attempts will be made to improve the techniques for removing product layers so that the oxidation products can be more completely identified. Reaction-rate determinations by gas chromatographic analysis will be initiated soon.

e. Anelasticity of Some Uranium Compounds. Table XIV shows the measured effect of density on the elastic modulus of UO_2 at room temperature. Although the range covered was small, there was a clear trend toward larger values of the elastic modulus as the density was increased.

The determination of the elastic modulus and internal friction did not alter the characteristics of the same, making it possible to determine the effect of grain size by heat treating the samples after each modulus determination. The results indicated that an increase in the grain size produced a decrease in the internal friction, but did not affect the Young's modulus.

The change of elastic modulus and internal friction with variations in the O/U ratio (see Progress Report for April 1965, ANL-7045, p. 29) is shown in Table XV. Metallographic studies made in

a specimen treated in a similar way to those used for the measurements, showed the presence of many microcracks. An increase in the number of etch pits was also observed, probably indicating an increase in the number of dislocations. The specimen with an O/U ratio of 2.02 showed a second phase which was unevenly distributed. This phase was not detected when the O/U ratio was 2.05.

Table XIV. Effect of Density on the Elastic Modulus of UO_2 at Room Temperature

Density (gm/cc)	$E_f^{(1)}$ (kilobar) ⁽⁴⁾	$E_e^{(2)}$ (kilobar)	$E_{av}^{(3)}$ (kilobar)
10.54	1980	2020	2000
10.55	2100	2070	2085
10.56	2065	2040	2052
10.57	2090	2070	2080
10.58	2035	2060	2048
10.59	2140	2090	2115
10.62	2110	2125	2117
10.72	2130	2180	2155
10.73	2160	2135	2147

(1) Young's modulus calculated from the flatwise flexural vibration.

(2) Young's modulus calculated from the edgewise flexural vibration.

(3) Average of E_f and E_e .

(4) Kilobar is equal to 10^9 dynes/cm².

Table XV. Effect of O/U Ratio on the Elastic Modulus and Internal Friction of Uranium Oxide at Room Temperature

Sample 30-53				Sample 32-55			
O/U	E_{av} (kilobar)	Percent Decrease	$Q^{-1}(\times 10^4)$	O/U	E_{av} (kilobar)	Percent Decrease	$Q^{-1}(\times 10^4)$
2.005	1921	-	1.01	2.005	2102	-	1.17
2.025	1763	8.2	-	2.02	1934	7.9	-
2.052	1404	26.9	38.5	2.049	1445	31.2	35.5

2. Liquid-Metal Corrosion

a. Polarization Studies. Anodic-polarization experiments with unalloyed zirconium rod samples in oxygenated sodium have been continued. Transfer of sodium to the cell has been done in a helium dry box, the helium moisture level being in the 1 ppm range. A higher moisture level exists in argon cover gas admitted subsequently to the cell, and this is to be reduced. However, corrosion film surfaces without the

troublesome, low-resistance-spot areas observed previously are consistently produced in overnight exposures in 540°C sodium after loading the sodium in this manner.

An apparent problem area persists at the sodium-argon interface in the cell, where a preoxidized zirconium rod surface extends through the sodium surface. The preoxidized region reproducibly displays a localized, low-resistance area in this region, but not elsewhere. Investigation of this phenomenon is continuing.

b. Dissolution Kinetics. Dissolution rates of tantalum in liquid tin have been determined at five temperatures between 800 and 1100°C at a speed of 10.0 rpm in rotating-disc experiments.

Trace amounts of oxygen contamination introduced into the argon cover gas during sampling of the liquid bath affected the results in one of the dissolution runs. This problem has necessitated the construction of a better gas-purification system and a modification of the sampling method. The next series of experimental runs will concentrate upon the effect of velocity on dissolution rates.

c. Lithium-corrosion Studies at Elevated Temperatures. Nitrogen is one of the major impurities in lithium. The solubility of nitrogen in lithium at 250 and 300°C has been reported to be 1290 and 1830 ppm, respectively. No information is available at higher temperatures. The presence of lithium nitride in lithium has been related to a considerable decrease in ductility of steel exposed to lithium (ORNL-2924). It has also been reported that the solubility of molybdenum in liquid lithium is highly dependent upon the concentration of nitrogen between 650 and 930°C (Acta Metallurgica 9, 519-520). The corrosive effect of nitride in lithium on molybdenum and other refractory metals at temperatures of interest has not yet been established.

Work on the lithium corrosion of tantalum and molybdenum at 1200°C for periods of 8-9 days indicated an increase in nitrogen content of the lithium in contact with molybdenum from 40 to 175 ppm of nitrogen. However, there was a decrease of nitrogen in the lithium to 5 ppm after test in tantalum capsules. The mode of corrosion attack on the tantalum was both intergranular and transgranular. There was only slight solution attack of the molybdenum. Detailed metallographic and microhardness examinations are in progress.

Sufficiently accurate data on the free energy of formation of these nitrides are not available. Standard free energies of reactions of tantalum and molybdenum with lithium nitride calculated from standard free energies of formation indicated -14.9 kcal/g-atom of nitrogen for TaN and +27.0 kcal/g-atom of nitrogen for Mo₂N. The driving forces for

these two reactions are opposite in direction. In view of the nitrogen analyses of lithium given above, it is apparent that tantalum was effective in gettering nitrogen from lithium, whereas lithium gettered nitrogen from molybdenum. The highly selective attack on tantalum makes it reasonable to believe that concentrated interstitial compounds, such as nitrides, may be present in the area of attack. The nitrogen concentrations in lithium of 1.75 ppm seemed to have no effect on the corrosion of molybdenum at 1200°C.

d. Corrosion by Sodium at Elevated Temperatures. The exposure of materials and components to sodium at 1200°F containing a known and controlled oxide content is pertinent to the development of future fast power reactors. In a surveillance loop, comparative performance of commercially available equipment can be evaluated in a common environment. Such a loop is being constructed. Piping, insulation, and the container tray are available. Portions of the piping will be sampled to supply "as received" specimens for metallurgical examination. The construction of the EM pump, which is the major long lead item, is 20% complete.

A preliminary design of a device for wear testing of materials in the environment of the loop has been completed. The device consists of a block and shaft test specimen suspended in the flowing sodium stream. Arrangements are being included which will allow the imposition of a constant measured force upon the specimen as the test proceeds.

Commercially available instruments and equipment for the above loop will be purchased to serve a dual function; as a part of the instrumentation evaluation program, and as measurement and control devices. An oxygen analyzer and a water-content monitor for the cover gas will be installed.

An ultrasonic hardness tester is being purchased to permit "in situ" hardness determinations on portions of components exposed in the sodium loop. The device produces minimal deformation of the tested material as compared to conventional hardness testers. As a consequence, a large number of measurements can be made over a small area.

D. Other Reactor Fuels and Materials Development

1. Nondestructive Testing

a. Determination of Elastic Moduli of High-temperature Materials by Ultrasonics. Room-temperature ultrasonic measurements with a tungsten rod machined to specified dimensions indicated that the amount of threading on the large diameter part of the rod (some parts of which were left smooth to accommodate O-ring seals for vacuum and water jacket) was sufficient to eliminate trailing pulses from that region of the rod. However, it was discovered that the echo from the shoulder between the large part of the rod and the index region was too small, so the index region is now being machined to a smaller diameter, thus providing a larger shoulder area. The index diameter was originally chosen larger because it was desired to separate trailing pulses which would be generated in each of the two rod sections of different diameter; now that it is known that no trailing pulses can be seen from the large part of the rod, we can redesign the bar for better performance.

A mirror system designed for use in temperature measurements in the Brew furnace has been tested and found to work very well. The optical path from sample to optical pyrometer has been calibrated against a standard lamp. Measurements of thermal gradients in a stainless steel dummy sample rod are now being made. When the remachined tungsten rod has been received, it will first be measured in the regular furnace to 1100°C, then it will be placed in the Brew furnace and measured to as high a temperature as possible.

Table XVI is a revision and an extension of the table presented last month (see Progress Report for April 1965, ANL-7045, p. 32, Table XI).

Table XVI. Room-temperature Elastic Properties

Material	Longitudinal Wave Velocity (10 ⁵ cm/sec)	Shear Wave Velocity (10 ⁵ cm/sec)	Density (g/cm ³)	Young's Modulus (10 ¹² dyne/cm ²)	Shear Modulus (10 ¹² dyne/cm ²)	Poisson's Ratio
W-25 w/o Re (vacuum arc cast; rolled)	5.31	2.93	19.6	4.29	1.68	0.283
W-25 w/o Re (powder metallurgy; pressed, sintered, rolled)	5.24	2.95	19.6	4.34	1.71	0.269
Re (rolled sheet)	5.63	2.91	20.99	4.68	1.78	0.318
UO ₂	5.52	2.81	10.63	2.22	0.839	0.326
				Acoustic Impedance, Longitudinal Waves [10 ⁵ g/(cm ² -sec)]	Longitudinal Attenuation (db/μsec)	Longitudinal Attenuation (db/cm)
UO ₂	5.52	-	10.63	58.5	18.5	33
Araldite 502 Epoxy	2.62	-	1.18	3.09	-	-
Araldite 502 Fiberglass-loaded	2.60	-	1.19	3.09	14	54
Araldite 502 Epoxy, Silver-loaded	1.67	-	2.70	4.50	14	84

b. Development of a Neutron-image Intensification System. A resolution analysis of a neutron-imaging system has been completed. The theoretical resolution limitation imposed both by the thickness of the input target of the present intensifier tube and by the minimum separation obtainable between an inspection object and the input target is in reasonable agreement with the experimentally observed high-contrast resolution of 0.5 mm. The television system as now used does not seem to limit the resolution of the system.

The new neutron-image intensifier tube, containing a thinner input target and decreased spacing between the target and the tube window, should permit a high-contrast resolution capability of about 0.25 mm. Some modification of the television system will probably be necessary in order to achieve this resolution through the neutron television system. A delivery date for the new tube has not yet been established.

