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Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

December 1965

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

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Development Report

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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

December 1965

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FOREWORD

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

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

The last six reports issued
in this series are:

June 1965	ANL-7071
July 1965	ANL-7082
August 1965	ANL-7090
September 1965	ANL-7105
October 1965	ANL-7115
November 1965	ANL-7122

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REACTOR DEVELOPMENT PROGRAM

Highlights of Project Activities for December 1965

EBR-II

Three separate runs were made, and the total integrated power accumulated is now 5050 MWdt and 1375.5 MWde. Ten subassemblies, five control rods, were replaced.

A minor chemical explosion occurred on December 26 during removal of the interbuilding coffin from the equipment airlock, but no personnel were injured, nor were any radioactive materials involved.

Plutonium Recycle Program (EBWR)

New theoretical calculations by the THERMOS code with the use of cylindrical, one-dimensional, 4-group diffusion-theory calculations led to a value of 20.1 elements for the initial clean critical mass of the plutonium zone as compared with the experimentally used 22 elements. The earlier non-THERMOS calculations predicted 17.7 elements. Also, the boric acid concentration necessary for criticality with the 36-element loading is now calculated to be 5.14 g/gal. compared to the non-THERMOS calculation of about 7 g/gal. The corresponding experimental value is 5.09 g/gal.

Measurements have been made of the worths of the central control rod in relation to boric acid concentrations for the 36-element loading of plutonium-bearing fuel. The minimum boric acid concentration for which the eight-rod shutdown criterion can be satisfied is 1.32 gm/gal.

AARR

The maximum reactivity effect of voiding of the ITC to various degrees has been measured in the criticality facility using the 810-fuel-plate, 810-poison-strip loading. The maximum effect is 1.5% reactivity at about 50% voids.

For the physics-reference preliminary core design, reactivity control by xenon and samarium has been computed at 2.85% for 100-MW and 3.5% for 240-MW operation, both at the beginning of core cycle. The graded loading of U^{235} necessary to achieve a maximum radial to average power distribution ratio of 1.8 has been calculated.

A reference Mark I Core Package design has been established.

Further development of circuitry for control drive magnetic clutch scram actuation has reduced the time from power interruption to initial rod motion to 4 msec.

The design and placement of nuclear instrumentation has been modified in coordination with beam tube space requirements to reduce the number of measuring decades required, to eliminate reactor vessel penetrations, and to enhance response time and accuracy by increasing thermal neutron flux "seen" by the instrumentation.

I. LIQUID-METAL-COOLED REACTORS

A. EBR-II

1. Operations

Reactor Runs 12, 13A, and 13 were completed. In Run 12, concluded as scheduled at 4750 MWdt, fuel handling was accomplished in 31 hr: 18 hr for seal melting and preparatory procedures, and 13 hr for fuel-transfer operations. A total of ten subassemblies were replaced, including five control rods. A joint ANL-GE experimental irradiation subassembly designated XA08 was loaded, along with the first three short pin driver fuel (Mark I-A, 52% U^{235}) subassemblies.

Run 13A was devoted entirely to the acquisition of physics data. Control rods were recalibrated, power coefficients determined, and relative worths of Mark-I-A subassemblies determined in Rows 3 and 4 (see Section I.A.5).

Run 13 was concluded after 300 MWdt power was accumulated, to bring the total integrated power to 5050 MWdt.

Seven inner blanket enriched and one core-type subassemblies were loaded into the reactor after conclusion of Run 13. The last inner blanket enriched subassembly which was to be loaded into the core was determined to have either a damaged or out-of-tolerance top adapter. The top adapter of this subassembly would not fit properly into the main gripper jaws, so it was returned to the fuel storage basket.

During a subsequent operation, which involved removing the interbuilding coffin from the equipment airlock, a minor chemical explosion, occurring on December 26, damaged the airlock door on the reactor side. There were no personnel injuries, and radioactive material was not involved. The explosive mixture was apparently produced by hydrogen from the storage batteries on the interbuilding coffin. No physical damage to the reactor or its auxiliary equipment or systems is apparent at this time.

The cause of the explosion is currently under investigation and will be reported more fully at a later date.

A complete mechanical and electrical recheck of the fuel-handling and reactor control systems is in progress. All reactor interlock and safety circuits have also been rechecked and found to be satisfactory.

The accumulated power from the start of Run 12 to the end of this reporting period is 770.0 MWdt; total integrated reactor power at month's end is 5050.0 MWdt and 1375.5 MWde.

2. Systems and Components

a. Primary Purification System. Tests of the primary-sodium-system plugging meter placed in the secondary system (see Progress Report for November 1965, ANL-7122, p. 1) were completed. The plugging meter was reinstalled in the primary system, and the first run indicated a plugging temperature of 370°F, a value higher than any previously measured in that system. It is assumed that the process of heating the cold trap to eliminate the plugging had redeposited some oxide in the primary systems.

When the cold trap was placed back in operation, it was found that if the minimum indicated trap temperature was held at 290°F or greater, there was no tendency toward plugging. The trap was held at this temperature to determine the equilibrium plugging temperature. After operation for approximately one week, the plugging temperature had decreased to less than 250°F, indicating an apparent temperature error in the cold trap of approximately 40°F. The plugging previously noted, therefore, may have been due, at least in part, to sodium freezing in the lower temperature section of the trap when operated at the normal indicated temperature of 250°F.

3. Reactor Improvements

a. Irradiation Subassembly Development. In order to gain maximum utilization of EBR-II as a fuel and materials irradiation facility, subassemblies must be provided to accommodate the special requirements and restrictions imposed on these irradiations. The subassemblies are uninstrumented and are designed to be handled by the normal fuel-handling equipment.

To date, nine irradiation subassemblies have been placed into the reactor, eight of which are currently in the reactor, one having been removed after completing its scheduled irradiation. These nine irradiation subassemblies have all been of the earliest design (Mark A) in which the subassembly accommodates only encapsulated specimens. The clad fuel or material specimens, therefore, are exposed to stagnant sodium, as opposed to flowing sodium. Figure 1 shows the Mark-A subassembly in which spacing of the 0.375-in.-OD capsules is accomplished by locating each capsule within a dimpled shroud tube which directs the coolant flow over the capsule surface. Each subassembly holds 19 capsules and, by use of the appropriate lower adapter, may be located anywhere in the core or radial blanket. Inside the lower adapter is an orifice plate which, in combination with the adapter inlet holes, can provide the desired sodium flow at any value up to about 64 gpm. The maximum allowable diameter of the clad specimen is approximately 0.323 in., and the maximum length approximately 37 in.

A preliminary design of the Mark-B irradiation subassembly, which will permit the use of either encapsulated or unencapsulated specimens, has recently been completed (see Fig. 2). The unencapsulated samples will be exposed to the flowing coolant. No shroud tubes are employed; spacing is accomplished by helical wire wrap. Four different models will permit loading with 61, 37, 19, or 7 capsules (or full-length specimens), the respective maximum allowable outside diameters of the four different models being 0.220, 0.290, 0.375, and 0.750 in. The overall length of the test capsules is approximately $60\frac{7}{8}$ in. Again, an internal orifice plate enables setting the desired coolant flow rate. The Mark-B design is commensurate with probable later development of remote reloading capability, which would enable multiple, successive steps of specimen irradiation and examination.

Fig. 1. Mark-A Irradiation Subassembly
(Dimensions in inches)

Fig. 2. Mark-B Irradiation Subassembly

The prototype of the 37-capsule Mark-B subassembly is currently being tested hydraulically in a water test loop. A preliminary design has been completed for the 19- and 61-capsule subassemblies.

Plans have been made for a Mark-C irradiation subassembly. Although quite similar to the Mark B, the Mark-C subassembly test specimens will be contained in a smaller hex tube to allow some of the sodium flow to be bypassed (between this hex tube and the larger one surrounding it). This will permit higher coolant temperatures within the test section. The high-temperature sodium leaving the test section will mix with the cooler bypass sodium and leave the subassembly within the allowable temperature range for the reactor.

A Mark-D irradiation subassembly is under consideration. Besides those features of Mark C, it will have fuel elements below the core section to raise the sodium temperature at the test region inlet. The use of these nuclear heating elements will necessarily limit the length and number of irradiation capsules allowable.

b. Constant Power Supply. The existing motor-generator sets are being replaced with solid-state inverters to be used with either static or electromechanical switches. The existing battery charger and the batteries will be kept.

The Exide Battery Company, in conjunction with Thompson-Ramo-Woolridge, manufactures a static, standby a.c. power system that will satisfy this requirement. Two such smaller units, in operation at the NRTS, were observed and investigated. They are well packaged, with access to all parts and components, and have been troublefree for over six months. Frequency and voltage stability have been checked, and the error found to be less than advertised.

c. Trace Heating System. The design of the trace heating scanning system to replace the Panascan in the secondary system has been completed, and the fabrication and assembling of component parts is approximately 85% completed. The detailed plans required for interchanging the old and new systems have been completed, and the interconnection drawings completed in part. Replacement control panels have been manufactured, and some wiring, prior to installation, have been completed. The work will be completed in two steps. In the first step, the scanning equipment will be replaced. In the second step, the power-handling equipment will be replaced. The first step is approximately 60% complete. The second step is approximately 5% complete.

d. Tank Immersion Heaters. Two additional redesigned primary tank heater plugs have been received from Central Shops. The last two of a total of six plugs have been shipped; the remainder of the heater elements (15) were shipped December 17.

The heater elements have been installed in two heater thimbles. One has been filled with sodium and is ready for installation in the primary

tank. The main-power-supply panelboard for the new primary tank heaters has been completed and is ready for installation in the reactor building.

Overall, this project has been divided into three phases, as follows:

1. aging new tank heaters;
2. proof-testing the silicon-controlled rectifiers and trigger circuit components;
3. designing and fabricating control components.

(i) Phase 1. The individual tank heaters, as they were received from the manufacturer, have been aged by energizing the heaters from a controlled power source. The minimum aging period has been set at 72 hr. Prior to aging, the individual heaters are checked for heating element resistance and for resistance to ground as a check on the mineral insulator. The power to the heating elements is increased in steps until the protective sheath reaches a temperature of approximately 700°F, which is held for 48 hr. Measurement of resistances is repeated for comparison with those taken earlier. A third set of readings is taken following cooldown of the heating elements. Of the units checked to date, all but two have improved by reading a much higher resistance to ground. Two of the units failed by essentially shorting between the heating element and the external protective sheath.

(ii) Phase 2. The primary control component in the voltage-control unit to be used for the heater control is a 235-Amp, silicon-control rectifier. Although the silicon-control rectifiers (SCR) have been tested by the manufacturer, field data are scarce, particularly on the large-capacity, high-voltage units. A test SCR unit has been set up in the Sodium Boiler Plant on a section of the surge tank heaters (110 kW).

The SCR's will definitely handle the full load primary tank heater current with a minimum of external cooling. The SCR control is satisfactory and will provide a variable voltage from essentially zero to full voltage for primary tank heat control. The heat sinks provided are adequate.

(iii) Phase 3. Following the SCR test, the equipment is packaged and will be used as one of six control units for the primary tank bulk sodium heaters.

e. Reactor Instrumentation. Solid-state amplifiers to replace the vacuum-tube millivolt-to-milliampere amplifiers for reactor instrumentation have been selected, and thirty units ordered. The amplifier selected has been checked for fail-safe features, zero stability, ease of maintenance and range change, and linearity over the operating range. The unit selected was the best in all categories tested.

Complete redesign of the flux and flow deviation circuits has been in progress. The design of the flow circuit is essentially completed. The design of the flux circuit will require approximately one man-month to complete.

The primary-tank bulk-sodium-level indicating and control equipment is being redesigned to reduce the spurious scrams resulting from malfunction of the existing equipment. The new design will be to reduce the operating range and provide a wider range of output signal for alarm (shut-down) amplifier operation.

4. Fuel Surveillance

Sodium bond heights in fuel elements from subassembly C162 were measured. This subassembly, which had been fabricated from previously irradiated and reprocessed fuel and had received a nominal 1.0% burnup after reprocessing, indicated no significant differences from the original Mark-I fuel exposed for the same burnup period.

5. Reactor Physics

A series of trapezoidal oscillations was conducted during Run 13A at 500 kW and 40 MW. The analysis indicated no change in the reactor feedback below a 20-sec time constant.

Control rods were recalibrated, rods 3 and 12 by period measurement and the remaining rods by intracomparison with these two. Table I gives the values measured during Run 13A as compared to measurements of the same rods taken during the wet critical experiments. The variation between control rod worths reflects the asymmetry in core loading, and the number of fuel subassemblies and experimental subassemblies adjacent to a given control rod.

TABLE I. Control Rod Worths

Even Rod No.	Inhours		Odd Rod No.	Inhours	
	Run 13A	Wet Critical		Run 13A	Wet Critical
2	139	158	1	129	127
4	135	158	3	118	130
6	150	157	5	122	128
8	162	156	7	135	128
10	165	161	9	-	128
12	157	157	11	137	127
Avg 151			Avg 128		

A power coefficient of 66 lh was measured for Run 13A. The power coefficient at the start of Run 13 was then remeasured and found to be 71 lh. The variation in power coefficient between the last several runs cannot be attributed simply to experimental uncertainty; since the results from the trapezoidal oscillations indicate that the prompt feedbacks are essentially unchanged (see above), the power coefficient changes are probably associated with structural effect. Use of the rod-drop techniques after control rod drive serial No. 9 is reinstalled will greatly help in our understanding the reason for the variation.

Prior to Run 13, the relative worths of Mark-I-A fuel in positions 3-F-2 and 4-B-3 were measured to be +34 and +26 lh, respectively, when compared to a standard fuel subassembly which had received 0.67% burnup.