The present intensifier tube will be used to observe thermal-neutron images obtained from several Cm-Be neutron sources. Initial studies will be made with an assembly of 12 sources in a water moderator tank. It is hoped that the thermal-neutron intensity obtained through a centrally located beam opening will be sufficient so that the intensifier can be used to optimize the locations of the sources within the moderator.

c. Infrared Systems for Nondestructive Testing. Movement of the chopping motor of the infrared-detection system farther from its former position close to the indium antimonide detector has yielded an improved and more stable output. This separation of the motor and detector has reduced the heating effect of the motor on the detector. Temperature differences as small as 0.2°C at an object temperature of 95°C now appear readily detectable.

d. Thermal Conductivity of Irradiated Fuel as a Function of Burnup and Temperature. Additional measurements have been made with uranium-fissium alloys (see Progress Report for April 1965, ANL-7045, p. 33). Rather than making a number of runs on one specimen each 2.54-cm long fuel pin was alternately sectioned in longitudinal and transverse cuts and measurements made on each section. The results reported in Table XVII have been corrected to account for the effect of the finite pulse duration on the specimens whose half time, $t_{1/2}$ (time for the back surface temperature to reach one-half of its maximum rise), is ≤ 25 ms. The correction factor is not needed for longer half times. Values are $\pm 10\%$.

2. Irradiation Testing

a. Delayed-failure Hydrogen Embrittlement and Irradiation Effects in High-strength Steels. The presence of hydrogen in a high-strength steel can lead to the phenomenon of delayed failure, generally referred to as

hydrogen embrittlement. With appropriate combinations of hydrogen content, temperature, and presence of stress concentrations, failure can occur under dead-weight loading at less than half the yield stress and in a period of hours. It is generally believed that the hydrogen must be in solution in the iron lattice for this phenomenon to occur, and during an incubation period it diffuses preferentially to regions of highest triaxial stress. There it concentrates and reduces the strength of the material. Catastrophic failure follows.

Table XVII. Thermal-Diffusivity and Conductivity Measurements of Various Fuel Alloys

Alloy	Section	$\alpha(\text{cm}^2/\text{sec})$	$K\left(\frac{\text{cal-cm}}{\text{cm}^2\text{-sec-}^\circ\text{C}}\right)$	$K\left(\frac{\text{cal-cm}}{\text{cm}^2\text{-sec-}^\circ\text{C}}\right)^{**}$
U + 3.3 w/o Fs + 1.7 w/o Zr; 10% Enriched	First	0.0794	0.0550	
	Transverse			
	First	0.0831	0.0540	
	Longitudinal			
	Second	0.0816	0.0560	
	Transverse			
U + 3.0 w/o Fs U + 5 w/o Fs; 20% Enriched	Third	0.0818	0.0554	
	Transverse			
				0.0541
	First	0.0601	0.0454	
	Transverse			
	First	0.0620	0.0440	
U + 5 w/o Fs U + 5 w/o Fs + 2.5 Zr; 20% Enriched	Longitudinal			
	Second	0.0631	0.0475	
	Transverse			
	Third	0.0628	0.0451	
	Transverse			
				0.0450
U + 5 w/o Fs + 2.25 w/o Zr U + 6.7 w/o Fs + 1.3 w/o Zr; 10% Enriched	First	0.0721	0.0516	
	Transverse			
	First	0.0752	0.0509	
	Longitudinal			
	Second	0.0744	0.0500	
	Transverse			
U + 5 w/o Fs + 2.25 w/o Zr U + 6.7 w/o Fs + 1.3 w/o Zr; 10% Enriched				0.0474
	First	0.0634	0.0454	
	Transverse			
	First	0.0610	0.0440	
	Longitudinal			
	Second	0.0614	0.0442	
U + 10 w/o Fs + 5.0 w/o Zr; 10% Enriched	Transverse			
	Second	0.0639	0.0457	
	Longitudinal			
	Third	0.0625	0.0460	
	Transverse			
U + 10 w/o Fs + 5.0 w/o Zr; 10% Enriched	First	0.0414	0.0357	
	Transverse			
	First	0.0419	0.0352	
	Longitudinal			
	Second	0.0454	0.0370	
	Transverse			
U + 10 w/o Fs	Third	0.0458	0.0369	
	Transverse			
	Fourth	0.0454	0.0368	
	Transverse			
				0.0345

*No value.

**Ziegler, S. T. and Nevitt, M. V., Structure and Properties of Uranium Fissium Alloys, ANL-6116 (July 1961).

If the hydrogen is not in solution, if the temperature is too low for adequate diffusion or too high to permit high triaxial stresses to develop, or if the hydrogen is trapped in such a way that diffusion cannot take place, hydrogen embrittlement will not be observed. Some of the potential conditions for hydrogen embrittlement are present in certain reactor applications, but the addition of irradiation to this problem has not been examined. Some hydrogen is born by fast-neutron reactions with iron, i.e., $\text{Fe}^{54}(\text{n}, \text{p})\text{Mn}^{54}$ and $\text{Fe}^{56}(\text{n}, \text{p})\text{Mn}^{56}$. The protons produced by these reactions could possibly cause hydrogen embrittlement if enough are generated. However, the large number of defects introduced into the lattice under neutron irradiation may tend to trap the hydrogen and prevent the diffusion necessary for embrittlement.

An experiment is being planned in which enough hydrogen can be generated by neutron irradiation to lead to hydrogen embrittlement. Since cross sections are small, it will take a high level of fast flux to reach this concentration in a reasonable period of time, even if use is made of a very high-strength steel with a known high susceptibility to hydrogen embrittlement. Current plans involve irradiation in ETR of notch-tensile specimens of Type 4340 steel* heat treated to the 190-kg/mm² strength level. The feasibility of the proposed irradiation depends on the gamma-ray heating rate in the ETR core position desired. Even specially plated steel will out-gas hydrogen at temperatures above 90°C. A measurement of the gamma-heating rate is under way.

Some related experiments that require a much lower integrated neutron flux are to be done in CP-5. These will involve hydrogenation of the steel by electrolytic techniques before and after irradiation. The ability of the steel to retain hydrogen during irradiation and the effectiveness of defects as traps will be evaluated. These tests will include ASTM specification A212-B pressure vessel steel in addition to Type 4340. It is conceivable that a tough steel like A212-B could become susceptible to hydrogen embrittlement after it has become hardened due to neutron irradiation. The implications of this type of effect to water-cooled reactor systems need not be overemphasized.

E. Engineering Development

1. Two-phase Flow

a. Void Fraction--Pressure-drop Facility. This experimental facility is designed to investigate the two-phase flow characteristics of boiling sodium; in particular, it is desired to obtain experimental information pertinent to the vapor volume fraction and two-phase frictional losses in an adiabatic test section.

*This steel contains 1.62 to 2.00 w/o Ni, 0.70 to 0.90 w/o Cr, and 0.38 to 0.43 w/o C.

Following test section repairs and flushing operations (see Progress Report for April 1965, ANL-7045, p. 35), the loop was preheated to 1200°F in preparation for further boiling runs. At this point a failure occurred in the electromagnetic pump tube and the loop was shut down. Inspection showed the failure due to a faulty weld, although the ultimate cause was certainly corrosion. Repairs will require approximately five weeks, and will delay the repeatability and higher quality runs needed to complete the experiment.

2. Boiling Liquid Metal Technology

a. Niobium-1% Zirconium Loop. The Nb-1% Zr loop construction is 50% complete. Pratt & Whitney-CANEL reports the pump duct has been received, leak tested, and is being welded into the loop.

The design of the lower loop-support structure has been approved for construction. The design of the clean storage area is complete, and construction will begin.

Final instrumentation designs are complete. Orders have been placed for the 109 thermocouple assemblies required for operation within a high-vacuum environment. A high-temperature vacuum furnace is available for calibration. An experiment to determine the diffusion bonding of Nb-1% Zr to itself and to tantalum is under way. The pressure transducers have been fabricated and initially calibrated.

The niobium parts for the EM flowmeter, supports, and a pump for the flowmeter cooling system are being fabricated.

An order has been placed for the automatic data-acquisition system, consisting of a crossbar scanner, an integrating digital voltmeter, a preamplifier, a tape punch coupler, punch power supply, and a teletype tape punch. The 160A computer routines for analyzing two-phase liquid metal heat transfer data are approximately 50% complete.

Initial assembly of the vacuum chamber revealed that the upper flange was considerably out of tolerance for flatness. The bell jar was taken down, the oven disassembled, and the vessel shipped to a local fabrication concern where the flange was refaced. After reassembly of the vacuum chamber, one of the roughing pumps was operated briefly to correct some problems in the control circuits.

The liquid-nitrogen manifold previously assembled was slightly modified to reduce the possibility of valve freeze up and has been partially insulated. The ball valves on the leak-detector lines developed leaks, and it was necessary to rebuild the lines using new valves.

The blower, air duct, capacities, autotransformer, insulation assembly, and stator assembly for the EM sodium pump system have been received. A housing has been fabricated for the transformer, some of the equipment has been set in place, and the installation design work has been completed.

b. Heater Experiments

(i) Electron Bombardment Heater. Modifications (see Progress Report for April 1965, ANL-7045, p. 37) were completed and operation of the system continued. However, a breakdown of a relay in the high-voltage power supply plus a breakdown of an alumina insulator in the heated section and a leak in the vacuum system have caused further delay. Both the relay and the alumina insulator have been replaced. The vacuum system is being examined to locate and eliminate the leak. Following repairs, the system will be degassed and operation continued to the maximum test power of approximately 10 kW.

3. General Heat Transfer

a. Heat Transfer in Double-pipe Heat Exchangers

(i) Liquid Metal Cocurrent Turbulent Flow. The research program described in previous progress reports and which consisted of both experimental and analytical investigations, has been completed and a draft of a report written. Since the major portion of the research was performed as an M.S. thesis for Marquette University, under the supervision of an ANL staff member, the report was presented to an appropriate committee of Marquette professors and ANL staff members. Various revisions were suggested and are now being included in the final draft of an ANL report.

In general, the research verified the validity of various results which idealized analyses had predicted previously. Most important of these is a new heat exchanger design relation which offers significant improvements in accuracy compared to methods currently in use. Since practical applications of this new method will be very convenient once certain quantities are available, a brief computing program has been initiated which will result in generalized tabulations of these quantities.

4. ANL-AMU Program

Other heat engineering experiments, performed as part of a joint program (not supported by the Division of Reactor Development and Technology) between the Laboratory and the Associated Midwest Universities (AMU), are described below.

a. Propagation of Void Waves in Air-Water Flow. This project was designed to investigate the behavior of void-fraction disturbances in an air-water system. All experimental data have been analyzed, and the results were compared with predictions based on the kinematic wave theory. A qualitative comparison shows good agreement; a quantitative comparison suggests modification of the theory.

F. Chemical Separations

1. Fluidization and Volatility Processes

a. Recovery of Uranium from Low-enrichment Fuels

(i) Laboratory Support Work. Experimental evaluation of bromine pentafluoride as a selective fluorinating agent for uranium in uranium-plutonium mixtures was continued (see Progress Report for April 1965, ANL-7045, p. 40). It is expected that the reaction between BrF_5 and mixtures of uranium and plutonium compounds will result in the conversion of uranium to volatile UF_6 while plutonium is converted to nonvolatile PuF_4 . Plutonium can then be converted to volatile PuF_6 in a subsequent step using elemental fluorine.

A series of experiments were performed in a horizontal tubular reactor to determine the kinetics of the reactions of BrF_5 with UF_4 , UO_2F_2 , and with mixtures containing UF_4 , UO_2F_2 , alumina, and fission products. The data were treated using a diminishing-sphere model in which F , the fraction reacted, is related to a rate constant k' and a reaction time t by the expression

$$(1 - F)^{1/3} = 1 - k't.$$

Values of k' for the reaction of UF_4 with BrF_5 in the temperature range 175-275°C varied from 0.0005 to 0.0344 min^{-1} . The integrated form of the Arrhenius equation for the reaction rate constants is

$$\log k' = 5.7194 - (3948.3/T).$$

The apparent activation energy for the reaction was calculated to be 18.1 kcal/mole UF_4 .

Values of k' for the reaction of UO_2F_2 with BrF_5 in the temperature range 175-300°C varied from 0.0040 to 0.0276 min^{-1} . The least-squares fit of the Arrhenius equation to these data established the following relationship:

$$\log k' = 1.4931 - (1747.1/T).$$

An apparent activation energy of 8.0 kcal/mole UO_2F_2 was calculated for the reaction.

In tests performed with solid mixtures containing UO_2F_2 and UF_4 (ratio 2:1), alumina, and fission products at temperatures between 175 and 250°C, rate constants from 0.0038 min^{-1} at 175°C to 0.017 min^{-1} at 250°C were obtained. A least-squares fit of the Arrhenius equation to these rate data resulted in

$$\log k' = 2.0197 - (1970.3/T).$$

Comparison of these rate data with values calculated for a $2\text{UO}_2\text{F}_2\text{-UF}_4$ mixture using the rate data obtained for UO_2F_2 and UF_4 alone showed reasonably good agreement. The rates for the mixtures containing UO_2F_2 , UF_4 , alumina, and fission products are about 20% greater than those calculated for the theoretical $2\text{UO}_2\text{F}_2\text{-UF}_4$ mixture. The apparent activation energy for the test mixture was calculated to be 9.0 kcal/mole, whereas that calculated for the theoretical $2\text{UO}_2\text{F}_2\text{-UF}_4$ mixture was 10.0 kcal/mole.

In experiments performed to determine the comparative reactivities of BrF_5 and fluorine toward uranium compounds, it was shown that the reaction rate constants for reactions involving fluorine were lower by an order of magnitude than the rate constants for comparable reactions with BrF_5 .

(ii) Decladding and Fluorination. Development studies are being performed in a 2-in.-dia fluid-bed reactor to determine the conditions for fluorinating $\text{UO}_2\text{-PuO}_2$ pellets that would result in a minimum retention of uranium and plutonium on the fluid bed of alumina particles. Three runs have been completed with $\text{UO}_2\text{-PuO}_2$ pellets containing 19 nonradioactive fission products to simulate fuel after 10,000 MWd/ton burnup. In each run, a 2-in.-deep bed of pellets was reacted by a 3-step process: (1) two-zone oxidation-fluorination step (see Progress Report for January 1965, ANL-7003, pp. 58-59) at 450-500°C, (2) a single-zone fluorination step for 5 hr at 450°C in which 10 v/o fluorine in nitrogen is introduced into the bottom of the reactor, and (3) a recycle-fluorination step with 80 to 95 v/o fluorine at 450°C for 5 hr, 500°C for 5 hr, and 550°C for 10 hr.

The final uranium and plutonium concentrations in the alumina bed material were about 0.004 and 0.007 w/o, respectively, representing uranium removals of >99.9% and plutonium removals of 97% for a single use of the alumina bed. Plutonium recoveries of >99% can be achieved by reuse of the alumina bed material for additional process cycles (see Progress Report for May 1964, ANL-6904, pp. 88-89). In these experiments, the concentrations of uranium and plutonium in the final bed were apparently independent of variations in operating conditions among the three experiments. No effects resulted from changes in fluorine concentrations, filter blowback frequency, superficial gas velocity during recycle-fluorination period, and type of alumina used as the fluid-bed material. The course of uranium and

plutonium removal was observed throughout the recycle-fluorination period. The results indicate that the recycle-fluorination time required to obtain satisfactorily low retention of uranium or plutonium on the alumina may be less than the 20 hr employed in these tests.

(iii) Cleanup of Cell Exhaust Air Contaminated with Plutonium Hexafluoride. Studies have continued on the cleanup of air contaminated with plutonium hexafluoride. A series of three experiments were made to determine surface reaction kinetics for the hydrolysis of a mixture of PuF_6 and UF_6 in a gas stream. Two air streams, one containing the mixed hexafluorides and the other containing moisture, were rapidly mixed and passed through a segmented bed of glass spheres. The quantities of plutonium and uranium retained on the glass spheres within the various bed segments were used to estimate rates of hydrolysis for UF_6 and PuF_6 in the mixture. Previous work (see Progress Report for December 1964, ANL-6997, p. 41) indicated that the hydrolysis rate for PuF_6 alone was similar to that of UF_6 alone.

The results showed that the rates of hydrolysis of the component hexafluorides in the mixture differed from the rates obtained when the hexafluorides were individually hydrolyzed. In experiments in which air of low moisture content (about 70 ppm) was used, the hydrolysis rates for PuF_6 in the mixture were markedly higher than rates obtained when PuF_6 alone was hydrolyzed. On the other hand, hydrolysis rates for UF_6 in the mixture were lower than rates obtained with UF_6 alone. In the mixed gas the hydrolysis rates for PuF_6 were always higher than the hydrolysis rates for UF_6 . The data indicate that the hydrolysis mechanism for mixed hexafluorides is more complex than that expected on the basis of mechanisms for individual hexafluoride species.

(iv) Engineering-scale Alpha Facility. In preparation for the next shakedown runs in the fluorinator and the converter of the engineering-scale alpha facility, the two gloveboxes were sealed and a complete check of the ventilation system was made. The test showed that the proper negative pressure (0.5 in. H_2O) was maintained in the boxes under conditions of design gas flow rates.

A device for blending and sampling liquid mixtures of UF_6 - PuF_6 has been constructed and tested. The device, which is housed in a heated enclosure, was designed to mix the liquids in the hexafluoride collection cylinder by rocking the cylinder. It is expected that this mixing will keep in suspension the solid particles of PuF_4 produced by α -decomposition of PuF_6 . The hexafluorides are kept in the liquid state under approximately 50 psia pressure by heating the cylinder to 90°C . A 10- to 20-g sample is obtained by discharging the cylinder into a small evacuated sample tube.

(v) Bench-scale Studies of Irradiated Fuels. Two experiments have been performed in the bench-scale high-activity-level facility with highly irradiated uranium dioxide fuel to define the behavior of fission products in the fluoride volatility process. The facility consists of a $1\frac{1}{2}$ -in.-dia fluid-bed reactor and a packed-bed filter with associated equipment for the collection of the hexafluoride products and disposal of process gases (see Progress Report for May 1964, ANL-6904, pp. 89-90). (Data on studies with irradiated enriched uranium alloy fuels were presented in the Progress Report for March 1965, ANL-7028, pp. 64-65.)

The UO_2 pellets used in the tests were irradiated for one year to a level of 20,000 MWd/ton and cooled for 18 months. The original uranium enrichment was 3.6%. Approximately 50 g of UO_2 were charged to the fluid-bed reactor in each test. The processing scheme involved two steps: (1) conversion of the pellet charge to a powder by reaction with a 48 v/o HF-52 v/o O_2 mixture for 4 hr at 400°C , and (2) fluorination of the fuel by reaction with 2 to 20 v/o F_2 in nitrogen for 3 hr at a bed temperature of 405 to 450°C .

It was observed that 0.3% of the gamma activity and 0.1% of the beta activity volatilized during the first step. These activities were primarily antimony and ruthenium. In the second step, 99.7% of the uranium charged to the system was removed from the reactor as volatile uranium hexafluoride. The UF_6 was purified by passage through a NaF trap at 400°C and collected in a NaF trap at 100°C . The activities which were found to be nonvolatile in the fluorination step were the rare earths, strontium, zirconium, and cesium. The elements which were partially volatile were antimony, ruthenium, and niobium, which were deposited in significant quantities in the 400°C sodium fluoride trap. Molybdenum and technetium were completely volatilized from the fluid-bed reactor and were sorbed with the uranium on the NaF in the trap maintained at 100°C . The decontamination factor for uranium from the principal fission product activities, excepting molybdenum and technetium, calculated from activities in the 100°C NaF trap, ranged from 4×10^3 to 9×10^4 .

b. Recovery of Uranium from Highly Enriched Uranium Alloy Fuels

(i) Pilot-plant Demonstration Runs. The final data on the disposition of uranium during processing of uranium-zirconium and uranium-aluminum multiplate fuel assemblies in the pilot plant facility have been collated. A description of the process and typical operating conditions were presented in the Progress Report for December 1964, ANL-6997, pp. 41-45. The main objectives of the pilot-plant program were (1) to demonstrate the recovery of over 99% of the uranium from the alloy fuel and (2) to define the mode of uranium losses. The uranium that is considered to be unrecoverable includes that which passes through the packed-bed filter into the pyrohydrolysis reactor during the hydrochlorination step and that which remains on the fluid-bed and packed-bed material at the end of the fluorination step.