6. Experimental Irradiations

Subassembly XA08, a joint ANL-GE irradiation subassembly, containing eight ANL mixed Pu-U carbide fuel capsules, nine structural capsules, and two GE superheat program structural capsules, was loaded into EBR-II grid position 4-F-2. The status of all experimental irradiations following completion of Run 13 follows:

Subassembly	Date Loaded	Accumulated Exposure (MWd)	Goal Exposure (MWd)	Experimenter	Grid Position
XA01	5/6/65	3420	14,000	ANL (MET)	6-D-2
XG02	7/16/65	2549	13,650	GE	7-A-1
XG03	7/16/65	2549	19,450	GE	7-D-1
XG04	7/16/65	2549	39,000	GE	7-B-1
XG05	9/3/65	2122	10,300	GE-ANL (MET)	4-C-2
XG06	9/3/65	2122	20,600	GE-ANL (MET)	4-E-2
XG07	10/27/65	1385	18,600	ANL (MET)	4-D-3
XG08	12/13/65	300	19,800	ANL (MET)	4-F-2
C220X	6/29/65	2549	3,149*	ANL (CEN)	1-A-1
C221X	7/24/65	2549	3,249*	ANL (CEN)	3-B-2
C222X	9/28/65	1728	4,328*	ANL (CEN)	5-B-2
U-1548X	5/6/65	3420	6,080	ANL (CEN)	9-B-5
U-1549X	5/6/65	3420	6,080	ANL (CEN)	11-B-6
U-1550X	5/6/65	3420	9,120	ANL (CEN)	13-B-7
U-1551X	5/6/65	3420	9,120	ANL (CEN)	15-B-8

*Minimum acceptable exposure is one-half above value.

The preliminary postirradiation examination information of the mixed PuO₂-UO₂ fuel capsules irradiated in subassembly XG01, which had a relatively short exposure of approximately 381 MWd, was received from GE. The capsule had apparently reached approximately the temperature predicted. No adverse effects were disclosed thus far in their examination.

7. Fuel-element-failure Test

A proposal has been prepared and assembly started on a second bare pin subassembly. The control rod subassembly, essentially identical to the first one irradiated on May 25, 1965 (see Progress Report for May 1965, ANL-7046, p. 1.), will be used in a second series of tests intended to yield more definite answers on the performance of the two fission product monitors.

8. Fuel Cycle Facility

A temporary second shift has been initiated to expedite the fabrication of subassemblies from Mark-I-A fuel pins produced by ANL-Illinois. Production is summarized in Table II.

TABLE II. FCF Production Summary, December 1965

Subassemblies received:	21*
Subassemblies dismantled:	15*
Subassemblies fabricated:	9
Subassemblies transferred:	14
Pins decanned:	1091
Melt refining runs:	
Irradiated fuel	8
Recycle metal	3
Average pouring yield:	
Irradiated fuel	92.8%
Recycle metal	93.4%
Injection casting runs:	7
Pin processing:	
Acceptable pins (688 additional Mark I-A)	495
Rejected pins	214
Pins welded (453 of these were Mark I-A):	1097
Leak testing (321 of these were Mark I-A):	
Accepts	1046
Rejects	12
Total	1058
Bond testing--completed runs	
(364 of these were Mark I-A):	
Accepts	747
Rejects	65
Surveillance subassemblies examined:	B306, C113, C162

*Includes C162, the first subassembly made from fuel reprocessed in the FCF, which was received and dismantled. It achieved a 1 a/o maximum burnup as part of the FCF surveillance program.

The fuel element decanning machine (see Progress Report for October 1963, ANL-6801, p. 21) has been improved by the addition of a redesigned fuel-chopper unit and a scrap-disposal unit. These units were changed to eliminate occasional jamming of the fuel pin between the decanner and chopper and to increase the efficiency of the scrap cutter. Since the units were installed, over 500 irradiated elements have been decanned and chopped without jamming. The efficiency of the scrap starting into the scrap cutter has increased from 50% to 100%.

Before the new units were installed, the fuel pin was chopped as it was being decanned. The brittleness of the fuel and the adhering property of sodium in a dry, inert atmosphere caused occasional jamming of the fuel-transfer tube which connected the decanner and chopping units. The entire fuel pin is now decanned and passes into a large-diameter tube which is later pivoted to a vertical position. The pin is then broken into short lengths by a reciprocating ram.

The redesigned fuel chopper increased the available space for the scrap-disposal unit. The opening through which the scrap must enter has been enlarged and positioned closer to the decanner tool. In addition, this unit has been made easier to repair remotely and has been equipped with remotely removable bearings.

The melt refining furnaces that failed due to induction coil melting (see Progress Report for October 1965, ANL-7115, p. 10) has been removed from the cell, decontaminated, rebuilt, and returned to the Argon Cell. It is now ready for routine use.

Metallographic examination of specimens removed from the hexagonal tube of EBR-II subassembly C104 (1.20 a/o maximum burnup) was made this month at the TAN Site. No cracking or other abnormality could be observed in any of the specimens. It is significant to note that material grain structure was characteristic of that displayed by annealed, Type 304 stainless steel. No sensitization of the material was observed. It is therefore concluded that the observation of possible cracks in the tubing material made while the subassembly was located in the FCF Air Cell cannot be substantiated. No further examinations are planned.

B. FARET

1. General

In accordance with the Commission's instructions, the FARET Project is being terminated. In view of this action, this will be the final reporting of FARET activities.

2. Summary of Major Project Milestones

The project was initiated in December 1962, with a feasibility study, and Public Law 88-72 was signed July 22, 1963 authorizing the work. During the ensuing period the major milestones were as follows:

June 1963	Start Title I
November 1963	Start Title II
April 1964	Assign Key Construction Contractor Personnel
May 1964	Complete Site Preparation Design Package for Start of Construction
May 1964	Submit Preliminary Safety Analysis Report (PSAR) to Commission for Approval
August 1964	Initiate Reactor Vessel Procurement (Design Package II)
March 1965	Complete Design Packages III and IV, Liquid Metal Pumps and Heat Exchangers
April 1965	Select Reactor Vessel Vendor
June 1965	Complete Title II Design
September 1965	Complete Reference Core I Design Report
November 1965	Complete Reactor Vessel Vendor Preliminary Design Report
November 1965	Complete PSAR Design Review by ACRS.

3. Status at Termination

The termination action came at a time when the plant design was essentially complete and the start of construction had already been long delayed. In general, the status of the work in terms of major activities was as follows:

a. The definitive design of the facility was completed by the Architect-Engineer, and all drawings and specifications had been available to the Construction Contractor since June 1965.

b. The construction plan was essentially completed by the Contractor, including erection procedures.

c. Start of construction could be initiated immediately upon authorization (this situation had existed for over a year).

d. The design documents had been assembled into construction packages ready for use by the Construction Contractor.

e. The prototype designs of in-cell equipment and reactor components were to a large extent nearing completion. In some instances prototype subcomponents were in fabrication.

f. The vendor had completed a preliminary design report for the reactor vessel and was prepared to initiate final design and major material orders.

g. The seismic analysis of the critical reactor coolant piping was complete.

h. Evaluation of the Preliminary Safety Analysis Report by the Advisory Committee for Reactor Safeguards was complete.

i. A number of significant tests to prove prototype equipment designs were about to start.

Based upon start of construction by February 1, 1966, the latest PERT Schedule indicated that FARET would have become operational in December 1970. At this time, all systems check outs, criticals, and other preparatory work would have been completed and the facility would be ready to start the first operational phase of fuel campaign No. 1.

All work is being terminated as rapidly as consistent with an orderly closeout of the job. The construction account should be phased out by the end of December 1965, with the possible exception of minor administrative tasks.

4. Control Drives

The assembly of the gripper test facility was completed. Operational checks were satisfactory.

Since the FARET control-drive development is of great importance to the development of prototype fast reactors, this work will continue with a redirection toward general application. The prototype design, as well as the platform design, is completed except for detail checking. This work is terminated.

C. General Fast Reactor Physics

1. ZPR-3

Criticality of Assembly 47 was achieved with a loading of 313.8 kg of plutonium ($\text{Pu}^{239} + \text{Pu}^{241}$). The corrected critical mass with all rods "in" is 312.4 kg of plutonium.

ZPR-3 control rod No. 10 has been calibrated several times; the calibration curve is essentially a straight line; the worth of the rod is 12.9 Ih/in. The temperature coefficient for this loading has been measured as $-3.5 \text{ Ih/}^\circ\text{C.}$

Figure 3 shows the location of three SEFOR control-rod mockups. Each rod consists of 21 matrix positions in each half of the reactor with a 15-in. stainless steel front drawer and 17-in. aluminum back drawer. Twenty inches of reflector are loaded in each half for a total rod length of 40 in. The drawer loading consists of seven $1/4$ -in. columns of nickel and one $1/4$ -in. column of 100% aluminum.

Fig. 3. Interface View of Assembly 47

In preparation for the SEFOR control rod calibration, all of the nickel in Rod No. 2 was moved 40 in. to the "out" position. Rod No. 1 was left in the "in" position. Also, all of the drawers as well as the nickel and aluminum were removed from Rod No. 3 so that gross reactivity changes may be measured in future experiments. GE-San Jose also made a correction in its sodium calculation in Region 2, the sodium, stainless steel, and void region surrounding the core. Therefore, more sodium was added to this region, and the new composition for Region 2 is now 0.392 g/cc of sodium and 1.79 g/cc of stainless steel. Additional plutonium was added to achieve criticality in this configuration.

The calibration of SEFOR Rods Nos. 1 and 2 was made by moving Rod No. 1 "out" and Rod No. 2 "in" to compensate for the reactivity change. Only the nickel, 415 kg in each rod, was moved, leaving the aluminum in place. Figure 4 shows the shape and the worth of the two rods in this case. The reactivity is given in cents, where 100 cents equals 320 lh.

Fig. 4
Calibrations of SEFOR
Rods 1 and 2

The worths of aluminum and stainless steel were measured for the total 40 in. of rod (see Table III).

TABLE III. Calibration of SEFOR Rods 1 and 2

	Cents/kg	
	Rod No. 1	Rod No. 2
Worth of nickel (averaged over rod)	0.248	0.314
Worth of aluminum (averaged over rod)	0.745	0.818
Worth of stainless steel (averaged over rod)	0.423	0.462
	<u>Overall Worth in Cents</u>	
Integral rod worth (see Fig. 4)	103.0	130.7
Change in Rod No. 1 worth due to addition of polyethylene behind boron carbide	+7.5	-
Change in Rod No. 2 worth due to polyethylene and boron carbide behind Rod No. 2	-	-6.4
Change in Rod No. 1 worth due to removing boron carbide only	-7.2	-

Due to the difference in nickel worth in the two rod positions, the effect of the boron carbide was investigated. The position of the boron carbide, shown in Fig. 3, was between the SEFOR reflector and the steel table of the ZPR-3 machine. The boron carbide region is 42 in. long and consists of drawers containing either enriched B_4C , natural B_4C , or Boral loaded so that the equivalent thickness of natural B_4C is one inch. First, polyethylene was loaded in drawers DD-12 through DD-20 in both halves. This increased the overall worth of Rod No. 1. Next, the entire region of boron carbide and polyethylene was moved to the region behind Rod No. 2. This change decreased the worth of Rod No. 1 and decreased the worth of Rod No. 2. The results are also given in Table III.

2. ZPR-6

The cell leakage (see Progress Report for November 1965, ANL-7122, p. 21) was reduced below the required 12% per 24 hr at 10 psig by applying an epoxy compound to various potential sources of leaks. The experimental program with Assembly No. 4Z (see Progress Report for May 1965, ANL-7046, p. 14) was restarted after the leakage rate was found to be acceptable.

The reduction of the data from the 1965 series of Doppler experiments with Assembly No. 4Z has been completed, and the results of 22 separate experiments are presented in Table IV. Apart from element HE-3, in which the sample was 10 in. long, all samples were 12 in. long. Three basic types of elements were used.¹ In one the sample was allowed to expand freely on heating, in a second the expansion was constrained axially, and in a third expansion was constrained both axially and radially.

The seventh column in Table IV indicates the reactor environment immediately around the sample. In every case the general environment was that of the test zone of Assembly No. 4Z, as the Doppler element was always positioned at the center of this assembly, but the environment within one drawer adjacent to the Doppler element was changed as indicated in this column. "Reactor" indicates that the drawers adjacent to the Doppler element have the average test-zone composition, but with the materials arranged somewhat more homogeneously than is true of the test zone itself. "Isolated" indicates that the U^{238} in the adjacent drawers was replaced by a quantity of molybdenum selected to hold the fluxes and adjoints in the Doppler-energy region the same as those in the "Reactor" arrangement; the "cold" U^{238} resonances in the test zone were separated in this way from the "hot" U^{238} resonances in the sample. "Reference Void" indicates an arrangement of materials approximately the same as the "Reactor" arrangement, but arranged such that when an inch of material adjacent to the Doppler element was removed to form the "Increased Void" case, the Doppler element was faced with a geometrically similar array of materials. The "1/2-in. B_4C " case indicates that 1/2 in. of natural boron carbide surrounded the Doppler element.

The results are listed as the reactivity change in inhours per kilogram of uranium metal as the material was heated from 293°K to the temperature listed.

Comparisons of the quoted results with theory are currently underway.

3. ZPR-9

The worths of selected rare earth metals and rhenium in the oxide fuel-aluminum oxide reflected Assembly No. 9 were measured. The reactivity worth of a single column of metal extending through the full axial length of the core was determined at the various radial positions. Each column consisted of five 1/16-in. plates, 2 by 2 in. square, located down the axial center of the core drawer in each half of the assembly. The reactivity effect of exchanging a drawer with the rare earth for a normally loaded drawer was determined at four different radial positions in the core.

¹Till, C. E., Gooh, E. F., and Lewis, R. A., Trans. ANS 8, 2, 455 (1965).