In general, high uranium recoveries--about 99% of the uranium charged to the fluid-bed reactor--were achieved with uranium-zirconium alloy fuel. The transport of uranium through the packed-bed filter was 0.2%. The uranium retained on the alumina-bed material was approximately 0.8% of the uranium charged. It was also observed that reuse of the alumina bed material for additional process cycles resulted in a significant decrease in the loss of uranium to the alumina bed.

The uranium loss to the pyrohydrolysis reactor was relatively high (1.5%) in runs with uranium-aluminum fuel; in these runs the packed-bed temperature was $>200^{\circ}\text{C}$ during the hydrochlorination step. Chlorination of the uranium to a species more volatile than UCl_4 (possibly through reaction with aluminum chloride) could account for the high loss. Based on the results of bench-scale tests,¹⁴ it is expected that uranium loss by this mechanism can be reduced to less than 0.5% by operating with a packed-bed temperature of 180°C . The retention of uranium on the fluid-bed alumina after fluorination was consistently less than 0.2% of the uranium charged to the system.

G. Sodium Coolant Chemistry

1. Control of Sodium Oxide Impurity

a. Distillation Analyzer. The problem of effective analysis of sodium samples for oxide content is dependent upon an accurately known quantity of sodium sample. One inherent difficulty associated with the distillation analyzer is the problem of obtaining a reliable sample volume. A possible solution to this problem lies in sampling the sodium after it is solidified by a method in which sodium, at temperature, is circulated through an arm of a loop. After circulation, the sodium is frozen with its impurities "in situ." This is followed by extruding a solid sodium cylinder of fixed length into the distillation analyzer. The ends of the sample are cut off and discarded to eliminate end effects.

A device was constructed to enable an operator to perform extrusions into a protective vacuum region. The sodium extruder, and cut-off and waste remover have been subjected to a series of operational tests to develop a suitable technique. Some of the troublesome problems are: development of vacuum leaks due to shifting of gasket seals when bellows-sealed manipulative rods are operated; galling between cut-off blade and its channel resulting from misalignment between them; lack of complete severance of cut-off segment of sodium after extrusion due to failure of blade to attain "home" position and sticking of severed sodium to the stainless steel orifice below the blade.

¹⁴Chemical Engineering Division Semiannual Report July-December 1963, ANL-6800, pp. 271-282.

2. Sodium Purification

Detection by mass spectrometry of high molecular weight organic compounds in various sodium samples was reported previously (Progress Reports for April and March 1965, ANL-7045, p. 43, and ANL-7028, p. 66). Two new graphite electrodes of the type employed in the electrolytic manufacture of sodium have been examined mass spectrometrically for evidence that they might be the source of these compounds. The results indicated that these electrodes (and presumably ones made in a similar manner) are not responsible for the heavy mass species previously observed. Only low-mass species (<103) were evolved up to 850 to 900°C, and these gradually disappeared with time. At about 1025°C, no peaks above background were observed. Further work is planned to determine the source of the high molecular weight organic compounds.

3. Carbon Analysis

The assignment of a blank correction is an important aspect of the analysis of sodium for carbon. Ideally, the blank would be determined by carrying out the analysis with carbon-free sodium. Because carbon-free sodium is not available, the blank is usually obtained by duplicating the analytical procedure with sodium absent. The carbon found (from the apparatus and reagents) is then subtracted from the carbon found in a sodium sample. The blank determined in this manner is inexact because the experimental conditions are not identical to those when sodium is present.

An alternative method now being used to obtain a more reliable blank correction involves the following: Sodium is analyzed for carbon over a wide range of sample weights, e.g., 0.1 to 2 g. The carbon content is plotted as a function of the sample weight, and a least-squares curve is fitted to the points. The intercept on the ordinate (zero sodium weight) is taken as the blank. Such a plot is shown in Figure 16 for sodium manufactured by the

Figure 16
Carbon Found vs. Weight
of Sodium Sample

United States Industrial Chemical Co. and analyzed for carbon by a modification of the method of Kallman and Liu.¹⁵ The slope of the curve, i.e., the carbon content, is 28.8 ± 4.4 μg carbon per gram of sodium; the intercept, or blank, is 6.0 ± 1.4 μg carbon. Blank determinations by the method first discussed gave a value of 2.1 ± 1.3 μg carbon. The reason for the discrepancy is not clear. The method of using multiple sample sizes will be employed to obtain more reliable blank corrections in future work, particularly where very low carbon contents are being assayed.

H. Plutonium Recycle Reactors

1. EBWR Facility

a. Primary Piping

(i) 8-in. Reboiler Piping. Sonic and confirming measurements of the thickness of wall of the 8-in. primary steam line showed no evidence of deterioration in service. All 26 welds in the tunnel between the containment shell and the primary reboilers were radiographed and evaluated; 14 welds require repairs before further service.

(ii) New Boric Acid Line. The new boric acid line was radiographed in place; 28 of the 45 welds are satisfactory; evaluation of the radiographic indications of the remaining 17 welds is in progress.

(iii) 20-MW, 6-in. Safety Valve Line (original). Faulty welds were repaired, and are ready for radiographic and magnaflux inspections.

(iv) 20-MW, 6-in. Steam Line (original). Faulty welds were repaired, and are ready for radiographic and magnaflux inspections.

(v) 80-MW, 6-in. Safety Valve Line. Ten welds were repaired and re-radiographed. All welds were acceptable for continued operations.

(vi) Start-up Heater Piping (3 in. and 4 in.). Radiographs revealed that 6 of 12 repaired welds were accepted for service.

(vii) 8-in. Steel Condenser Line. Three of the five repaired welds were accepted; evaluation of radiographic indications for the remaining two are in progress.

b. Feed Water Filters

(i) Feed Water Filter No. 1. Grinding explorations of the carbon steel weld crack in the lower head-to-shell seam were completed.

¹⁵Kallman, S. and Liu, R., Anal. Chem. 36, 590 (1964).

The crack in the carbon steel terminated at the Type 304 SS cladding interface. Welders are being qualified for the weld-repair procedure.

The flanged cover dome was sent to the Central Shops for a build-up of the head-to-flange weld to reduce stresses in the "hub" region of the connection.

(ii) Feed Water Filter No. 2. A welding neck orifice flange was salvaged from this filter to replace a slip-on flange on Feed Water Filter No. 1.

(iii) Feed Water Filters Nos. 3 and 4. The design review of the filters was completed. A simple remedy was found for reducing the computed stress in the filter body flanges. A 4-in. high circular ring by 1/2-in. thick wall, with ID equal to OD of flange, when butt welded to the flange reduced flange stresses to 17,000 psi, which is below the maximum allowable ASME Code Stress for SA 105-Gr II forgings.

c. Reactor Pressure Vessel Cladding. Cracking in a composite 2-in. thick test plate consisting of 304 SS spot welded to SA 212B steel base plate purchased in 1959 to enable determination of the quality of practice machining, flame cutting, and welding operations, has been observed. In 1959, there were no noticeable cracks in the stainless steel cladding of this plate. The plate had been exposed to the weather during the period of storage and was cracked more extensively than the EBWR pressure vessel cladding (see Progress Report for March 1965, ANL-7028, p. 66).

A dye check examination, for cracking, of a 1954, 4-in. thick EBWR 304 SS-clad SA 212B head plate revealed cracking in the cladding. This piece had not been in the reactor vessel.

A plug of $2\frac{7}{16}$ -in., 304 SS-clad SA 212B steel, cut from the EBWR pressure vessel wall in 1960, showed no cracks in the cladding. This plug had been in the reactor vessel 4 yr and stored outdoors for 3 yr since then.

Subsequent dye penetrant re-examination of the cladding inside the vessel revealed two of the three panels, previously reported as crack free contained minute superficial, surface cracks and that only one panel was totally crack free.

Metallographic examination of samples of the cracked 304 SS cladding, which had been in the EBWR for 10 yr and now removed for examination, revealed that each grain of steel was surrounded by brittle carbides which had precipitated from the solid-solution stainless steel. This brittle carbide precipitation was caused by the 1150°F temperature to which the vessel was heated for stress relief during its manufacture.

Metallographic examination of cracks propagated in the 1959 test plate revealed that the cracks stopped (1) at areas between the welded areas of SA 212B base plate and 304 SS cladding (interface), and (2) at areas of dendritic structure. This structure is the herringbone structure of the weld caused by rapid cooling of the molten weld metal. Samples cut from the cladding panels of the EBWR pressure vessel wall have not, as yet, been examined for details of crack propagation.

Other studies in progress include (1) Strauss corrosion resistance testing of cladding from the pressure vessel, the test plate, the head plate, and the plug; (2) chemical composition of each 304 SS panel section in the upper one-half of the pressure vessel; (3) chemical analysis of the interface oxide deposits formed between the 304 SS cladding and the SA 212B steel wall. For these studies, ten cladding strips, one from each panel of stainless steel cladding, were removed from the reactor vessel wall.

2. Fuel Development and Fabrication

The fuel loading for the Plutonium Recycle Experiment consists of 148 elements in three separate zones. The central zone, which is the plutonium test fuel, is surrounded by a driver of uranium fuel of both 6% enriched and natural uranium. The natural uranium fuel is located on the periphery at the core loading and the enriched or shim fuel is positioned between the plutonium test fuel and the natural uranium driver fuel.

A total of 1,512 plutonium test fuel rods containing a mixed oxide of 1.5 w/o of PuO_2 in depleted UO_2 loaded into Zircaloy-2 tubes was fabricated at Battelle Pacific Northwest Laboratories (PNL). These rods were fabricated by a combination of pneumatic impaction of mixed oxide and a vibratory-compaction loading process to effect a density of 86 to 89% of theoretical density in loaded rods. All fuel rods were thoroughly tested, inspected, and accepted at PNL, and delivered to ANL for loading into fuel elements.

All plutonium test fuel rods were assembled into fuel elements at ANL and are available for use in the reactor. A total of 36 test core elements and six spare elements were assembled. Each element contains fuel rods in a 6 x 6 array loaded into Zircaloy-2 square cans of 0.157-cm nominal wall thickness.

In addition to the regular PuO_2 - UO_2 fuel for the central zone, 25 test fuel rods were fabricated at PNL and delivered to ANL for physics measurements on the core. These rods contain plutonium-aluminum alloy, special isotopic plutonium-aluminum alloy, natural UO_2 without plutonium, and mixed-oxide fuel with controlled isotopic plutonium content.

The driver fuel loading consists of 60 enriched-uranium fuel elements and 52 natural uranium fuel elements. The uranium fuel rods contain UO_2 pellets in Zircaloy-2 tubes and are assembled into elements in a 6 x 6 array the same as the plutonium test fuel. The above driver fuel elements plus four spare enriched-uranium elements and six spare natural-uranium elements and all components for the plutonium test-zone element assembly were fabricated by United Nuclear Corporation. The last shipment of spare uranium fuel was received on May 14, 1965, and all of the uranium fuel is available for use in the reactor.

It is possible that the innermost row of enriched uranium shim fuel elements may have to be replaced during the course of the experiment to restore reactivity to the overall system. All necessary components for an additional 32 elements, except oxide fuel, are also being furnished. These components are all completed with the exception of five Zircaloy-2 fuel element cans, which were delayed for lack of material. These cans are now in process.

Detailed records have been compiled for all materials used for structural and fuel components; inspections and tests for all structural components; and for all fuel fabrication, testing, inspection, and loading. Each fuel rod and each fuel element have individual identification markings cross referenced with all fabrication and inspection records.

III. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Critical Experiments

a. Theoretical Analysis. A set of 16-group cross sections has been established for each of the three basic core loadings on which experiments have been performed: (1) 315 fuel foils (16.1 kg U^{235}); (2) 615 fuel foils (31.4 kg U^{235}), plus 420 B-SS poison strips (220.5 g B); (3) 810 fuel foils (41.4 kg U^{235}), plus 810 B-SS strips (425 g B). The procedure involves: the use of TEMPEST¹⁶ computations for the two thermal groups; the assignment of an equivalent Σ_{spot} (homogeneous) per atom of U^{235} (with the assumption that the fuel foils act like isolated slabs at U^{235} -resonance energies); and the use of curves by Hansen and Roach¹⁷ to obtain $\bar{\sigma}_c U^{235}$ and $\bar{\sigma}_f U^{235}$ for the resonance-energy groups. A more detailed discussion of the approximations of getting group-averaged cross sections is available (see Progress Reports for March 1965, ANL-7028, pp. 73-74 and April 1965, ANL-7045, p. 49).

For each case, an additional set of cross sections was determined with the alternative, extreme assumption that U^{235} foils are black to neutrons of all resonance energies. Preliminary 16-group REX¹⁸ computations indicate that $\Delta k/k$ (black foil $\rightarrow$ isolated fuel) is negligible for loadings of 810 fuel foils or less. This result for the 810-foil loading is much smaller than the value of +0.4% reported in the Progress Report for April 1965 (ANL-7045, p. 49) and the analyses are being rechecked.

In the following, results of preliminary REX 16-group computations are summarized. Certain results, such as the variation of coolant void coefficient of the fuel zone with increased fuel content, are being studied in greater detail. Additional calculations are in progress on the excess reactivity of the unpoisoned system (no boron-stainless steel strips) as a function of fuel-foil loading.

(i) 315-fuel-foil Loading. Without correction for the gross heterogeneity of the fuel foils in this loading, $k_{eff} = 1.039$. The estimated reactivity loss due to this heterogeneity is $\sim 3\%$; including this correction, the calculated k_{eff} is within 0.5% of the measured value, 1.004 (see Progress Report for March 1965, ANL-7028, p. 74).

¹⁶Shudde, R. H., and Dyer, J., TEMPEST-II, AMTD-111, NAA Program 1, TID-18284.

¹⁷Hansen, G. E., and Roach, W. H., Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies, LAMS-2543 (Dec. 1961).

¹⁸Butler, M. K., and Cook, J. M., UNIVAC Programs for the Solution of One-dimensional Multigroup Reactor Equations, ANL-5437 (Oct 1956).

(ii) 615-fuel-foil Loading. Two cases have been calculated: (α) 10 v/o Plexiglas in the beryllium reflector, distributed in horizontal layers; (β) no Plexiglas. The neutron-poisoning effect of the 420 B-SS strips was included, but not the effect of heterogeneities of fuel foils and of Plexiglas.

For case (α), the calculated value of k_{eff} was about 1.033, in comparison with that measured, ≈ 1.039 (see Progress Report for December 1964, ANL-6997, p. 55). The calculated reactivity effect of removing a homogeneously distributed 10 v/o Plexiglas is 1.4%; the measured effect is 1.2% (see Progress Report for March 1965, ANL-7028, p. 75).

For this loading, the calculated reactivity worth of boron in the fuel zone was -0.048% per g boron.

(iii) 810-fuel-foil Loading. For this loading, with 425 g B in the core, the calculated value of k_{eff} was 1.029. The measured value was 1.030 (see below, Section III.A.1.b). Note that there is no Plexiglas in the beryllium reflector. The calculated k_{eff} does not include a correction for fuel-foil heterogeneity, which is expected to be small.

The calculated reactivity worth of boron in the fuel zone was -0.035% per g boron.

(iv) 1215-fuel-foil Loading. It has been estimated that a loading of 1215 standard fuel foils, containing a total of ~ 62.0 kg U^{235} , plus a loading of 1650 B-SS strips (866 g B) would be supercritical by $2\frac{1}{2}\%$. This provides a basis for such a core loading.

b. Measurements. Experimentation continued with the 810-fuel-foil core loading consisting of 41.4 kg U^{235} and 425 g natural boron (see Progress Report for April 1965, ANL-7045, pp. 50-52). The measured excess reactivity of this loading is estimated to be 2.9%, on the basis of calibration of control blades by the rising-period method.

Measured worths of the control blade ranged from 1.25% to 1.45% in unshadowed configurations, with the differences being attributed to locations and orientations. For the four types of control blades in the system the following worths were obtained: each of peripheral control blades Nos. 1, 3, and 5 controls, 1.30%; each of peripheral blades Nos. 2, 4, and 6 controls, 1.44%; each of interior control blades Nos. 7, 9, and 11 controls, 1.25%; and each of interior blades Nos. 8, 10, and 12 controls, 1.43%. These results indicate a reduction of 5-10% of the absolute worth of each control blade with the increase in loading from 615- to 810-fuel-foils. Earlier measurements had indicated a reduction of 10% or more in blade worth when the fuel loading was increased from 315 to 615 fuel foils.

Measurements of the reactivity effect of voids in the water were made in the 810-fuel-foil core and in the ITC, using Teflon to displace water. The average for the fuel region was between values obtained in the 315- and 615-fuel-foil loadings, as shown in Table XVIII. The absolute magnitude of the reactivity coefficient of voiding of core water had been expected to increase as the fuel loading was increased. The removal of Plexiglas from the beryllium reflector may have had a larger effect on this void coefficient than was expected, a possibility which will be studied computationally. The measured value of void coefficient in the ITC was unchanged from coefficients measured in earlier core loadings.

Table XVIII. Reactivity Coefficient of Voids in Water
(%/void in the water)

Loading of Fuel Foils	Core	ITC
315	-0.16	Not Measured
615	-0.23	+0.05
810	-0.19	+0.05

Measurements of void worth at different core radii were taken in this loading for eventual comparison with similar data to be obtained in the fully loaded cores (see Table XIX).

Table XIX. Radial Distribution of Void Coefficient
in the 810-Fuel-foil System (%/void in the water)

Radius, cm	Void Coefficient of Reactivity	
0.0	+0.05	} ITC
1.6	+0.05	
3.2	+0.05	
4.6	+0.05	
5.3 (corner)	+0.04	
6.8	-0.14	} Fuel Zone
10.5	-0.24	
14.6	-0.25	
20.3	-0.14	

The mean prompt-neutron lifetime inferred from a Rossi- α measurement in the 810-fuel-foil system was between values obtained with lighter core loadings. It is being remeasured. The various possibilities which might affect this result are being carefully rechecked. Available data are given in Table XX.

Table XX. Rossi- α Data

Loading of Fuel Foils	Rossi- α , sec ⁻¹	Mean Prompt-neutron Lifetime, μ sec
315	187 $\pm$ 5	38.8
615	274 $\pm$ 7	26.5
810	250 $\pm$ 6	29.0
(preliminary)		

In the 315-fuel-foil loading and in the 615-fuel-foil loading (with and without Plexiglas in the beryllium), U-Al vertical traverses (U^{235} fission) had been obtained with a full top water reflector (approximately 37 cm of water above the fuel). For the 810-fuel-foil loading, the vertical traverses were made with the control blades withdrawn and criticality attained by adjustment of the water level (see Figure 17). It is of interest to note that the peak activation appears at the same vertical level in the four regions investigated. It was at this level that the radial traverses were taken (approximately 3.2 cm below the core midplane). A new set of U-Al foils has been intercalibrated on the wheel in the Argonne Thermal Source Reactor graphite stack, and it is expected that radial and vertical traverses will be made with these foils. The radial locations of the vertical traverses are shown in Figure 18.*

Radial traverses were made with manganese foils with the reactor made critical by adjustment of water level. In addition, in the 615-fuel-foil loading without Plexiglas in the beryllium, U-Al radial traverses (U^{235} fission) were made. Figures 19 and 20 show the radial traverses with manganese foils in the 810-fuel-foil loading for the cases of criticality with water-level adjustment and with control blade positioning, respectively. Locations of the foils used for Figure 19 are identical to those previously described for the 615-fuel-foil loading without Plexiglas in the beryllium (see Progress Report for April 1965, ANL-7045, p. 51). The radial locations of the foils for Figure 20 are shown in Figure 21.