TABLE IV. Results of Doppler Experiments in ZPR-6 Assembly No. 42

Description of Experiment									Reactivity Change Versus Temperature									
Expt. No.	Doppler Element	Diameter of Cylindrical Sample (in.)	Material	Ratio of U^{238} to U^{235}	Expansion	Environment around Sample	Date	σ (lh)	Temp, $^{\circ}K$	293	546	845	1099					
1	N1	1	UO ₂	0.993/0.007	Free	Reactor	6/7/65	± 0.003	lh/kg U	0	-0.125	-0.214	-0.273					
2	N1	1	UO ₂	0.993/0.007	Free	Reactor	8/16/65	± 0.003	Temp, $^{\circ}K$	293	352	398	453	532	650	808	988	1093
3	N8	1	UO ₂	0.993/0.007	No Axial	Reactor	6/29/65	± 0.003	lh/kg U	0	-0.031	-0.057	-0.083	-0.115	-0.157	-0.202	-0.253	-0.273
4	N1	1	UO ₂	0.993/0.007	Free	1/2-in. B ₄ C	7/14/65	± 0.003	Temp, $^{\circ}K$	293	396	536	701	890	1083			
5	N9	3/4	UO ₂	0.993/0.007	Free	Reactor	7/13/65	± 0.006	lh/kg U	0	-0.043	-0.117	-0.175	-0.224	-0.275			
6	N5	1/2	UO ₂	0.993/0.007	Free	Reactor	6/23/65	± 0.011	Temp, $^{\circ}K$	293	546	798	1094					
7	N6	1/2	UC	0.996/0.004	Free	Reactor	7/9/65	± 0.008	lh/kg U	0	-0.031	-0.038	-0.052					
8	LE1	1	UO ₂	0.875/0.125	Free	Reactor	6/9/65	± 0.004	Temp, $^{\circ}K$	293	483	693	712	904	1090			
9	LE1	1	UO ₂	0.875/0.125	Free	Isolated	6/14/65	± 0.003	lh/kg U	0	-0.106	-0.195	-0.211	-0.262	-0.316			
10	LE1	1	UO ₂	0.875/0.125	Free	Reference Void	6/17/65	± 0.003	Temp, $^{\circ}K$	293	477	673	873	1093				
11	LE1	1	UO ₂	0.875/0.125	Free	1-in. Void Increase	6/18/65	± 0.003	lh/kg U	0	-0.089	-0.183	-0.252	-0.311				
12	LE3	1	UO ₂	0.875/0.125	No Axial	Reactor	7/6/65	± 0.004	Temp, $^{\circ}K$	293	521	776	1098					
13	LE2A	1	UO ₂	0.875/0.125	No Axial or Radial	Reactor	9/3/65	± 0.003	lh/kg U	0	-0.118	-0.206	-0.295					
14	E33	1	UO ₂	0.667/0.333	Free	Reactor	8/6/65	± 0.003	Temp, $^{\circ}K$	293	443	574	688	800	952	1098		
15	E50	1	UO ₂	0.500/0.500	Free	Reactor	8/10/65	± 0.004	lh/kg U	0	-0.060	-0.114	-0.145	-0.175	-0.210	-0.245		
16	E66	1	UO ₂	0.333/0.667	Free	Reactor	8/4/65	± 0.004	Temp, $^{\circ}K$	293	463	667	863	1096				
17	HE1	1	UO ₂	0.011/0.976	Free	Reactor	7/23/65	± 0.003	lh/kg U	0	-0.086	-0.156	-0.201	-0.245				
18	HE2	1	UO ₂	0.011/0.976	No Axial	Reactor	7/29/65	± 0.003	Temp, $^{\circ}K$	293	462	678	874	1098				
19	HE4	1/2	UO ₂	0.011/0.976	Free	Reactor	8/31/65	± 0.007	lh/kg U	0	-0.080	-0.154	-0.207	-0.259				
20	HE1	1	UO ₂	0.011/0.976	Free	1/2-in. B ₄ C	7/19/65	± 0.003	Temp, $^{\circ}K$	293	478	675	873	1103	1180			
21	HE3	1	U-Metal	0.057/0.931	Free	Reactor	8/11/65	± 0.0015	lh/kg U	0	-0.085	-0.148	-0.198	-0.245	-0.269			
22	No Sample				Free	Reactor	9/13/65	± 0.003	Temp, $^{\circ}K$	293	380	468	592	697	823	833	964	1087
									lh/kg U	0	-0.040	-0.071	-0.114	-0.141	-0.177	-0.177	-0.213	-0.239
									Temp, $^{\circ}K$	293	323	364	404	453				
									lh/kg U	0	-0.016	-0.033	-0.053	-0.069				
									Temp, $^{\circ}K$	293	477	764	1084					
									lh/kg U	0	-0.047	-0.098	-0.135					
									Temp, $^{\circ}K$	293	486	681	884	1095				
									lh/kg U	0	-0.033	-0.066	-0.086	-0.106				
									Temp, $^{\circ}K$	293	483	781	1174					
									lh/kg U	0	-0.009	-0.024	-0.041					
									Temp, $^{\circ}K$	293	487	672	875	1075				
									lh/kg U	0	+0.030	+0.043	+0.051	+0.057				
									Temp, $^{\circ}K$	293	321	368	421	471	616	827	1073	
									lh/kg U	0	+0.005	+0.011	+0.021	+0.027	+0.043	+0.056	+0.063	
									Temp, $^{\circ}K$	293	498	1091						
									lh/kg U	0	+0.016	+0.035						
									Temp, $^{\circ}K$	293	497	684	885	1099				
									lh/kg U	0	+0.030	+0.036	-0.042	+0.054				
									Temp, $^{\circ}K$	293	474	706	829	969	1020			
									lh/kg U	0	-0.001	-0.026	-0.061	-0.103	-0.114			
									Temp, $^{\circ}K$	293	1098							
									lh/kg U	0	+0.003							

The results of measurements with Re, Eu, Gd, and Dy are shown in Table V for the four different mean radii of the measured positions. The last two columns give the central worths for a light and heavy sample of the same metal measured in the center of the core. The light samples were between 20 and 30 g, and the heavy samples ranged from 300 to 1270 g. These last two columns are intended to show the effect of sample size on self-shielding. It can be seen that increasing sample sizes by factors of 10 or more decrease the reactivity worth per kilogram by ~10% excepting for the very black europium. The spatial dependence of the worths for these materials follow very closely the shapes that were measured for small samples reported for Assembly No. 7 (see Progress Report for November 1965, ANL-7122, pp. 21-31).

TABLE V. Measured Worths (lh/kg) of Rare Earths and Rhenium in Assembly No. 9 of ZPR-9

Mean Radius from Core Axis (cm)	Worth of 20-in. Axial Column				Central Worth	
	0.0	11.07	22.14	27.68	Light Sample	Heavy Sample
Re (817.2 g)	-59.8	-51.4	-41.6	-66.6	-70.7	-58.8
Eu (202.9 g)	-197.1	-169.5	-146.4	-303.6	-237	-220
Gd (299.02 g)	-51.2	-43.1	-37.1	-108.7	-70.7	-56.9
Dy (322.91 g)	-45.5	-38.7	-38.1	-114.3	-62.2	-51.2

Rossi-alpha measurements with Assembly No. 9 indicate that the predominant decay components in the Rossi-alpha (α_3 component) is associated with a 4.25- μ sec prompt-neutron lifetime. This is very close to the previously reported value for Assembly No. 7. The α_1 component, estimated to be greater than 10^6 sec^{-1} , and could not be measured with any statistical precision. The measured α_2 value was $(1.67 \pm 0.40) \times 10^5$, somewhat larger than the equivalent value of α_2 in Assembly No. 7. This may be the consequence of not being able to extract a unique value of α_1 from the measured data. The value of α_3 was measured to be $(1.55 \pm 0.03) \times 10^3 \text{ sec}^{-1}$.

4. ZPPR

The supplement to the ZPPR Preliminary Safety Analysis Report is in preparation for anticipated submission to the AEC in January 1966. The supplement will include proposed additions to the roof structure of ZPPR, safety data on filtration through sand, and details of various types of accidents being evaluated.

A review of the report on the coring of the Gravel Gertie structure was made with Eberline and Associates late in the month. This report should be ready at about the same time as the supplement mentioned above.

The following is a status report on reactor components:

a. Reactor Bed and Tables. A revised check list for inspecting the bed and table units was received and will be reviewed again. The manufacturing procedures are now being processed through the production-control, methods, and time-study areas. Orders for materials are following these procedures to the shops and to the electrical engineering group.

b. Matrix Drawers. Two ANL representatives visited the vendor, Mechanical Products Manufacturing Co., Seattle, Washington, because none of the initially submitted drawers was entirely within the specifications; therefore, they were not approved. Two additional drawers will be submitted for approval.

c. Matrix Tubes. Two ANL representatives visited the vendor, Van Vetter, Inc., Seattle, Washington, to inspect the first 100 ZPPR matrix tubes. An additional 400 tubes were near completion. Gauges, test equipment, and test procedures were inspected and reviewed, and minor items were changed to conform to the quality control procedure agreed to in a letter written to the vendor in November. Workmanship and finish on the tubes were good. ANL will be notified after every 600 tubes have been completed and inspected.

d. Fuel Rod Drives. All machining on the drives has been completed. Assembly of the first unit has started. Two ANL representatives visited the vendor, McGee and Hogan Machine Works, Salt Lake City, Utah, to inspect parts and assure proper quality control procedures in fabricating the fuel rod drives. Spot checks were made to confirm accuracy of inspection records. These checks were acceptable.

D. General Fast Reactor Fuel Development

1. Metal Fuels

a. U-Pu-Ti and U-Pu-Zr Alloys

(i) Thermal Conductivity. The thermal conductivities of U-10 a/o Pu-13 a/o Ti, U-11 a/o Pu-30 a/o Ti, U-15 a/o Pu-15 a/o Zr, and U-15 a/o Pu-25 a/o Zr have been measured. Qualitatively, the values listed in Table VI correlate with the volume fractions of the phases present in the alloys. The (U,Pu) alpha and (U,Pu)Zr₂ phases have about twice the conductivity of the (U,Pu) zeta and the (U,Pu)₂Ti phases. The addition of plutonium to these alloys will increase the zeta phase and decrease the alpha phase. Both changes lower the thermal conductivity. These data were obtained in a comparative type of apparatus² with Armco iron as the standard.

TABLE VI. Thermal Conductivities of Several U-Pu-Zr and U-Pu-Ti Alloys (a/o)

Temp (°C)	Thermal Conductivity ^a (W/cm-°C)			
	U-11 Pu-30 Ti	U-10 Pu-13 Ti	U-15 Pu-15 Zr	U-15 Pu-25 Zr
100	0.087 ^b	0.10 ^b	0.11 ^b	0.092 ^b
200	0.117	0.139	0.142	0.117
300	0.142	0.171	0.177	0.144
400	0.163	0.201	0.210	0.171
500	0.184	0.228	0.241	0.197
600	0.201	0.254	0.264	0.219
700	0.216	0.278	0.280	0.234
800	0.23 ^b	0.302	0.293	0.248
900	-	0.33 ^b	0.30 ^b	0.260 ^b

^aProbable accuracy, $\pm 10\%$.

^bExtrapolated.

(ii) Phase Transformation. The U-Pu-Zr and U-Pu-Ti alloys were repeatedly thermally cycled through their phase transformations. They were heated to 700 or 830°C and cooled to 350 or 550°C one hundred times with essentially no effect. A few, small blowholes were present in some of the pins, but the thermal cycling produced no distortion. The data show small, unrelated changes in density and length. The density changes were both positive and negative, and were probably related more to homogenizing the cast structure than to damage produced by thermal-transformation cycling. The length changes were all positive, but measurement errors on warped ends and bowed rods were probably the major cause of the increases.

²Annual Report for 1961, Metallurgy Division, ANL-6516, p. 403.

The lack of correlation leads to the conclusion that the cycling effects (if any) are masked by these other factors. Perhaps a thousand cycles or more refined measurements could produce definite effects.

2. Cladding and Jacket Development

a. Fabrication of Vanadium Alloys. The processing scale-up of V-15 w/o Ti-7.5 w/o Cr is continuing (see Progress Report for September 1965, ANL-7105, pp. 26-27). The processing generally follows procedures established for the V-20 w/o Ti binary alloy, with some variations. The heat-treated alloy can be readily cold worked to reductions of 60% with no evidence of cracking. The as-rolled hardness of 0.178-cm sheet after 60% reduction is about 300 DPH (30R_C), dropping to about 220 DPH (95 R_B) after a one-hour anneal at 900°C.

Considerable investigative work on this alloy, associated with fabricability, is in progress. Although results are preliminary, it has been determined that:

- (i) A 900°C anneal of cold-worked V-15 Ti-7.5 Cr results in optimum room-temperature mechanical properties consistent with ductility and fabricability.
- (ii) The ternary alloy will be expected to be more resistant to grain coarsening at elevated temperatures than the V-20 Ti binary during short-time exposure to elevated temperatures.
- (iii) Anneals at higher (above 1200°C) may affect the precipitate morphology to the extent that ductility is lowered and fabricability jeopardized.
- (iv) The ternary alloy is amenable to tube reducing based on a 50% tube-wall reduction accomplished on an ANL-designed three-roll tube reducer.

b. Properties of Vanadium-base Alloys

(i) Thermal Conductivity of TV-20. The thermal conductivity of the vanadium-20 w/o titanium alloy was measured in a comparative type of apparatus. Inconel and Armco standards were used to check the specimen. The values calculated from the Inconel standard are generally higher than those from the Armco standard, but the difference is usually within $\pm 5\%$ of the mean values listed in Table VII. These data are probably accurate to within $\pm 10\%$.

TABLE VII. Thermal Conductivity of TV-20^a

Temp (°C)	Thermal Conductivity ^b (W/cm-°C)
100	0.153
200	0.192
300	0.220
400	0.241
500	0.257
600	0.271
700	0.283
800	0.296
900	0.306
1000	0.317
1100	0.33 ^c

^aVanadium-20 w/o titanium alloy.

^bProbable accuracy, $\pm 10\%$.

^cExtrapolated.

hydrogen contents increased by a factor of four, while carbon and nitrogen increased by a factor of two during testing. These are significant increases in interstitial levels and may influence the creep properties.