Note that there is a marked flux tilting due to the poisoning of one third of the reactor with control blades (see Figures 20 and 21).

*Figure 18 of the present report shows locations of foils for vertical-activation measurements. These locations also apply to Figure 12, ANL-7045. Note that in the latter figure the hexagon symbol is incorrectly positioned, and the symbols $\square$ and $\triangle$ were omitted. The unshaded triangles in Figures 11 and 13 of ANL-7045 should be shaded (solid) circles, corresponding to the locations in Figure 12. Also, in Figure 13, the two arrows should be directed to the two vertical lines, not to the one line. In Figure 14, the unshaded circles correspond to the hexagon shown in Figure 12, with the location of the hexagon revised as noted above.

Figure 17
Vertical Activation Traverses with
Manganese Foils (810-fuel-foil Core)

Figure 18
Foil Locations for Vertical Activation Measurements

Figure 19
Radial Activation Traverses with Manganese
Foil (810-fuel-foil Core)

Figure 20
Radial Activation Traverse with Manganese
Foil (810-fuel-foil Core; Criticality
Achieved with Control Blades--see
Figure 16)

Figure 21. Foil Placement for Radial Activations with Manganese Foils (Blade Control)

2. Heat Transfer

a. Analytical Studies. A simplified steady-state heat transfer study was initiated to investigate sample cooling in rabbit tubes located in the internal thermal column. The study was undertaken in order to gain a better insight into the problems associated with sample cooling, to estimate sample limitations, and to determine the influence of rabbit-tube design on cooling requirements.

A digital program was written to study a cylindrically-shaped rabbit. The material being studied is assumed to be uniformly dispersed in the rabbit or in pellet form within a tube. The program allows for heating the coolant, tube housing, and sample during investigation.

An analog program was set up to evaluate the problems associated with the use of flow reversal to remove a rabbit from the core following irradiation.

b. Experimental Program. Difficulty has been encountered in the steady-state loop in that actual burnout of specimens has occurred before power to the apparatus can be shut off. The steady-state test program

has been temporarily halted pending outcome of an investigation of the power supply. Efforts are being directed toward reducing the time lag of switching circuits.

Studies of transient heat transfer were continued, during which measurements were made of the power rise time, which is defined as the time required to achieve 95% of the full-power setting. By trial-and-error series-parallel arrangements of the rectifiers, the rise time has been reduced from 100 to 60 msec. Although this decrease in time does not represent the ultimate, it encourages further attempts at reduction.

Design of a Shutdown Cooling Test Loop was started. Two heat exchangers for the loop have been designed and construction drawings started. A preliminary investigation has shown the test section fabrication technique to be feasible.

3. Primary Coolant System

Computer programs were written to study the effect of various leak rates on the pressure in the primary coolant system. Calculations were made by use of the orifice and exponential gas law equations, neglecting inertial, distributed storage, and vena contracta effects for conservatism and simplicity. The model used is illustrated in Figure 22.

Figure 22
Model of Primary Coolant System

Two programs were employed: one expressed the problem without regard for flow resistance at the surge tank outlet, the other took this resistance into consideration. Families of curves are shown in Figure 23 and 24. The curves of Figure 23 break sharply at, and of Figure 24 terminate at, a cut-off point. This point occurs when flow due to expansion of gas in the surge tank is interrupted.

Pressure decreases faster initially in the curves of Figure 24, but takes a longer overall time to reach the surge-tank cut off. Decay of pressure after surge-tank cut off was not computed for Figure 24 since it is due only to compression effects and would be the same as in Figure 23.

Additional useful information derived from these results include estimates of minimum allowable scram times, and rates and amounts of makeup and minimum sizes of system components.

Figure 23. Decrease of Pressure with Time
(no flow resistance)

Figure 24. Decrease of Pressure with Time
(flow resistance)

4. Fuel and Material Development

Initial irradiation tests of eight subsized fuel specimens in the ETR-G12 facility were completed (see Progress Report for December 1964, ANL-6997, p. 51, and Progress Report for March 1965, ANL-7028, p. 71). Programming of these material samples achieved the burnup rates, nominal temperatures, and average heat fluxes shown in Table XXI. The ratios of maximum to average heat flux for the tests did not exceed 1.5 to 1. The data are applicable to the fuel material only and not to the AARR fuel-assembly configuration, for which there is no dimensional simulation in the samples irradiated.

Table XXI. Irradiation Test Data for 37 w/o UO₂-SS-type 347 Fuel Plate Specimens for AARR.
Nominal Oxide Density was 80-85% of Theoretical.

Plate No.	Cycle 70				Cycle 71			
	Avg Fission Rate (Fiss/g U-sec x 10 ⁻⁴)	Avg Heat Flux ⁽¹⁾ (Btu/hr-ft ² x 10 ⁻⁶)	Avg Burnup (% U ²³⁵)	Surface Temp, Avg/Max (°F)	Avg Fission Rate (Fiss/g U-sec x 10 ⁻⁴)	Avg Heat Flux (Btu/hr-ft ² x 10 ⁻⁶)	Avg Burnup (% U ²³⁵)	Surface Temp, Avg/Max (°F)
M-1-926	1.49	1.37	8.6	240/282	1.57	1.33	21	226/295
M-1-923	1.49	1.37	8.6	240/282	-	-	-	-
M-1-933	-	-	-	-	1.57	1.45	~13	-
M-1-925	1.49	1.37	8.6	240/282	1.57	1.33	21	226/295
B-6-947	2.11	1.69	12.8	314/368	2.17	1.56	27	295/327
B-6-952	-	-	-	-	2.17	1.67	~15	304/343
B-6-943	2.11	1.69	12.8	314/368	-	-	-	-
B-6-945	2.11	1.69	12.8	314/368	2.17	1.56	27	295/327

⁽¹⁾Max heat flux for these tests was held down to 2×10^{-6} Btu/hr-ft².

Preliminary investigation at ETR indicated no measurable change in physical dimensions, using instruments accurate to ± 0.001 in. Some discoloration of the fuel plate specimens was observed after irradiation, which was attributed to Al₂O₃ deposition on the heat transfer surface. The depth of this deposit was not ascertained, but was believed to be less than 0.001 in., since it was undetected when the plates were measured for change in thickness.

Several full-scale plates and control rods were ordered to supplement the technology verification studies. In addition, new contracts were let for:

- (a) burnable poison addition to 37 w/o UO₂-stainless steel;
- (b) very low UO₂ inventory fuel plate fabrication verification studies;
- (c) oxide evaluation and improvement;
- (d) improvement of advanced-assembly brazing technology;
- (e) reactor internal components material compatibility studies.

Fabrication of a beryllium test block was completed for use in a high-velocity-flow study in the hydraulic test loop. A variety of coolant hole sizes and chamfers was used so that the effect, if any, on erosion rate in the reflector can be determined.

5. Studies of Reactor Vessel Stress

Investigation of stresses in the region of the cylinder-to-ellipsoid junction of the reactor pressure vessel continued. The ellipsoidal shell was considered to be an assemblage of tapered curved beams on elastic foundation, supported by "hoop rings." Results indicated that the present configuration is feasible.

An attempt was made to evaluate stresses at the beam tube penetrations. The large number of penetrations in close proximity makes accurate theoretical analysis difficult; further, use of beam tubes tangent to an 18-in.-diameter inner circle rather than normal to the shell complicates the problem. Approximate stresses at beam tube penetrations have been obtained. Stresses in the vicinity of a particular nozzle were computed by ignoring the existence of other nozzles, and total stresses were mapped by the method of superposition.

Inelastic buckling of a beam tube under external radial pressure was investigated analytically. Collapse pressure was determined to be 6700 psi.

6. Development of Reactor Components

A 1/4-in.-thick, full-size core-support plate template was machined on the tape-controlled milling machine. It was cut to prove out the technique prior to machining a full 6-in.-thick aluminum support plate, which will be fabricated for experimental stress studies. Dummy fuel assemblies are being fabricated to study core assembly and disassembly.

IV. NUCLEAR SAFETY

A. Reactor Kinetics

1. Fast Reactor Safety

a. Coolant (Water) Expulsion Studies. In order to eliminate extraneous voltage signals being picked up by the recording instrumentation (see Progress Report for April 1965, ANL-7045, p. 60), an isolated ground rod has been placed on the north side of Building 308. In addition, the entire apparatus has been isolated from the building. However, an a.c. signal of approximately 2 mV (as opposed to 10 mV before) is still being picked up. The source of this signal is now being investigated.