Of the five alloys tested thus far, V-5 w/o Ti-15 w/o Cr and V-15 w/o Ti-7.5 w/o Cr have the best creep resistance under the stated test conditions. Next in order are V-20 w/o Ti, V-30 w/o Ti-10 w/o Cr, and V-30 w/o Ti-5 w/o Cr. The lower the titanium-to-chromium ratio the lower is the minimum strain rate. Since elastic-property terms appear frequently in expressions used in dislocation mechanics, it might be expected that they influence creep behavior and may therefore serve as a useful index of creep resistance. In the absence of reliable elastic-property data for these alloys in the heat-treated condition, the arithmetic mean bulk moduli of several V-Ti-Cr alloys have been calculated by a method proposed by Smith³ and are plotted against creep rate in Fig. 6.

³Smith, J. F., Elastic Constants and Their Relation to Cohesion and Stability in Intermetallic Phases, Nuclear Metallurgy Series, Vol. X, p. 397 (1964).

(ii) Uniaxial Creep. The V-5 w/o Ti-15 w/o Cr alloy has been creep tested at 650°C and 15-kg mm⁻² stress. Sheet specimens of this alloy, 0.76 mm thick and 4.2 cm long by gauge length, were machined from specimen blanks that had been rolled from arc-cast buttons by the ANL Foundry and Fabrication Group. After a one-hour anneal at 900°C, the total interstitial content (C, N, O, and H) of these alloys was about 1200 ppm. The strain-versus-time curves for the recrystallized alloys tested in a vacuum environment of less than 1×10^{-6} mm Hg pressure are shown in Fig. 5.

The interstitial analyses for the V-15 w/o Ti-7.5 w/o Cr alloy before and after 700 hr of testing are given in Table VIII. Oxygen and

Fig. 5. Creep of Annealed V-Ti-Cr Alloy Sheet Specimens Tested at 15 kg/mm² Stress and 650°C

Although a relationship between these two quantities has not yet been established experimentally, the apparent trend is toward a lower creep rate with an increasing bulk modulus.

TABLE VIII. Interstitial Analyses for V-15 w/o Ti-7.5 w/o Cr before and after Creep Testing in Vacuum ($<1 \times 10^{-6}$ mm Hg) at 650°C and 15 kg mm^{-2} Stress

Condition	Interstitial Analyses, ppm			
	C	O	H	N
Before	400	540	5.2	296
After	800	2300	23	490

Fig. 6. Log Minimum Strain Rate versus Arithmetic Mean Bulk Modulus for V-Ti-Cr Alloys Annealed at 900°C for 1 hr and Tested at 15 kg mm^{-2} Stress at 650°C in $<1 \times 10^{-6}$ mm Hg Vacuum.

(iii) Effect of Sodium on Uniaxial Creep. A series of creep tests are being conducted on 0.45-mm-thick sheet specimens of V-20 w/o Ti preexposed to 650°C sodium in a dynamic loop. Vacuum distillation analyses of two sodium samples yielded oxygen contents of 18 and 20 ppm. A test now in progress indicates that the preexposed samples may have a faster creep rate than unexposed V-20 w/o Ti, but the control specimens must be tested to verify this.

(iv) Linear Thermal Expansion. The average thermal expansion coefficient for V-20 w/o Ti has been determined from 25 to 960°C from data already reported (see Progress Report for June 1965, ANL-7071, p. 21). Figure 7 is a plot of the average expansion coefficient (α) versus weight fraction of titanium (N) for the V-Ti binary alloy. The coefficients for vanadium and titanium are handbook values for material of an unknown thermo-

mechanical history, and the straight line connecting these end points represents the coefficient of the binary alloy as a function of composition predicted by a simple law of linear mixing. The average coefficient for

recrystallized V-20 w/o Ti appears to be in good agreement with the predicted value, whereas the highly oriented rod shows considerable deviation from the predicted value. Samples of vanadium, titanium, and several binary compositions are being prepared to check this model for predicting the expansion characteristics of V-Ti alloys in greater detail.

Fig. 7

Average Linear Thermal Expansion Coefficient versus Weight Fraction of Titanium in Vanadium from 25 to 960°C

E. General Fast Reactor Fuel Reprocessing Development

1. Skull Reclamation Process

Although the skull reclamation process is being developed specifically for the recovery of the 5 to 10% of the uranium charge that is left in the crucible after melt refining discharged fuel pins from EBR-II, it is anticipated that the chemistry and technology developed in this process will also be applicable to the processing of a variety of uranium-bearing fuels.

Recent modifications have simplified the process and the operation of equipment (see Progress Report for June 1965, ANL-7021, pp. 24-25). Current testing of the simplified process is being carried out in plant-scale (4 to 5 kg U per batch) equipment consisting of: (1) a reduction furnace for carrying out reduction of the skull oxide and partial purification of the uranium in a large (14-in. OD by 26 in. high) tungsten crucible, and (2) a retorting furnace for isolating the uranium metal for return to the main fuel stream by evaporating magnesium and zinc metals away from the uranium. These two furnaces or modified versions of them will be installed in the EBR-II Fuel Cycle Facility at Idaho.

A new series of runs in the reduction furnace is in progress. A major objective of these runs is the continuation of equipment evaluation, but the runs have been designed also to verify the higher decontamination from fission products previously obtained when calcium fluoride was omitted from the flux (see Progress Report for November 1965, ANL-7122, p. 38) and to determine if oxide reduction is affected by the omission of the calcium fluoride. For these runs, the flux is 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl . (In the preceding runs with fluxes containing calcium fluoride, the flux consisted of 47.5 m/o CaCl_2 -47.5 m/o MgCl_2 -5 m/o CaF_2 .) In addition, longer contact periods at higher temperatures and more vigorous mixing will be used in the noble-metal extraction and the reduction steps

to improve fission product decontamination and uranium recovery. To improve the mixing intensity, three vertical mixing baffles were installed in the tungsten crucible and a second impeller was added to the mixer shaft.

Steps have also been taken to overcome the problem of fuming of metal and salt vapors which had been previously observed during opening of the reduction furnace at operating temperatures for charging additional materials and, to a lesser extent, during and following transfers of metal and salt solutions from the reduction furnace (see Progress Report for September 1965, ANL-7105, p. 33). For collection of the fumes during intermediate charging operations, an exhaust manifold that fits around the furnace charging port is provided; this manifold performed very well in prior runs (see Progress Report for October 1965, ANL-7115, p. 37). Fumes that accompany transferred materials will be confined in the hood of the recently installed prototype transfer receiver in which transferred materials are collected and weighed. Both the furnace exhaust manifold and the hood are connected to a filter-exhauster assembly system.

In the first runs of the new series, fume control was very satisfactory. The quantity of fumes that escaped to the inert atmosphere enclosure from the charging port and from the transfer receiver hood was reduced to a negligible value. All the fumes were removed from the exhaust gas stream (argon) by AEC-type high-efficiency filters, and the argon was returned to the inert atmosphere enclosure.

For operation of the furnace in the EBR-II Fuel Cycle Facility, a further modification in the furnace-charging equipment is being developed to reduce the fume load on the filter-exhauster system. The modification consists of incorporating a 4-in.-dia ball valve as a closure at the bottom of the charging hopper; the valve will be opened only to charge process reagents. In this way, the length of time that the hot ($\sim 750^{\circ}\text{C}$) furnace is open will be reduced from almost an hour to about a minute. The fume control system and this modification in charging equipment should satisfactorily eliminate the escape of fumes to the Argon Cell atmosphere of the Facility.

2. Processes for Future Fast Reactor Fuels

Previous reports (Progress Reports for September and November 1965: ANL-7015, p. 34, and ANL-7122, p. 39) have indicated that uranium-plutonium-fission product separations can be effected by salt-transport processes involving liquid copper-magnesium and zinc-magnesium alloys in mutual contact with molten MgCl_2 at 800°C . Salt-transport processes are those in which the fissile and fertile constituents of the fuel are selectively and incrementally transferred from one liquid metal solvent to another by cycling a molten salt phase between the two metal solvents. The selective transport of the valuable fuel constituents takes place as a result

of differences in distribution coefficients and solubilities of the fuel constituents in the two metal/salt solvent systems. Further evaluations of distribution coefficient and solubility data show that similar separations should be possible with cadmium-based alloys of zinc and magnesium as the metal solvents, and 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl as the

Fig. 8. Plutonium or Uranium Concentration in 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl Salt above Saturated Alloy at 600°C

molten salt. Figure 8 presents the estimated equilibrium concentrations of plutonium and uranium in 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl salt above uranium- or plutonium-saturated alloys of specified composition at 600°C. It is evident from these data that appreciable concentrations of uranium (>1 w/o) can be achieved in the equilibrated salts by using the solvents Cd-Mg and Cd-Zn-Mg at very low magnesium concentrations.

Cadmium-based materials offer the advantages of lower operating temperatures ($\sim 600^\circ\text{C}$), possible use of steel vessels, and the absence of nonvolatile metals that cannot be removed by retorting. They have the disadvantage, however, that fission product separation factors are lower, and multistage contacting is therefore required, particularly for the plutonium-rare earth separation.

In Fig. 9 is shown a conceptual salt-transport process for recovering plutonium from EBR-II blanket material, using the skull-reclamation equipment. The uranium blanket rods are first hydrided-dehydrided, or oxidized and reduced, or dissolved in zinc-magnesium alloy to make the plutonium available for reaction. The salt-transport principle is then used to transfer the plutonium selectively from a magnesium-rich zinc alloy to a small volume of Zn-5 w/o Mg alloy in a second crucible suspended in a molten salt over the magnesium-rich alloy.

Another conceptual salt-transfer process (see Fig. 10) is suggested for a slightly enriched uranium oxide fuel. Oxidative decladding is followed by reduction of the oxides with a magnesium-rich zinc alloy in the presence of a molten chloride salt. Plutonium is recovered in a Zn-5 w/o Mg alloy by the salt-transport procedure and then separated from rare earth fission products by a multistage extraction with a molten salt. Plutonium metal is

Fig. 9
Conceptual Salt-transport Process
for EBR-II Blanket Metal

*For example, a 50 m/o MgCl₂-30 m/o NaCl-20 m/o KCl Salt.

Fig. 10
Conceptual Salt-transport Process for
Low-enriched Uranium Oxide Fuels

recovered from the zinc-magnesium solution by retorting and reoxidized to form PuO_2 . The uranium is separated from noble and refractory metal fission products by another salt-transport step, collected in a zinc-magnesium or cadmium-magnesium alloy, recovered as the metal by retorting, and re-oxidized to UO_2 .

Scouting experiments were performed to determine if UC could be converted to UCl_3 by the following reaction:

which might be useful for dissolving UC fuel elements. Tests were made at 700°C with 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl , both in the presence and absence of zinc. The reaction did not proceed to a significant extent, indicating that this procedure would not be a satisfactory method for dissolving UC fuels. Other oxidation reactions are being investigated.

Engineering work related to the development of salt-transport processes is underway. In a demonstration run reduction and salt-transport steps at 800°C were used to recover uranium from a 6-kg unirradiated charge consisting of a mixture of oxides of uranium (5 kg), cerium, zirconium, molybdenum, and ruthenium. In the reduction step, uranium, zirconium, and nobler metal oxides in a 30 m/o MgCl_2 -30 m/o NaCl -40 m/o KCl salt were reduced by Cu-10 w/o Mg. The reduced uranium metal was present largely as a precipitated phase in the Cu-10 w/o Mg alloy. The bulk of cerium remained in the halide salt. The salt containing cerium and MgO reaction product was removed and discarded as waste.

In the salt-transport step, uranium in the molten Cu-10 w/o Mg alloy was transferred incrementally to a molten Mg-35 w/o Zn alloy by cycling MgCl_2 between the two alloys. This was accomplished by having the molten metal alloys in adjacent vessels and then alternately pressuring a charge of molten MgCl_2 through a transfer line from one alloy to the other. In each vessel the salt and metal phases were mixed to promote chemical equilibration and then the phases were allowed to separate by settling. Equilibration and settling times were 6 and 10 min, respectively, in the Cu-Mg vessel, and 6 and 5 min, respectively, in the Zn-Mg vessel.

The transfer of reduced uranium proceeded in accord with theory until 14 transfer cycles were completed. At this point a weld failed in the transfer line, and it was necessary to terminate the run. The net uranium transfer achieved at this point was about 75% of the 5 kg charged. A 99% uranium recovery would have been achieved after about 22 transfer cycles. The rate of uranium transport tails off as the source solvent becomes highly depleted in uranium. The copper concentration in the transferred uranium was 2.8 w/o of the uranium. This indicates that the entrainment of copper was excessive, and this problem of copper entrainment must be overcome. A very satisfactory zirconium removal of 97.5% was achieved. Except for

the problem of copper entrainment, the engineering-scale run is very encouraging and has successfully demonstrated the salt transport principle.

3. Eddy Current Induction Probe

Testing of a 14-in. eddy current liquid-level probe with liquid cadmium has been completed to supplement earlier studies with a 6-in. probe (Progress Report for January 1965, ANL-7003, pp. 21-22). The longer probe did not have the same absolute accuracy as the shorter probe, but the average error in measured liquid depth was about the same percentage ($\pm 2\%$) of the probe length with both probes.

4. Materials Evaluation

A supporting program to test the corrosion resistance of a new crucible material, vitreous carbon, to molten metal solutions containing zinc-magnesium-uranium/salt systems and copper-magnesium-uranium/salt systems is continuing. An analysis of all runs completed to date indicates the material is resistant to both systems (containing up to 10 w/o uranium) at 800°C. In the most recent experiments, none of the contact surface was wetted by a zinc-2 w/o uranium/salt system after 208 hr of exposure at 800°C, 10% appeared to be wetted after a further 209 hr (for a total of 417), and 50% after 238 more hours (for a total of 655),⁴ but in none of these cases did the solidified content stick to the crucible. No wetting of the vitreous carbon occurred after 321 hr of exposure to a copper-10 w/o magnesium-2 w/o uranium system at 800°C. It is concluded that vitreous carbon has excellent potential as a container material for these systems.

Another new container material, pyrolytic boron nitride, is also being tested. The interest in this material stems primarily from the fact that it can be deposited in thin sections on graphite to provide a coating that is impervious to gases. This material, deposited on a graphite crucible as a 2- to 5-mil coating, was tested by exposure to a zinc-5 w/o magnesium-2 w/o uranium/MgCl₂-30 m/o NaCl-20 m/o KCl salt at 800°C for about 400 hr. At the end of this time the crucible appeared sound, and there was no evidence of salt or metal leakage.

In other corrosion work, Croloy 16-1, a ferritic stainless steel, was exposed to a Cu-10 w/o Mg-2 w/o U/MgCl₂-30 m/o NaCl-20 m/o KCl system for 197 hr at 800°C. (Croloy 16-1 is similar in composition to Type 405 stainless steel, which has been shown to be a satisfactory containment material for many molten metal/salt systems, but Croloy 16-1 is much more readily available in tubing and pipe than is Type 405 stainless steel.) The corrosion rates in the salt and vapor phases were acceptably small; however, samples located in the metal phase and at the metal-salt interface showed evidence of the formation of an intermetallic compound, possibly a U-Fe intermetallic, to a depth of up to 16 mils.

⁴ In the first 208 hr of exposure the salt was MgCl₂-47 1/2 m/o CaCl₂-5 m/o MgF₂; in the remaining 447 hr of exposure, the salt was MgCl₂-30 m/o NaCl-20 m/o KCl.

II. GENERAL REACTOR TECHNOLOGY

A. Experimental Reactor and Nuclear Physics

1. Computer Code for Reduction of Coincidence-counting Data

A computer code COINC (ANL RP-258) has been developed to reduce most types of coincidence-counting data. Input data is presently handled either by manually or automatically punched cards. Modifications to allow the use of paper-tape input are in progress.

Complex adjustments of the accidental coincidence rates affected by deadtime are made. Both Poisson statistics for total accumulated counts and error propagation for correlated and uncorrelated quantities are compounded in determination of standard deviations.

System parameters of deadtimes and coincidence gate times for each channel, background rates, decay constants, and associated standard errors are used in formulae based on derivations published in recent literature. The output contains corrected single-channel and coincidence rates, specific disintegration rates at "zero" reference time, as well as efficiencies, error limits, and a weighted mean when more than one case is run at one time.

A very simple input format is available, allowing application to essentially any coincidence decay scheme. The program was designed for absolute $\beta\gamma$ coincidence counting, but it has also been used for $\gamma\gamma$ and neutron-fission coincidence work.

Figure 11 displays an example of the results obtained from applying the COINC code to measurements of a Mn^{56} sample, which was followed for more than 15 half-lives in the Mn^{56} coincidence-counting facility.⁵

2. High Conversion Critical Experiments (Hi-C)

Microparameter measurements in the 1.349-cm square-pitch, stainless steel-clad zone Hi-C core have nearly been completed. This core contains a central zone of 400 stainless steel-clad fuel pins surrounded by about 450 aluminum-clad fuel pins. The microparameter measurements are made at a radius of about 5 cm in the 15-cm-radius, stainless steel-clad central zone.

The U^{238} to U^{235} fission ratio, δ^{28} , has been determined as in previous cores by irradiating depleted and enriched uranium foils in flux-symmetric positions. Three irradiations were performed, each containing several sets

⁵DeVolpi, A., Porges, K. G., and Larsen, R. N., Mn^{56} Coincidence Counting Facility, ANL-6760 (Aug 1963).

of foils. Twelve independent determinations gave a value of

$$\delta^{28} = 0.0742 \pm 0.0011,$$

where the error quoted is the standard deviation of the mean.

Fig. 11. Mn^{56} Decay (followed for 39 hr [15 half-lives] in 4π $\beta\gamma$ coincidence system) with Data Processing by COINC Computer Code for Automatic Punched-card Readout. The lines of the semilog plot are simply fitted to the majority of the data with a slope corresponding to the 2.577-hr half-life used in the code. The scale factor for each curve is indicated by an arrow at the appropriate abscissa point. The deadtime correction for the first point on each curve is approximately 40%.

Data for the determination of the initial conversion ratio and the U^{238} capture cadmium ratio have been taken, but the data analysis is not complete. Data for the determination of the thermal disadvantage factor are now being collected.

3. Nuclear Constants

a. Radiative Capture. Measurements of the neutron capture cross sections of Re^{185} and Re^{187} (see Progress Report for May 1965, ANL-7046, p. 36) and of Lu^{175} and Mo^{98} (see Progress Report for September 1965, ANL-7105, p. 38) have been extended by determinations of the cross sections for Pr^{141} , La^{139} , and Yb^{176} .

In Fig. 12 the cross sections determined for Pr^{141} are compared with the values of Gibbons *et al.*,⁶ Johnsrud *et al.*,⁷ and Konks *et al.*⁸ Data for La^{139} are presented in Fig. 13 along with the data of other investigators.⁷⁻⁹ Good agreement is evident. Figure 14 shows the cross sections for Yb^{176} , for which no other data are available for comparison.

Fig. 12
Radiative Capture Cross Section
of Praseodymium-141

Fig. 13
Radiative Capture Cross Section
of Lanthanum-139

⁶ Gibbons, J. H., Macklin, R. L., Miller, P. D., and Neiler, J. H., Phys. Rev. **122**, 182 (1961).

⁷ Johnsrud, A. E., Silbert, M. G., and Barsehall, H. H., Phys. Rev. **116**, 927 (1959).

⁸ Konks, V. A., Popov, Yu. P., and Shapiro, F. L., Zhur. Eksp. Teor. Fiz. **46**, 80 (1964).

⁹ Macklin, R. L., Gibbons, J. H., and Inada, T., Phys. Rev. **129**, 2695 (1963).

Fig. 14
Radiative Capture Cross Section
of Ytterbium-176

B. Theoretical Reactor Physics

1. WIGLE-1 Calculations

Investigation into the performance of the WIGLE-1 code has continued (see Progress Report for November 1965, ANL-7122, pp. 43-45). The output of the code has been modified further. The values of the fluxes and precursor concentration are now punched on cards at every tenth time step. This supplements an earlier alteration to the code to plot the fast-flux distribution for each tenth time step on the oscilloscope.

An auxiliary code was written which uses the values of the flux and precursor concentration to calculate a neutron inventory, that is, the leakage, absorption, fission source, neutron balance, and "instantaneous" multiplication.

The following procedure is used. The material components and dimension of a one-region, one-dimensional slab are first chosen. A slab containing U^{235} and carbon with a 40-cm half-thickness has been chosen as the standard system for the present investigation.

With the standard system, a search for a specified value of k_{eff} was made with the MACH-1 code and the 16-group, Set #201 cross sections. The code yields the values of the material concentration, the flux distribution and spectrum, neutron lifetime, and collapsed two-group cross sections, with the neutron velocity for each group. The concentrations and two-group cross sections are then read into MACH-1 to recalculate the value of k_{eff} . The use of the two-group set yields a k_{eff} different than that initially specified. The output of the two MACH-1 calculations provides the input to WIGLE-1. The input data includes the two-group macroscopic cross sections, initial values of the fluxes and their corresponding equilibrium precursor concentration, mesh spacing, and the number and length of the time increments.

The neutron inventory is calculated from the punched output of WIGLE-1 by means of the auxiliary code. The calculation of the neutron inventory has shown a discrepancy in the precursor concentration. The source of the problem has not been determined.

One objective in the investigation of the WIGLE code is to determine its sensitivity to the values of the mesh-point intervals and time increments. The standard problem was run for three mesh intervals (1.0, 2.0, and 4.0 cm) and three time increments (10^{-4} , 10^{-3} , and 10^{-2} sec). Within the range of the investigation, the mesh interval did not influence the magnitude or distribution of the flux nor the neutron inventory. Increase of the time increment produced a change in the magnitude but not the distribution of the flux.

2. Evaluation of Cross Sections

The least-squares fitting code LEGENDRE has been used to obtain expansion coefficients of the differential elastic-scattering cross sections for Fe, Ni, Cr, Na, U^{235} , and U^{238} , with the Aldermaston-Winfrith data from the ENDF library serving as input. In most cases a tenth-order Legendre expansion was used, although higher-order expansions were necessary in some cases at high energies in order to obtain an adequate fit. These expansion coefficients will be combined with those previously obtained for more restricted ranges of energy for use in the MC² library.

Work is continuing on the study of multilevel interference effects in fissionable isotopes. A plotting program has been written, involving the Calcomp plotter, which will plot various multilevel and single-level cross sections as a function of energy.

3. The Argonne Reactor Computation (ARC) System

Figure 15 is a schematic diagram of the ARC System displaying details of the system capability beyond the initial (Phase 1) effort (see Progress Report for October 1965, ANL-7115, pp. 45-48).

The interaction between the ARC cross section library and the Brookhaven ENDF is displayed schematically at the left. The ANL version of ENDF is obtained on tape from the BNL ENDF center. The link between the ANL ENDF and the Argonne Master Cross Section library is by means of the RETRIEVAL PROGRAM, which is currently being developed at BNL and which will be available at Argonne when completed. The input to the retrieval program will probably consist of ENDF acquisition numbers, energy ranges, and scale factors which would be specified by the user for the purpose of generating cross section-energy correspondences in the format useful to the Standard library tape.

Fig. 15. Schematic Representation of ARC Emphasizing System Capability beyond the Phase-1 Effort

The organization of the DATAPOOL is still being considered. Some general thoughts concerning the structure of the DATAPOOL follow.

The DATAPOOL will consist of a number of files outlined below.

I. PERMANENT FILE

Contains the computational modules, the Argonne Master Cross Section library, and other semipermanent data common to all ARC runs.

II. SCRATCH FILE

This is a storage area in the DATAPOOL which is overwritten by each ARC run and used for temporary storage during a given run.

III. PROBLEM FILE

Contains the permanent record of all data to be preserved from a given ARC run. The PROBLEM FILE is organized into CATEGORIES, TYPES, RECORDS, and WORDS. These levels of information are illustrated below.

A. CATEGORY

Defined for each general class of quantity such as fluxes, cross sections, events, option flags, and geometrical specifications, within the PROBLEM FILE. Note that some CATEGORIES are dependent upon the generating module. For example, fluxes from diffusion theory must be stored in a separate storage area than that used for fluxes from transport theory, so that there is more than one FLUX CATEGORY. Other quantities, such as mesh spacing, are unique and can be contained in one CATEGORY area.

B. TYPE

Refers to a specific type of quantity within a given CATEGORY. For example, in the CATEGORY CROSS SECTION, different TYPES would be fine-group microscopic capture, coarse-group macroscopic fission, fine-group microscopic inelastic scattering, etc.

C. RECORD

A RECORD in general contains all of the data with respect to a running index contained in a given TYPE. For example, in the TYPE FINE GROUP MICROSCOPIC CAPTURE, a RECORD would contain $\sigma_c^m(1)$, $\sigma_c^m(2)$, ..., $\sigma_c^m(J)$, where 1, 2, ..., J are energy group indices and m is a material index. Each new material m would belong to a different RECORD.

D. WORD

This is the lowest level in the DATAPOOL and consists of the individual quantities making up a RECORD. For example, $\sigma_c^m(17)$ is a WORD in the RECORD MATERIAL M MICROSCOPIC CAPTURE.

Each new ARC run will generate a new PROBLEM FILE, and results from previous runs can be obtained by recalling particular WORDS, RECORDS, etc., from a particular previously generated PROBLEM FILE.

In order to begin work on ARC, specifications are required for the individual modules. In particular, the interface quantities for each module must be specified.

A glossary of the computational module interface quantities is being collected and organized consistent with the different structural levels of the DATAPOOL. Some of the DATAPOOL nomenclature referred to above is utilized in various of the module specifications. Thus, reference is made to WORD or RECORD "numbers" in the DATAPOOL since it has so far been arbitrarily assumed that locations will be assigned in the DATAPOOL by a number code. It may turn out more convenient to recall quantities from the DATAPOOL by name. Resolution of such detailed questions must await further study of the System as related to the DATAPOOL handling problem.

4. Application of the ARC System to Problems of Fuel Burnup

Two major items to be resolved in drawing up preliminary specifications for the ARC modules in the BURNUP sequences are:

- a. allowable chains of isotopic burnup;
- b. allowable schemes of fuel rearrangement.

The calculations show isotopic burnup chains can be of arbitrary length and complexity so long as the burnup matrix is triangular, that is, no reaction in an isotope labeled i gives rise to production of an isotope $j < i$. This means that n , $2n$ and n, α reactions can not be accounted for as sources of isotopes if n, γ reactions are also included. If a full-burnup matrix is required, only one algorithm may be used, and the chains may have to be restricted in length.

It is also shown that any scheme of fuel management can be allowed so long as there is atom conservation. The algorithm is independent of the number of regions or, indeed, of reactors. Efficient execution, however, will require designation of those blocks of regions which cycle among themselves, with an indication of their relative importance.

- a. Burnup. Let the abundance of isotope i in region j be denoted by N_{ij} ; since we need discuss only one region, drop the subscript j . In general,

$$\frac{dN_i}{dt} = -\sigma_{ai}\phi N_i + \sigma_{n,\gamma,i-1}\phi N_{i-1} - \lambda_i N_i + \text{other terms representing isomeric transitions, etc.}$$

Written in matrix form the equations are $\dot{N} = BN$, where B is, for example, a matrix with nonzero terms only on the main diagonal and immediately below it, if only total absorption and capture are included. An exception is the fission yield and this we propose to handle separately. The equation for N may be solved:

$$N = e^{Bt}N(0) = TN(0).$$

The matrix T may be found by straight exponentiation of B , or by first diagonalizing B to P , exponentiation of P , followed by retransformation. As long as B is lower triangular (or upper triangular) the procedure for finding T by diagonalization is independent of the number of nonzero bands in B . The only variable is the amount of storage required. Based on experience at KAPL, we expect the diagonalization routine to be the fastest routine for long chains (~30 isotopes). Straight exponentiation can be used for any irreducible nonsingular matrix B , and experimental computations are under way to determine when exponentiation is superior to diagonalization. Analytic solutions may also be used if B is lower triangular, but may be subject to cancellation errors. Clever manipulation may minimize these.