A computer program is now being written based on a physical model of coolant expulsion. When operating, the program will be able to compute pressure pulses in the coolant and velocity of expelled coolant, given the input information of the initial state of the system (temperature, pressure, and liquid level) geometry, physical properties of the coolant and cladding, and the energy generation rate. Although the model will accommodate any functional forms of energy generation rates, three specific cases will be initially studied: impulse, step, and step-exponential.

b. Calculations of Pressure Pulses. The finite-difference pressure pulse code (see Progress Report for March 1965, ANL-7028, pp. 80-81)

Figure 25. Calculated Pressure and Distance of Expulsion

was run on the CDC-3600 to calculate a series of cases in which part of a sodium-filled channel was heated. The heated section contained 620 g of sodium, was 60 cm in length, 12.187 cm 2 in area, and was initially under a pressure of 2.7 atm. Expulsion was assumed to be symmetric above and below the channel center, so the calculation was performed on an equivalent unidirectional expulsion of 310 g of heated mass in a channel 30 cm long. Tabulated sodium material properties were used. Figure 25 shows results for the reference case of 3-MW constant-power input and total coolant mass of 2.5 kg. Pressure is approximately 60 atm when the coolant leaves the heated section and power input ceases. The remainder of

the curves correspond to power continuing indefinitely; under this condition a peak pressure of 83×10^6 dyne/cm² (approximately 82 atm) is attained occurring at an expulsion distance of 152.7 cm. The initial portion of the pressure curve in the figure is concave upward; this results from the nonlinear relationship between vapor pressure and the enthalpy of the saturated liquid. The concave downward portion of the pressure curve results from appreciable amounts of energy going into vaporization. At an expulsion distance of 30 cm, the amount of mass vaporized was computed to be about 0.6 gm. A summary of results is given in Table XXII, including the calculated pressure at 30 cm, the pressure at 30 cm which was estimated from the reference problem result and a simple approximate scaling law, and the percent error in the scaled estimate. The scaling law, based on a model using constant properties and power as well as neglecting power going into vaporization (see Progress Report for December 1963, ANL-6810, pp. 53-54), predicts that pressure rise during expulsion from a fixed channel length is proportional to

$$(\text{power})^{2/3} / (\text{area} \times \text{mass})^{1/3}.$$

Table XXII. Summary of Pressure-pulse Problem

Problem	Total Mass (kg)	Power (ergs/sec)	Heated Channel Area (cm ²)	Pressure at 30 cm (10 ⁶ dyne/cm ²)*	"Scaled" Pressure at 30 cm (10 ⁶ dyne/cm ²)*	"Scaling" Error
1	25	3×10^{13}	12.187	59.6	-	-
2	10	3×10^{13}	12.187	39.4	44.5	12.9
3	10	1×10^{14}	12.187	117.6	96.1	-18.3
4	1	1×10^{14}	12.187	41.7	16.0	15.6
5	13.95	3×10^{13}	9.678	50.7	46.2	7.6
6	13.95	1×10^{14}	9.678	136.4	99.7	-26.9

*One atm = 1.016×10^6 dyne/cm²

It should be pointed out that the value of 60 atm at expulsion is based on a tacit assumption that the cross section of the channel does not change at the end of the heated section. If, for example, the flow cross section of the heated section is 40% of that beyond the end of the section, then the equivalent piston will only have moved about 12 cm rather than 30 cm when the heated portion is empty. In this case (see Figure 25) the pressure at expulsion is about 42 atm rather than 60 atm.

B. TREAT

1. Operations

Two metal-water reaction samples of SPERT-I-type fuel were irradiated in transparent capsules to investigate the feasibility of using high-speed motion pictures to gain additional information on the phenomena that influence the extent and rate of metal-water reactions obtained when fuel

samples are rapidly heated in water. Analysis of the motion pictures showed that the technique is feasible, but the fuel-sample container tube must be redesigned because the square quartz tube used in these experiments darkened during the transient. Apparently impurities were introduced into the quartz during the forming operation, since flat quartz windows have not shown any tendency to darken during previous transients.

Metal-water reaction scale-up experiments 202 and 203 were irradiated. Capsule 202 contained a cluster of nine Zircaloy-clad UO_2 pins; capsule 203 contained a similar cluster of pins clad in stainless steel.

Two EBR-II-type fuel pins were irradiated in package sodium loops. One sample was tested in flowing sodium and the other one was tested in stagnant sodium.

2. Large TREAT Loop

Installation of the main 3-in. piping, tanks, and valves for the loop has been completed, with the exception of three expansion bellows that have not been received from the supplier. All welds have been radiographed, and the completed sections of the piping system have passed a helium leak test. Installation of pipe heaters is in progress.

The Keller Corporation (the fabricator selected for the construction of the surge suppressor) reports that the necessary tooling for fabricating the diaphragm segments is 80% complete. The metal for the fabrication of these diaphragms is on site. Stamping is to be started by the end of this month, with assembly welding to follow immediately.

The remainder of the test section is estimated as 66% complete.

Additional components which have been placed on order include the two bellows which are needed as replacements for the units damaged in previous operations. The delivery of these units from the Fulton Sylphon Company is expected during the last week of May.

The liner, which serves to isolate the test-section region from the reactor core, has been redesigned. The redesign employs Zircaloy instead of stainless steel in order to reduce neutronic perturbation. This substitution resulted in a number of physical changes which are now integrated into the system. Vendors are being contacted to supply the necessary materials.

3. Integral Sodium Loop

The first package sodium loop was used to carry a fuel specimen just to the threshold of failure in sodium, as an initial transient experiment in TREAT. The loop functioned satisfactorily during the experiment, and a

postmortem examination of the fuel specimen furnished a basis for specifications of the test conditions for the first multi-loop series of meltdown experiments. The purposes of this series (Series XLVI) of TREAT experiments may be summarized as follows:

- (1) to provide a further check on the threshold of failure, as determined from the initial experiment;
- (2) to measure the magnitude of pressure pulses associated with sample failure;
- (3) to determine the radiation sensitivity of the pressure instrumentation under experimental conditions for each loop, both under flow and no-flow conditions;
- (4) to measure the transient flow perturbations during the experiments;
- (5) to examine the modes of sample failure and determination of fuel movement.

Some minor modifications were made in the loops following the first experiment, to facilitate loading and post-experiment handling, but the basic design of the loops remains unchanged. As in the first experiment, each loop loading consists of a single EBR-II fuel sample, surrounded by six steel dummy EBR-II pins, to present a flow passage closely comparable to that for an EBR-II pin inside an EBR-II subassembly.

Two completed package sodium loops of the series have been loaded and shipped to the TREAT facility for testing. A third loop will be shipped in the near future.

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

1. Reactions of Fuel and Cladding Metals with Liquid Sodium

A program is underway to determine the nature and magnitude of any safety problems that might arise in the event that abnormally hot fuel and cladding materials come in contact with the coolant (usually sodium) in a fast reactor. The methodology and experience gained in work on metal-water reactions are being exploited in these studies.

Studies of the interaction of fuel and cladding materials with molten sodium, using the levitation method (see Progress Report for May 1964, ANL-6904, p. 102), have been initiated. The experiment consists of heating and levitating a sample of the fuel or cladding (about 0.1 cc in volume) to temperatures between 2100 and 2400°C and then dropping the sample into a container of liquid sodium maintained at 250°C. The apparatus has been installed in a glovebox with an argon atmosphere.

Niobium, niobium-1% zirconium, tantalum, and molybdenum samples were dropped as solid pellets; no chemical interaction of these materials with the sodium was observed.

Zirconium, stainless steel 304, nickel, and mild steel samples were dropped as molten pellets. Zirconium pellets, when heated to temperatures just above the melting point (1860°C) before dropping, remained in the form of spheres. At higher initial temperatures, the spheres flattened somewhat, cracked in spots, and at the highest temperatures studied developed the appearance of blown-out shells. Stainless steel pellets, when heated to 1600°C and dropped into sodium, formed slightly flattened spheroids. At 2100°C , the stainless steel pellets formed pancake-shaped solid residues; at 2400°C , they fragmented violently into particles which ranged in size from about 0.7 mm to about 0.04 mm (as determined by sieve screen analysis). Considerable violence accompanied the pellet breakup, and approximately 25 cc of sodium was ejected from the container. Similar behavior occurred when nickel and mild steel pellets were dropped into the sodium, although the breakup at the highest temperatures (2100 and 2400°C for nickel and mild steel, respectively) appeared to be somewhat less violent than that which occurred with stainless steel.

It appears at the present time that the violence occurring during pellet breakup results from the transient formation of sodium vapor by rapid subcooled boiling. Visual examination of the fragmented metal particles indicated that they had not reacted or alloyed with sodium. Chemical and metallographic studies are being carried out to determine the validity of this observation.

V. PUBLICATIONS

Papers

CORROSION OF STAINLESS STEELS BY ZINC VAPOR

G. A. Bennett, P. A. Nelson, and Leslie Burris

Trans. Met. Soc. AIME, 233, 1032-1036 (May 1965)

REACTION OF FLOWING STEAM WITH REFRACTORY METALS.

I. MOLYBDENUM (1100-1700°C)

Martin Kilpatrick and S. K. Lott

J. Phys. Chem. 69, 1638-1640 (May 1965)

THE URANIUM-URANIUM DIOXIDE PHASE DIAGRAM AT HIGH TEMPERATURES

A. E. Martin and R. K. Edwards

J. Phys. Chem. 69, 1788 (May 1965) Letter

TEMPERATURE TRANSDUCER FOR THERMOGRAVIMETRIC STUDIES

J. G. Schnizlein, John Brewer, and D. F. Fischer

Rev. Sci. Instr. 36, 591-593 (May 1965)

CONTROL DRIVE MECHANISMS FOR THE EBR-I REACTOR

B. C. Cerutti

Proc. Mtg. on Fast Reactor Control Mechanisms Held at Germantown, Md., Sept. 16-17, 1964. WASH-1054, Pt. 1, pp. 16-34

REVIEW OF FAST REACTOR CONTROL FUNDAMENTALS

R. R. Smith

Proc. Mtg. on Fast Reactor Control Mechanisms Held at Germantown, Md., Sept. 16-17, 1964. WASH-1054, Pt. 1, pp. 2-14

VIBRATORY COMPACTION. I. COMPACTION OF SPHERICAL SHAPES

J. E. Ayer and F. E. Soppet

J. Am. Ceram. Soc. 48, 180-183 (April 1965)PHASE STUDIES IN THE SYSTEMS UP- UO_2 AND UP-UN

Y. Baskin

Bull. Am. Ceram. Soc. 44(4), 393 (April 1965) AbstractSOLID SOLUBILITY AMONG NaCl -TYPE URANIUM COMPOUNDS

Y. Baskin and G. D. White

Bull. Am. Ceram. Soc. 44(4), 395 (April 1965) AbstractREACTION SINTERING OF URANIUM SESQUISILICIDE (U_3Si_2)

A. J. Carrea and J. H. Handwerk

Bull. Am. Ceram. Soc. 44(4), 395 (April 1965) Abstract

SUPERHEATED STEAM CORROSION OF ZIRCONIUM ALLOYS CONTAINING SMALL AMOUNTS OF IRON AND COPPER OR NICKEL

Sherman Greenberg and C. A. Youngdahl
Corrosion 21, 113-124 (April 1965)