If the full matrix is required, exponentiation must be used, the matrix B must be reducible to nonsingular submatrices, and a convenient convergence test devised. In general we expect this system to work efficiently only for small chains because of the many multiplications involved.

Thus, if B is lower triangular, any of three algorithms may be used to find T ; testing will be done to determine the most efficient range for each algorithm. If B is not triangular, then only exponentiation may be used, and it is probable that only short chains can be used to avoid excessive error accumulation and running time.

Clearly, if more than one burnup chain is present in a region (i.e., B is reducible), then each chain may be handled independently.

b. Fuel Management. In region j , after irradiation for time t at flux ϕ , the concentration or output of isotope i_j is given by

$$O_{i_j} = T_{i_j}^{\ell_j}(t, \phi) N_{\ell_j},$$

where N_{ℓ_j} is the concentration of isotope ℓ in region j before irradiation, and the repeated upper and lower index means a sum over isotopes. Fuel management consists of taking a fraction A_i of each isotope from region k

and putting it in region j ; then enough feed is added in region j to bring the atom density to its original level (some of the feed may be negative representing sold material) and the process restarted. Thus the new concentration N_{ij}' is given by

$$N_{ij}' = A_{ij}^{ik} O_{ik} + C_j F_{ij}(\lambda) - R_{ij},$$

where F_j is the feed vector for region j (a fixed vector), C_j is a single coefficient for region j , and R_{ij} is the amount of isotope i rejected from region j , and λ is a parameter to adjust the isotopic ratios in F for criticality. Other parameters internal to F may be used to specify more detailed reactivity controls. The A 's are the fractions mentioned earlier, the sum being over regions. Three conditions must be met in each region:

I. Burnup: $(B_{ij}^{ij} O_{ij}) / (G_{ij}^{ij} N_{ij}) = Q_j$, an input constant;

II. Equilibrium: $N_{ij}' = N_{ij}$, or equivalently,

$$O_{ij} = T_{ij}^{\ell j} A_{\ell j}^{k} O_{\ell k} + T_{ij}^{\ell j} (C_j F_{\ell j} - R_{\ell j}).$$

III. Atom Conservation: $\sum_i N_{ij} = \sum_i N_{ij}; \quad R_{ij} = O_{ij} - A_{i\ell}^{ij} O_{ij},$

sum on ℓ , not on j ; and $C_j \geq 0$.

Condition I determines ϕ ; if there is radioactive decay, ϕ is determined by power level. Conditions II and III determine C_j , R_{ij} , and O_{ij} .

So long as $A_{i\ell}^{ik}$ are positive and their sum over region ℓ is less than 1, any arbitrary matrix A can be used. If, however, A is in block form, that is, material is cycled only within certain regions, such as only in the core or only in the blanket, then the solution can be expedited by using the following generalization of the current CYCLE technique, for example:

The N regions are divided into blocks $N_1, N_2, \dots$, such that material in any region in block N_j is only cycled into another region in N_j ; equilibrium is first calculated for a block of highest importance, other compositions being held constant. Then with the use of the average composition for the first block, the process is repeated for the second; and so on. Obviously the process may be repeated.

Fuel management at commensurate cycles is possible by making A_{ij}^{ik} equal to zero unless the irradiation time in region k is some fixed multiple of the irradiation time in region j .

Finally, the vectors R_{ij} can be viewed as inputs to a processing plant which produces a homogenized feed vector H_j . Then the substitution $C_j F_{ij} + H_j$ that replaces F_{ij} , can be made and overall system equilibrium determined.

Clearly the N regions may have any dimensionality and need not be part of the same reactor; we need only know the one-group fluxes ϕ and the elements of T , A , etc.

C. High-temperature Materials Studies

1. Ceramics

a. Thermal Conductivity of UP. The thermal conductivity of a uranium phosphide specimen, 2 cm in diameter by 8 cm long, and 93% dense, was measured in the comparative apparatus described in the 1961 Annual Report.¹⁰ Runs were made with both Armco iron and Inconel as standards. The data presented in Table IX are averages from the Armco and the Inconel runs; the Armco values were 10% higher and the Inconel values 10% lower than these means. The temperature differences measured along the uranium phosphide rod were not consistent, and average values were calculated to obtain the thermal conductivity data. Since the specimen was 93% dense, the values in column two were multiplied by 1.113 to approximate the thermal conductivity of 100% dense uranium phosphide. The probable accuracy of these data is $\pm 15\%$.

TABLE IX. Thermal Conductivity of
Uranium Phosphide

Temp (°C)	Thermal Conductivity, ^a (W/cm-°C)	
	93% dense	100% dense ^b
100	0.126 ^c	0.140 ^c
200	0.126	0.140
300	0.126	0.140
400	0.126	0.140
500	0.130	0.145
600	0.137	0.152
700	0.151	0.168
800	0.167	0.186
900	0.181	0.201
1000	0.188 ^c	0.209 ^c

^aEstimated accuracy, $\pm 15\%$.

^bFrom the formula

$$K_{th} = K_m \left[\frac{1 + (V/2)}{1 - V} \right] = 1.113 K_m,$$

where

V = volume fraction of the pores;

K_m = measured thermal conductivity;

K_{th} = theoretical conductivity.

^cExtrapolated.

¹⁰Annual Report for 1961, Metallurgy Division, ANL-6516, p. 403.

2. Modified Falex Wear Tester

The first model of the modified Falex wear tester has been received and is being checked for proper operation. Initial wear tests with materials will be performed in air and heated inert gas to gain knowledge of the wear tester characteristics. Subsequently, the model will be operated in a pot of static 1200°F sodium to observe the nature of the performance of the device in this medium.

Materials are on order for another five units to be assembled in the near future.

D. Other Reactor Fuels and Materials Development

1. Nondestructive Testing

a. Elastic Moduli of High-temperature Materials by Ultrasonics.

Ultrasound measurements have been completed at several temperatures from room temperature up to 1030°C with the first Type 304 stainless steel specimen (see Progress Report for November 1965, ANL-7122, p. 52). Above 1030°C no echoes could be seen, and the noise level of internally scattered sound was very high. Calculations of the moduli are in progress; another sample is being prepared, and measurements of it will soon be made.

b. Ultrasonic Instrument and Transducer Development. Work on backing members for ultrasonic transducer probes that contain ceramic piezoelectric materials has continued. Additional sample discs of pressed and sintered Type 316 stainless steel have been received. Measurements of the longitudinal wave velocity in these samples are in process.

c. Development of a Neutron-image-intensification System. The relative neutron-gamma response of the second neutron intensifier tube has been determined. A cobalt-60 gamma-radiation intensity of about 1800 R/hr produced an output brightness on the phosphor screen equal to that produced by a thermal-neutron intensity of 2×10^5 n/cm²-sec. These values were determined by measuring the brightness with a calibrated CdS cell.

The neutron-gamma response of the second tube compared well with that of the first tube, even though the first tube determination was made in a slightly different manner. For the first tube, an irradiated fuel capsule was used for the gamma source and the brightness equivalent was visually estimated. The value determined for the first tube indicated that a 4000-R/hr gamma intensity was equivalent to about 5×10^5 thermal n/cm²-sec. The relative neutron-gamma responses of the two tubes should be equal, since the thickness of the neutron scintillator in each tube is about the same.

d. Thermal Conductivity of Irradiated Fuel as a Function of Burnup and Temperature. The thermal-pulse method of measuring thermal diffusivity and conductivity is being utilized to determine the thermal properties of irradiated fuel pins as a function of burnup. This method will enable the diffusivity and conductivity to be measured in both the axial and radial directions at many points along the fuel pin and, thus, will indicate any anisotropy in the thermal properties as well as any variation of these properties in the axial and radial directions along the length of the fuel pin.

About half of the completed irradiated uranium-5 w/o Fs specimens have been received and transferred into the cave. Room-temperature measurements of the thermal diffusivity and thermal conductivity are in progress.

E. Engineering Development

1. Boiling Liquid-metal Technology

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

Work on the vacuum chamber consisted of revamping the work platform on the outside of the chamber together with constructing a leaktight through-flange for power feed about the entering electrical cables. The Nb-1% Zr loop, which will be mounted in the vacuum chamber, is being fitted with heaters, and the loop is being wrapped with Nb-1% Zr flat strip foil to deter oxidation of the loop itself. When this loop is mounted in the vacuum chamber, it will be temporarily protected by an inflatable plastic cover which will be supplied with filtered air from the cooling fan of the electromagnetic pump.

The pressure transducers have been completed and calibrated. The flowmeters have been attached to the loop structure. The data-acquisition system is operating properly, and a preventative-maintenance schedule has been set up.

To date, all nine thermocouple assemblies received from the vendor have been rejected because they do not meet the specifications. A calibration system is ready as soon as the vendor completes the assemblies satisfactorily.

The sodium-handling system, which is 90% complete, is dependent only upon receipt of the valve gaskets. The condenser-shutter drive mechanism has been designed and is being fabricated.

b. Heater Experiments

(i) Electron-bombardment Heater Experiment. Improvements in the heater have resulted in longer times of electron emission. The greatest improvements were attaining a higher vacuum by eliminating some bends in the diffusion-pump piping system and also by careful cleaning of the heater during assembly. Additional modifications are expected to extend emission time to useful levels.

2. General Heat Transfer

a. Heat Transfer in Double-pipe Heat Exchangers

(i) Countercurrent Turbulent Liquid-metal Flow. Difficulties related to proper convergence of a particular computational technique (see Progress Report for November 1956, ANL-7122, p. 57), have led to an alternative procedure, originally planned some time ago, for machine computation. During the past month, however, a new technique, originated by Brown¹¹ was used successfully, so that the practical significance of the study of countercurrent flow can be illustrated more definitively than before.

For a concentric tube exchanger with a "narrow" annular space, operating conditions can be characterized by three dimensionless parameters: H , K , and K_w , defined as follows:

$$H = C_2 W_2 / C_1 W_1;$$

$$K \approx (k_1 k_1^+ / k_2 k_2^+) (a_2 / a_1);$$

$$K_w = (k_1 k_1^+ / k_w) (b' / a_1);$$

where

C_i = specific heat of fluid "i";

W_i = mass flow rate of fluid "i";

k_i = thermal conductivity of fluid "i";

k_w = thermal conductivity of wall;

a_2 = width of annular space;

¹¹Brown, R. W., Jr., Resident Student Research Associate, ANL.

a_1 = radius of tube;

b' = log mean tube-wall thickness;

k_i^+ = functions of Peclet number which account approximately for the effects of turbulence.¹²

In the above subscript $i = 1$ represents the tube side of the exchanger, and $i = 2$ the annulus side. The case $H = 0.5$, $K = 0.1$, and $K_w = 0$ (Ref. 13) was treated in some detail. The significance of results obtained can be illustrated by comparing predictions of heat-exchanger lengths required for a specified heat-exchanger efficiency based on the customary design relations with those predicted by the analysis. The customary relations were based on the use of fully developed heat-transfer coefficients appropriate to the boundary condition of uniform heat flux. For the case being illustrated, the customary design relations always predicted larger lengths than would actually be required. The percent differences of these predicted lengths are summarized as a function of the specified heat-exchanger efficiency in the following:

Specified Efficiency (%)	Error in Predicted Length (%)
30	216
40	144
50	112
60	88
70	69
80	54
90	43
99	21

For certain conditions, Brown's procedure converges very slowly so that further exploratory computations with alternative procedures are desirable. Also, because of the unusual aspects of the mathematics of counter-flow systems, verification of results by completely independent procedures and/or further theoretical investigations are required.

¹²See, for example, Stein, R. P., "Heat Transfer Coefficients in Liquid Metal Concurrent Flow Double Pipe Heat Exchangers," Chem. Eng. Prog. Symp. Series No. 59, 61, 64 (1965).

¹³Values of $K_w < 0.05$ are for all practical purposes equivalent to the case of $K_w = 0$, except possibly for very small heat-exchanger efficiencies.

F. Chemical Separations

1. Fluidization and Volatility Separations Processes

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

(i) Fluorination of UO_2 - PuO_2 -Fission Product Pellets. Development studies are being performed in a 2-in.-dia fluid-bed reactor to determine optimum conditions for fluorinating UO_2 - PuO_2 pellets containing fission products. Three runs have been completed to determine the effect of the pellet to alumina weight ratio on the uranium and plutonium volatilization during fluorination. In these runs, pellet charges of 0.33, 0.65, and 1.32 kg were used with 1.1 kg of alumina, corresponding to pellet-to-alumina weight ratios of 0.3, 0.6, and 1.2, respectively. The corresponding pellet bed depths were 1, 2, and 4 in. Cesium fluoride was added to the alumina bed in proportion to the quantity of pellets charged and in the amount expected from fission product cesium.

The reaction sequence for the three runs involved: (1) a two-zone oxidation-fluorination step at 450°C, (2) a single-zone fluorination step at 450°C with 10 v/o fluorine, and (3) a recycle-fluorination step with 90 v/o fluorine at 450°C for 1 hr, at 500°C for 3 hr, and at 550°C for 8 hr. The duration of the first two steps varied with the weight of pellets charged, totalling: 2 hr for 0.33 kg pellets; 4 hr for 0.65 kg pellets; and 6 hr for 1.32 kg pellets.

The concentrations of uranium and plutonium in the final alumina beds are presented in Table X. In all runs, greater than 99.9% of the uranium was volatilized from the alumina bed during fluorination.

TABLE X. Effect of Pellet-to-alumina Weight Ratio on Fluorination of Uranium and Plutonium

Equipment:		2-in.-dia fluid-bed reactor				
Inert Bed Material:		1.1 kg sintered alumina				
Pellets:		0.48-in.-dia by 0.41-in. cylinders of UO_2 - PuO_2 -fission products; 0.4 w/o Pu, 86 w/o U, 1 w/o F.P.				
Pellet Charge (kg)	Cesium Fluoride (g)	Pellet-to-alumina Weight Ratio	Concentration of Residual Uranium and Plutonium in Alumina Bed after Fluorination			
			Uranium (w/o)	Uranium Removal (%)	Plutonium (w/o)	Plutonium Removal (%)
0.33	0.3	0.3	0.010	>99.9	0.010	92.5
0.65	0.6	0.6	0.020	>99.9	0.014	94.7
1.32	1.2	1.2	0.030	>99.9	0.024	95.3

Although the plutonium concentration in the alumina bed increased slightly with an increase in the weight of pellets charged to the reactor, the fraction of the plutonium charged which volatilized was increased by increasing the pellet-to-alumina charge ratio from 0.3 to 1.2. The percentage of the plutonium charged that is retained by the alumina is expected to decrease to an acceptably low value with reuse of the alumina. Experiments to demonstrate this concept with the system used in these runs are planned.

(ii) Phase Diagram of UF_6 - PuF_6 System. The solid-liquid phase diagram for the binary system UF_6 - PuF_6 is being studied by thermal analysis. Melting-point data for 8 mixtures are reported in Table XI. It is apparent from these results and from results reported previously (see Progress Report for August 1965, ANL-7090, p. 48) that the solid-liquid equilibria in the system UF_6 - PuF_6 involves a continuous series of solid solutions. Preliminary observations of all the thermal-analysis data indicate that the system shows a slight deviation from ideal behavior.

TABLE XI. Temperatures of Thermal Halts^a
in UF_6 - PuF_6 Mixtures

Mole Fraction UF_6	Solidus Point ^b (°C)	Liquidus Point ^b (°C)
0.0506	51.6 ± 0.1	53.0 ± 0.1
0.323	53.8 ± 0.1	54.7 ± 0.2
0.501	56.1 ± 0.2	57.8 ± 0.1
0.503	55.8 ± 0.4	57.5 ± 0.4
0.581	57.1 ± 0.6	59.4 ± 0.9
0.684	58.3 ± 0.2	59.6 ± 0.1
0.851	61.3 ± 0.0	62.4 ± 0.3
0.924	61.9 ± 0.1	63.6 ± 0.1

^aPoints determined by heating curves.

^bEach point is an average of two to five determinations.
The uncertainty value is the standard deviation of the average.

In connection with these studies, the triple point of pure PuF_6 was measured. The observed triple point of a 1.8-g sample of PuF_6 was $51.4 \pm 0.1^\circ\text{C}$, which compares favorably with the published value¹⁴ of 51.59°C obtained by extrapolation of vapor-pressure data.

¹⁴Weinstock, B., Weaver, E. E., and Malm, J., J. Inorg. Nucl. Chem. 11, 104-114 (1959).

b. Engineering Work

(i) Engineering-scale Alpha Facility. Process-development studies with plutonium-containing materials are being conducted in the engineering-scale alpha facility. Preliminary results from the first fluorination experiment with plutonium fuel material (see Progress Report for November 1965, ANL-7122, pp. 63-65) showed that after fluorination with fluorine the concentrations of uranium and plutonium in a grab sample of the alumina bed taken while the bed was fluidized were 0.015 and 0.025 w/o, respectively. These values correspond to uranium and plutonium removals of 99.9 and 95% of the material charged to the reactor. When the alumina bed was removed from the reactor, it was observed that a finely divided solid material containing plutonium had been held up in the reactor above the fluid bed. The plutonium content of this material was equivalent to about 15% of the plutonium in the $\text{UO}_2\text{-PuO}_2$ charge. Several methods are being investigated to reduce this holdup of fines and to return the fines to the bed during fluorination.

(ii) Process Development Studies for Uranium Dioxide Fuels. A development program is underway using a $1\frac{1}{2}$ -in.-dia fluid-bed reactor to establish optimum processing conditions for the fluorination of UO_2 fuel with BrF_5 . In this process (see Progress Report for August 1965, ANL-7090, pp. 47-48), the pellet UO_2 fuel is pulverized by reaction with oxygen and the U_3O_8 powder is fluorinated with BrF_5 to form volatile UF_6 .

A series of four tests was performed to determine the effect of fluorination temperature on the extent of conversion of uranium to UF_6 by BrF_5 . In each test, 270 g of UO_2 pellets were added to 470 g of alumina and oxidized to U_3O_8 powder by reaction with 30 v/o oxygen in nitrogen for 3 hr at 400°C . The powdered U_3O_8 was fluorinated to UF_6 by reaction with 8 to 14 v/o BrF_5 in nitrogen for 2.5 hr at temperatures of 380, 290, 200, and 110°C . The uranium concentrations in the final alumina beds were 0.0027, 0.0048, 0.165, and 10 w/o, respectively. These data indicate that a fluorination temperature of $>200^\circ\text{C}$ is required to assure essentially complete conversion of U_3O_8 to UF_6 at the BrF_5 concentrations used.

In future work in this program, the effect of BrF_5 concentration on the rate of fluorination of U_3O_8 will be studied.

G. Sodium Coolant Chemistry

1. Nature of Carbon in Sodium

In order to characterize the carbonaceous impurities in relatively pure sodium (<150 ppm carbon content), various direct and indirect methods of analysis are being employed. Frequently, the experiments attempt to

uncover the presence of carbon-containing compounds, either specific or typical of a class, which have been reported to exist in sodium. The results of recent experiments with United States Industrial Co. (U.S.I.) reactor-grade sodium (containing ~40 ppm total carbon) are given below.

Previous mass-spectrometric studies¹⁵ indicated that stable organic compounds may exist in sodium. Thus, extraction of such compounds by a hydrocarbon solvent might be feasible. This concept was tested by repetitive extraction of U.S.I. sodium with high-purity n-nonane under a helium atmosphere at 125°C in a Pyrex glass vessel. The n-nonane extracts were filtered and examined spectrophotometrically using n-nonane in the reference cell. No absorption was found between 2200 and 4000 Å. The sensitivity of the method of detection was estimated by using xylene as a stand-in for aromatic compounds which absorb strongly in the ultraviolet region. The results showed that the sodium contained less than 5 ppm of carbon in the form of extractable organic compounds that absorb in the ultraviolet.

The possibility exists that there are organic compounds in sodium which are not extracted by hydrocarbon solvents and which are stable even after hydrolysis of sodium. An example would be the sodium salt of hexahydroxybenzene, $\text{Na}_6(\text{CO})_6$, which is known to be a product of the reaction between sodium and carbon monoxide. Accordingly, U.S.I. sodium was dissolved in distilled water and the clear solution was examined spectrophotometrically with dilute sodium hydroxide in the reference cell. No significant absorption was found in the ultraviolet and visible regions (2150 to 7000 Å), indicating that the sodium did not contain any significant amount of stable, sodium hydroxide-soluble, aromatic compounds. The sensitivity of detection was such that sodium hydroxide solutions containing hexahydroxybenzene equivalent to ~1 ppm of carbon in sodium would give discernible absorption peaks in the ultraviolet.

Indirect information concerning the nature of carbonaceous species in sodium can be obtained by analysis of the products of sodium hydrolysis. The gases evolved on dissolution of U.S.I. sodium in water were absorbed on a liquid-nitrogen-cooled, silica gel trap and analyzed by gas chromatography. Methane (0.7 to 1.6 ppm equivalent carbon) was detected as an off-gas from the alkaline solution. The methane may have existed as such or as an organometallic compound in the sodium. When the alkaline solution was acidified, carbon dioxide (~6 ppm equivalent carbon) was evolved. The carbon dioxide probably originated from Na_2CO_3 in the sodium. Acetylene, ethylene, and ethane were not detected as off-gases from either alkaline or acid solution; carbon monoxide will be determined in future experiments. A grey-white residue, partially soluble in dilute HCl, remained after the dissolution. The residue, after being collected on a fine-porosity quartz frit, was analyzed for total

¹⁵Reactor Development Program Progress Reports: (a) ANL-7046, May 1965, p. 62; (b) ANL-7045, April 1965, p. 43; (c) ANL-7028, March 1965, p. 65.

carbon and found to contain 14 ppm carbon based on the original sodium. Summarizing, approximately 55% of the carbon in U.S.I. sodium has been accounted for by three different hydrolysis products.

Analyses for cyanide in the NaOH solution obtained by dissolution of U.S.I. sodium in water revealed less than 1 ppm cyanide. Using the same analytical procedure, less than 1 ppm cyanide was found in samples of sodium from the primary and secondary sodium coolant systems of the EBR-II.

H. Plutonium Recycle Program (EBWR)

1. Theoretical Calculations

New theoretical calculations have been made by means of the THERMOS code to generate the thermal group cross sections. The group cross sections at higher energy were left unchanged. Through use of cylindrical, one-dimensional, 4-group diffusion-theory calculations, the initial clean critical mass for the plutonium zone was calculated to be 20.1 elements as compared with the experimental value of 22 elements. The earlier non-THERMOS calculations predicted 17.7 elements. Also, the boric acid necessary for criticality in the 36-element evading was calculated to be 5.14 g/gal compared with the non-THERMOS calculation of about 7 g/gal. The corresponding experimental value is 5.09 g/gal.

Further calculations using two-dimensions are in progress to check the physical geometry more closely.

2. Worth Measurements

Measurements have been made of the worth of the central control rod and of boric acid for the 36-element loading of plutonium-bearing fuel. The procedure was to obtain the differential worth of the central rod from period measurements as the boric acid concentration was reduced from approximately 5 g/gal. It was also found from this procedure that the minimum boric acid concentration for which the eight-rod shutdown criterion is satisfied is 1.32 g/gal.

The partial differential worths of the central control rod were obtained with the other eight rods banked at three different heights. These differential worths are given in Figs. 16, 17, and 18. The error bars in these figures represent the uncertainty in the differential worth arising from uncertainties in the period (approximately 10%) and in the difference in the height of the rod (0.025 in.) when the reactor was critical and when it was on a period due to drift in critical rod position.

Fig. 16
Differential Worth of Central Rod for Eight-rod Bank Height of 48 in. Boric Acid Concentration 4.82-4.00 g/gal

Fig. 17. Differential Worth of Central Rod for Eight-rod Bank Height of 28.35 in. Boric Acid Concentration 4.00-2.78 g/gal

Fig. 18. Differential Worth of Central Rod for Eight-rod Bank Height of 19.62 in. Boric Acid Concentration 2.38-1.33 g/gal

tive agreement with the calculations which predicts about -1.3% in Δk for a 1-g/gal addition to the unpoisoned system and about -1% for a 1-g/gal addition to the system which contains 9 g/gal.

For certain control rod configurations an inverted rod-worth effect was noted. When the central rod was partially inserted and the eight-rod bank was fully withdrawn to 48 in., insertion of the bank caused a slight increase in reactivity. These effects arise because the lower 13 in. of the control rod follower is stainless steel, a moderately strong neutron absorber, instead of Zircaloy. In addition, the rod cross section in the stainless steel region is smaller than in the Zircaloy

The reactivity worth of a change in boric acid concentration was obtained from the change in the critical height of the central control rod and the worth of this change in height. Because the curves for differential rod worth are nearly linear, the reactivity worth of a change in rod position is well approximated by the product of the differential worth of the rod at its average position and the distance it was moved. The boric acid worths, $\Delta\rho/\Delta c$, are tabulated in Table XII. The uncertainties in the boric acid worths reflect unavoidable uncertainties in the differential rod worth and in the boric acid concentration itself. Due to limitations on the precision of titration, the uncertainty in the change in boric acid concentration is at least 0.04 g/gal. In view of the large uncertainty we can only state that the boric acid worth between 4.8 and 1.3 g/gal appears to be approximately -0.012 $\Delta k/\text{g/gal}$. This boric acid worth is in qualita-

TABLE XII. Boric Acid Worth as a Function of Boric Acid Concentration at Ambient Temperature

Boric Acid Conc (g/gal)	Boric Acid Worth $\Delta\rho/\Delta c$ [$(-\Delta k/\text{g/gal}) \times 10^2$]
4.82-4.42	1.19 $\pm$ 0.14
4.42-4.24	1.17 $\pm$ 0.20
4.28-3.99	1.25 $\pm$ 0.19
4.00-3.77	1.29 $\pm$ 0.19
3.77-3.515	1.42 $\pm$ 0.22
3.515-3.12	1.32 $\pm$ 0.18
3.12-2.78	1.25 $\pm$ 0.19
2.78-1.98	1.14 $\pm$ 0.17
1.98-1.54	1.33 $\pm$ 0.15
1.54-1.33	1.39 $\pm$ 0.25

follower section, so that more boric acid in the water is present at this region of the rod. With the eight-rod bank or the central rod partially inserted, the flux is pinched down to the lower part of the system causing the stainless steel portion of the followers of the fully withdrawn rods to be in a region of greater importance. When the rods are inserted, initially, the positive reactivity effect of removing stainless steel and boric acid from a region of high importance is greater than the negative reactivity effect of inserting the control rods into a region of small importance. The rod positions where the inverted worth effect occurs have been examined experimentally, and the mechanical stops on the rods have to be reset to prevent the raising of the rods to such a height that their insertion would cause an increase in reactivity.

3. Temperature Coefficient of Reactivity

During the course of the experiment for the determination of boric acid worths, indications of a positive temperature coefficient of reactivity were found. This effect was investigated by raising the temperature of the water in the reactor vessel to 142°F by use of the startup heater; as the temperature of the water was lowered, the critical height of the central rod with the other rods banked at 48 in. was observed along with the differential worth of the central rod.

Table XIII summarizes the results of this experiment. The uncertainty given for the critical height of the central rod arises primarily from the uncertainty in the boric acid concentration of about 0.03 g/gal. The uncertainty in the temperature due to incomplete mixing was about 0.4°F. From Table XIII we see that because the central rod is withdrawn to maintain criticality as the temperature is lowered the temperature coefficient of reactivity is positive over the temperature range studied. However, the coefficient is quite small, and because of uncertainties in the boric acid concentration it cannot be determined precisely. From the change in the critical height of the central rod and the differential worth of the central rod, the average temperature coefficient between 87 and 126°F is about 5×10^{-5} k/°F. It is planned to continue this experiment to higher temperatures to determine where the temperature coefficient becomes negative for several boric acid concentrations.