BEHAVIOR OF URANIUM SULFIDE FAST-REACTOR-TYPE FUEL SPECIMENS UNDER TRANSIENT HEATING IN TREAT

L. E. Robinson, C. E. Dickerman, Richard Carlander,
Charles August, and C. J. Mueller
Nuclear Appl. 1(2), 168-175 (April 1965)

DEFORMATION CHARACTERISTICS OF URANIUM DIOXIDE

C. R. Tottle
Bull. Am. Ceram. Soc. 44(4), 393 (April 1965) Abstract

FABRICATION AND EVALUATION OF W- UO_2 CERMETS FOR ROCKET REACTOR APPLICATION

D. E. Walker, Y. K. Yoon, and R. A. Noland
AIAA Bull. 2, 247 (May 1965) Abstract

OXIDATION STUDIES IN AIR AND WATER-SATURATED ARGON OF COMPOSITIONS IN THE SYSTEM UC-US

G. D. White
Bull. Am. Ceram. Soc. 44(4), 393 (April 1965) Abstract

PHASE CHANGES IN THE SYSTEM UC-UP

G. D. White and Y. Baskin
Bull. Am. Ceram. Soc. 44(4), 395 (April 1965) Abstract

AN EXPERIMENTAL INVESTIGATION OF AIR BUBBLE MOTION IN A TURBULENT WATER STREAM

J. L. Baker and B. T. Chao
A.I.Ch.E. Journal 11(2), 268-273 (March 1965)

FARET CONTROL DRIVES

Ernest Hutter
Proc. Mtg. on Fast Reactor Control Mechanisms Held at
Germantown, Md., Sept. 16-17, 1964. WASH-1054, Pt. 1,
pp. 78-86

EBR-II CONTROL DRIVES

Ernest Hutter
Proc. Mtg. on Fast Reactor Control Mechanisms Held at
Germantown, Md., Sept. 16-17, 1964. WASH-1054, Pt. 1,
pp. 36-70

REFRACTORY METAL FAST REACTORS FOR NUCLEAR ROCKET PROPULSION

J. F. Marchaterre, J. J. Dickson, B. M. Hoglund, and V. M. Kolba
AIAA Bull. 2, 207-208 (May 1965) Abstract

NUCLEAR ROCKET PREHEATER CONCEPTS FOR THE GRAPHITE AND FAST REACTORS

W. H. Rettig and P. F. Fox

AIAA Bull. 2, 249 (May 1965) Abstract

IMPROVED RATIONAL ESCAPE PROBABILITY IN LUMPED ABSORBERS

C. N. Kelber

Nucl. Sci. Eng. 22, 244-247 (June 1965)

A SIMPLE ESTIMATE OF THE EFFECTS OF RESONANCE INTERFERENCE

C. N. Kelber

Nucl. Sci. Eng. 22, 120-121 (May 1965) Letter

RECENT DEVELOPMENT IN THE PHYSICS DESIGN OF SOME NUCLEAR ROCKETS

W. B. Loewenstein, K. K. Almenas, R. C. Doerner, and W. G. Knapp

AIAA Bull. 2, 249 (May 1965) Abstract

A COMPUTER CONTROLLED DISPLAY SYSTEM FOR NUCLEAR EXPERIMENTS

J. F. Whalen

Proc. European-American Nuclear Data Committee Conf. on the Automatic Acquisition and Reduction of Nuclear Data, Karlsruhe, Germany, July 13-16, 1964, ed. K. H. Beckurts, W. Gläser, and G. Kruger. Gesellschaft für Kernforschung, Karlsruhe, 1964, pp. 399-408.

AN INTRODUCTION TO MULTICHANNEL ANALYZERS

G. S. Stanford

Nucl. Instr. Methods 34, 1-9 (1965)

INFLUENCE OF THE RADII OF NEIGHBORING IONS ON THE OPTICAL ABSORPTION OF THALLIUM IN ALKALI HALIDES

A. K. Ghosh

J. Chem. Phys. 42, 2978-2979 (April 15, 1965)

ULTRAVIOLET ABSORPTION OF GLASS

A. K. Ghosh

J. Chem. Phys. 42, 2623-2624 (April 1, 1965)

THERMOELECTRICITY IN IRRADIATED DIELECTRICS. I. SOME GENERAL FEATURES

F. C. Hardtke

J. Chem. Phys. 42, 3000-3009 (May 1, 1965)

The following appeared in Atoms for Peace Conference, 1964. Third United Nations Intern. Conf. on the Peaceful Uses of Atomic Energy, Geneva, August 31-September 9, 1964. Report by the U. S. Delegation, U. S. Atomic Energy Commission, Washington, D. C.

SODIUM-COOLED FAST BREEDER REACTORS

L. J. Koch, F. S. Kirn, G. W. Wensch, C. E. Branyan, and
E. L. Alexanderson
pp. 84-85 Abstract

DEVELOPMENTS IN NUCLEAR SUPERHEAT

Meyer Novick, C. B. Graham, D. H. Imhoff, R. E. Rice, and J. M. West
p. 89 Abstract

COMPONENTS FOR SODIUM REACTORS

H. O. Monson, W. J. Hallett, J. J. Moribito, and F. A. Smith
pp. 93-94 Abstract

FUEL CYCLE ECONOMICS OF FAST REACTORS

L. E. Link, G. J. Fischer, and E. L. Zebroski
pp. 95-96 Abstract

FABRICATION OF SOLID FUELS FOR FAST REACTORS

R. E. Macherey, F. J. Leitz, and A. A. Shoudy
p. 100 Abstract

IRRADIATION BEHAVIOR OF CERAMIC FUELS

T. J. Pashos, D. R. DeHalas, D. L. Keller, and L. A. Neimark
pp. 100-101 Abstract

IRRADIATION BEHAVIOR OF METALLIC FUELS

J. H. Kittel, T. K. Bierlein, B. R. Hayward, and W. C. Thurber
p. 101 Abstract

RADIATION-INDUCED CHANGES IN THE PROPERTIES OF NON-FUEL REACTOR MATERIALS

J. C. Tobin, A. D. Rossin, and M. S. Wechsler
pp. 102-103 Abstract

CORROSION IN AQUEOUS SYSTEMS

J. E. Draley, J. A. Ayres, W. E. Berry, E. Hollner, and S. P. Rideout
p. 103 Abstract

CLADDING MATERIALS FOR NUCLEAR FUELS

W. P. Chernock, R. M. Mayfield, and J. R. Weir, Jr.
p. 106 Abstract

PHYSICS OF FAST REACTORS

Robert Avery, P. Greebler, H. H. Hummel, R. N. Hwang,
David Meneghetti, P. A. Moldauer, J. B. Nims, and A. B. Smith
pp. 107-108 Abstract

FAST CRITICAL EXPERIMENTS AND THEIR ANALYSIS

F. W. Thalgott, G. H. Best, S. G. Carpenter, W. G. Davey, W. Y. Kato,
J. K. Long, and H. A. Morewitz
pp. 110-111 Abstract

SOME NUCLEAR AND SAFETY CONSIDERATIONS IN THE DESIGN OF LARGE FAST POWER REACTORS

David Okrent, K. B. Cohen, and W. B. Loewenstein
p. 112 Abstract

FLUID DYNAMICS, STABILITY, AND VAPOR-LIQUID SLIP IN BOILING REACTOR SYSTEMS

P. A. Lottes, W. H. Cook, K. F. Neusen, R. M. Wright, S. M. Zivi,
and N. Zuber
pp. 117-118 Abstract

FLUORIDE VOLATILITY PROCESSES FOR THE RECOVERY OF FISSIONABLE MATERIAL FROM IRRADIATED REACTOR FUELS

R. C. Vogel, W. H. Carr, G. I. Cathers, Jack Fischer, L. P. Hatch,
R. W. Horton, A. A. Jonke, R. P. Milford, J. J. Reilly, and
G. Strickland
p. 119 Abstract

PYROMETALLURGICAL AND PYROCHEMICAL FUEL PROCESSING

Leslie Burris, K. M. Harmon, G. E. Brand, E. W. Murbach, and
R. K. Steunenberg
pp. 119-120 Abstract

CRITICAL FLOW PHENOMENA IN TWO-PHASE MIXTURES AND THEIR RELATIONSHIPS TO NUCLEAR SAFETY

H. S. Isbin, H. K. Fauske, Michael Petrick, C. H. Robbins,
R. V. Smith, S. A. Szawlewicz, and F. R. Zaloudek
pp. 122-123 Abstract

SAFETY ASPECTS OF THE EBR-II REPROCESSING FACILITY

Milton Levenson, C. E. Stevenson, and J. C. Hesson
p. 125 Abstract

FAST REACTOR SAFETY RESEARCH

W. J. McCarthy, C. E. Dickerman, A. E. Klickman, and
R. B. Nicholson
pp. 127-128 Abstract

Book

NONDESTRUCTIVE TESTING

Harold Berger

U. S. Atomic Energy Commission, Division of Technical Infor-
mation Extension, Oak Ridge, Tennessee, April 1965

ANL Reports

- ANL-6750 IRRADIATIONS OF U-20Pu-10Fs ALLOY FUEL RODS
W. N. Beck, J. H. Kittel, and R. J. Fousek
- ANL-6781 ANALYTICAL AND EXPERIMENTAL INVESTIGATION OF
A NUCLEAR REACTOR SUPPORT STRUCTURE
A. H. Marchertas, S. H. Fistedis, and R. O. Brittan
- ANL-6901 ENGINEERING DEVELOPMENT OF FLUID-BED FLUORIDE
VOLATILITY PROCESSES. Part 5. Description of a Pilot-
scale Facility for Uranium Dioxide-Plutonium Dioxide
Processing Studies
G. J. Vogel, E. L. Carls, and W. J. Mecham
- ANL-6963 ELECTRICAL END LOSSES IN AN MHD CHANNEL OF
LINEARLY VARIABLE CROSS SECTION
Jerzy R. Moszynski
- ANL-6979 ENGINEERING DEVELOPMENT OF FLUID-BED FLUORIDE
VOLATILITY PROCESSES. Part 7. The Corrosion of Nickel
in Process Environments
A. A. Chilenskas and G. E. Gunderson