TABLE XIII. Critical Height of the Central Rod as a Function of Water Temperature

Temperature (°F)	Boric Acid Concentration (g/gal)	Critical Height of Central Rod (in.)	Temperature (°F)	Boric Acid Concentration (g/gal)	Critical Height of Central Rod (in.)
87.7	5.19	41.65 ± 0.85	125.8	5.13	36.15 ± 0.34
94.6	5.18	40.60 ± 0.69	125.6	4.96	33.31 ± 0.24
102.7	5.18	39.48 ± 0.54	134.9	4.94	32.55 ± 0.22
109.9	5.17	37.66 ± 0.39	142.6	5.19	32.08 ± 0.21
117.8	5.20	36.94 ± 0.35			

III. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. General

The architect-engineer has continued the preparation of architectural layouts of various buildings of the complex, and is also initiating building-elevation drawings. All area requirements have been completed. Also, economic and engineering analyses of structural framing and architectural treatment of the buildings have been initiated.

The subsurface boring and testing work has proceeded satisfactorily, and a meeting was held on site with the soil consultant, the architect-engineer, and the Laboratory to review progress and scope. The on site drilling work is approximately 70% completed.

Several extensive meetings were held with the architect-engineer to review process systems, electrical, instrumentation and nuclear work accomplished.

2. Reactor Physics Experiments

Sufficient fuel foils were not available to permit reloading to a 1215-fuel-foil system. However, all of the remaining foils to be sent have been inspected and approved for shipment. Therefore, no further delay is anticipated in delivery of fuel foils from the fabricator.

Experiments have been continued with the present core (810 fuel foils and 810 boron-stainless steel poison strips). Three sets of experiments have provided significant data regarding (1) reactivity effects of voidings in the Internal Thermal Column (ITC); (2) vertical activation traverses at several radial positions in the beryllium reflector, as a function of the total height of the beryllium stack; and (3) reactivity effects of inserts (beryllium and Lucite) in beam-tube simulations, and neutron-flux distributions in these tubes. Figures 19 and 20 show the reactivity effects of inserts as a function of their location.

Preliminary data for ITC voidings were previously reported (see Progress Report for November 1965, ANL-7122, pp. 73-74). Starting with approximately 74% void in the ITC, in the form of 19 voided, hollow Lucite tubes, various sequences of selective flooding of individual tubes were investigated. Figure 21 shows the radial dimensions and locations of the Lucite tubes in the ITC; identifying numbers are keyed to the results shown in Table XIV and in Fig. 22. Each sequence is normalized to zero reactivity effect with all 19 tubes voided. Note that, in each sequence, flooding of the first few tubes results in a total net reactivity gain. As

more tubes are flooded, the total reactivity effect changes sign. This is shown also in Fig. 22, a graphical depiction of the results in Table XIV. In addition to the actual void volume of the interiors of the Lucite tubes, the tube walls provided a 1.7% reduction in hydrogen content of the ITC. Inclusion of this additional void therefore would shift the reference zero by 0.06% in reactivity and would increase the maximum percentage of ITC void by approximately 1.7%.

Fig. 19. Reactivity Effect of 12-in. Long Beryllium Insert in Through Tube

Fig. 20. Reactivity Effect of 4-in. Long Lucite Insert in Through Tube

Fig. 21. Locations and Numbering of Lucite Tubes for Voidings of the ITC

TABLE XIV. Reactivity Effects of Flooding Void Tubes in the ITC

Reference Curve in Fig. 20	Sequence of Floodings	Tube No. ^a	Differential Change in Reactivity (%)	Total Net Change in Reactivity (%)	Average Differential Change in Reactivity per Tube (%)
A	1	8 (corner)	0.064	0.064	0.064
	2	17 "	0.049	0.113	0.049
	3	19 "	0.043	0.146	0.043
	4	12 "	0.044	0.190	0.044
	5	3 "	0.038	0.228	0.038
	6	1 "	0.034	0.262	0.034
	7	10 (center)	-0.008	0.254	-0.008
	8	16 (edge)	-0.030	0.224	-0.030
	9	Remaining 11	-1.431	-1.207	-0.130
B	1	10 (center)	0.062	0.062	0.062
	2	15 "	0.038	0.100	0.038
	3	11 "	0	0.100	0
	4	6 (inside ring)	-0.004	0.096	-0.004
	5	5 "	-0.060	0.036	-0.060
	6	9 "	-0.040	-0.004	-0.040
	7	14 "	-0.086	-0.090	-0.086
	8	8 (corner)	+0.008	-0.082	+0.008
	9	16 (edge)	-0.056	-0.138	-0.056
	10	13,17	-0.149	-0.287	-0.075
	11	18	-0.103	-0.390	-0.103
	12	19	-0.136	-0.526	-0.136
	13	4	-0.140	-0.666	-0.140
C	1	8 (corner)	0.067	0.067	0.067
	2	16 (edge)	0.055	0.122	0.055
	3	13,17,18,19	0.046	0.168	0.012
	4	12,7,3,2,1,4	-0.247	-0.079	-0.041
	5	10 (center)	-0.160	-0.239	-0.160
	6	Remaining 6	-0.962	-1.201	-0.160
D	1	8 (corner)	0.070	0.070	0.070
	2	10,6	0.080	0.150	0.040
	3	16 (edge)	0.022	0.172	0.022
	4	12	0.002	0.174	0.002
	5	7	-0.021	0.153	-0.021
	6	5,9,14,15,11	-0.410	-0.257	-0.082
	7	Remaining 8	-0.891	-1.148 ^b	-0.011
E	1	5,9,14	0.126	0.126	0.042
	2	6,11,15	-0.065	0.061	-0.022
	3	17,13,8,4,1,2	-0.387	-0.326	-0.065

^aThe tube numbers refer to tube locations shown in Fig. 19.^bPreliminary.

Fig. 22. Reactivity Change upon Flooding Tubes in ITC, Starting with a 74% Void

The maximum reactivity gain of partial voidings of the ITC, within the vertical bounds of the active fuel zone, is estimated to be 1.5% at approximately 50% void (see Fig. 22). For other sequences of voidings, the maximum reactivity gains due to voidings are smaller than 1.5%, and the peaks occur at higher percentages of void volume. Although the 19-tube array afforded a reasonably comprehensive survey of the reactivity-versus-void characteristics, experiments with smaller tubes might yield somewhat different results.

The isothermal reactor temperature coefficient of reactivity was measured to be $-0.007\%/^{\circ}\text{C}$ with all 19 Lucite tubes voided. This is $3\frac{1}{2}$ times the magnitude reported for the case where there was no void in the ITC (see Progress Report for June 1965, ANL-7071, pp. 52-54). A reevaluation of this experiment reveals the possibility that the Lucite tubes expanded with increasing temperature, thereby introducing an additional negative reactivity effect. The experiment was an attempt to isolate the temperature coefficient of reactivity of coolant in the fuel zone. If such a measurement is attempted later in the experimental program, it might be advisable to select a void configuration in the ITC where small additional voids in the ITC would have a negligible effect on reactivity.

A total of 8 "blind" beam tubes and 2 horizontal through tubes are to be included in the AARR. The vertical locations and the azimuthal orientations of these tubes have been established. Two of the blind beam tubes are located with their longitudinal axes in a plane $3\frac{1}{2}$ in. above the top of the active fuel zone. Consequently, the height of the beryllium reflector will be increased so as to enhance the thermal-neutron flux in these two tubes. Information needed to guide the selection of the beryllium height has been supplied by experiments in the Criticality Facility. For each of several different beryllium heights, vertical activation traverses have been obtained with bare manganese foils. Figure 23 displays the geometric configuration for these vertical traverses. Traverses shown in Fig. 24 were obtained with the original 19-in. height of beryllium reflector. Figure 25 shows vertical traverses with an additional height of 12 in. of beryllium, and Fig. 26 for a total beryllium height of 35 in.

Note:

1. Material in Channel of Control Blade No. 1:
 Figures 4, 5, & 6, Aluminum Dummy.
 Figure 7, Regular AARR Control Blade; Tip of the
 Hafnium Located at the Top of the Active
 Portion of the Fuel Zone.
2. Dashed Curves are Cadmium-Covered Points.

Fig. 23. Foil Locations of Vertical Activation Measurements in the Beryllium Reflector Region (810-fuel-foil System)

Fig. 24. Vertical Activation Traverses with Manganese Foils in the Beryllium Reflector (810-fuel-foil System; Beryllium Height, 19 in.)

Fig. 25. Vertical Activation Traverses with Manganese Foils in the Beryllium Reflector (810-fuel-foil System; Beryllium Height, 31 in.)

Fig. 26. Vertical Activation Traverses with Manganese Foils in the Beryllium Reflector (810-fuel-foil System; Beryllium Height, 35 in.)

In the experiments summarized in Figs. 24-26, an aluminum dummy blade was placed in the control channel of the adjacent peripheral control blade. In Fig. 27, the results of vertical traverses are shown when a hafnium blade was positioned at the top of the active fuel zone. Comparison of Figs. 25 and 27 reveals that the control blade does affect (reduce) the neutron flux where the two top-most blind beam tubes are to be located. The tips of these beam tubes will be close to radial position A (see Fig. 23).

Fig. 27. Vertical Activation Traverses with Manganese Foils in the Beryllium Reflector and Hafnium Blade at Top of Fuel Zone (810-fuel-foil System; Beryllium Height, 31 in.)

Further measurements are planned for the case where the adjacent peripheral control blade will penetrate the active fuel zone by $1\frac{1}{2}$ in. The flux-depression effect of operating with the peripheral (safety) control blades in a position of significantly large differential reactivity worth will be measured.

3. Theoretical Reactor Physics

Analysis of the reactor physics characteristics of the preliminary, physics-reference design Mark-I is continuing.

At an operating power level of 100 MWt, equilibrium xenon (Xe^{135}) has been calculated to control 2.1% in reactivity at the beginning of the core cycle. Under these same conditions, samarium (Sm^{149}) would control 3/4% in reactivity. At 240 MWt, equilibrium xenon and samarium are calculated to control $\sim 3\frac{1}{2}\%$ in reactivity.

A computation has been made of the mass of boron required to control the full estimated available excess reactivity ($\sim 16\%$) of the fresh reactor. From this, the average differential reactivity worth of natural boron has been computed to be $-0.024\%/g$ B. Thus, to control 6% in reactivity, a total of ~ 250 g B would be needed, or approximately 1 g B per 240 g U^{235} .

The effective delayed-neutron fraction was computed to be $\beta_{\text{eff}} = 0.00720$.

In Fig. 28 is shown the computed gross radial distribution of power in the fresh reactor, normalized to unity at a radial position of 10.0 cm. The reactor was brought to criticality by a uniform distribution of boron

Fig. 28. Radial Distribution of Fission-Neutron Production Rate (Normalized to Unity at 10.0 cm)

in the core. In Figs. 29-31 are presented some of the radial distributions of neutron flux obtained by multigroup, one-dimensional calculations, re-normalized to unit flux in group No. 16 (0-0.1 eV) at the vertical axis of the ITC.

Fig. 29. Semilog Plot of Radial Distributions of Neutron Flux in the Preliminary Reference Design of AARR (Normalized to Unit Group-16 Flux at Center of ITC)

Fig. 30. Cartesian Plot of Radial Distributions of Neutron Flux in the Preliminary Reference Design of AARR (Normalized to Unit Group-16 Flux at Center of ITC)

Fig. 31. Radial Distributions of Neutron Flux for Groups 15 and 16 in the Preliminary Reference Design of AARR (Normalized to Unit Group-16 Flux at Center of ITC)

Note that in Fig. 28 the peak power in the gross radial distribution is only ~ 1.8 times the average power. This was set deliberately, by selecting the gradation in U^{235} content. This extra margin of $\sim 10\%$ below a gross maximum-to-average ratio of 2 accounts for the actual distribution of the U^{235} in the cermet, as a correction to the approximation in which the fuel and all of the metal and water in the region are homogeneously distributed.

In Table XV is shown the graded U^{235} loading corresponding to the results of Figs. 28-31. In Table XVI is summarized the group-by-group fractional distribution of the production rate of fission spectrum neutrons in the bulk of the reactor core. The calculated median neutron energy for such events is in group No. 11, which covers the energy range from 10 to 30 eV. The number of fission-spectrum neutrons, ν_j , produced per fission by a neutron in energy group j is somewhat larger at higher neutron energy. Therefore, this median neutron energy is higher than the median energy for fission.

TABLE XV. Graded Loading of U^{235} in the Core of the Preliminary Reference Design

Number of Fuel-zone Annulus	Radial Dimensions of Annulus (cm)	Fractional Loading ^a of U^{235}
1	6.83-7.02	0.22
2	7.02-7.20	0.28
3	7.20-7.38	0.37
4	7.38-7.56	0.47
5	7.56-7.73	0.60
6	7.73-7.90	0.75
7	7.90-8.06	0.91
8	8.06-23.67	1.00
9	23.67-23.87	0.78
10	23.87-24.06	0.57
11	24.06-24.26	0.42

^aThe fractional loading is the ratio of the U^{235} content in this zone to the U^{235} content in the bulk of the reactor where the fuel-plate core contains 37 w/o highly enriched UO_2 .

TABLE XVI. Fractional Contribution of Each Neutron-energy Group to Production Rate of Fission-spectrum Neutrons^a

Number of Neutron-energy Group	Minimum Neutron Energy in Group	Fractional Contribution ^{b,c}
1	3 MeV	0.014
2	1.4 MeV	0.027
3	0.9 MeV	0.015
4	0.4 MeV	0.022
5	0.1 MeV	0.026
6	17 keV	0.032
7	3 keV	0.040
8	0.55 keV	0.065
9	100 eV	0.128
10	30 eV	0.099
11	10 eV	0.102
12	3 eV	0.078
13	1 eV	0.049
14	0.4 eV	0.066
15	0.1 eV	0.138
16	0 eV	0.099

^aThis fractional contribution is calculated in the bulk of the reactor core, where the U²³⁵ loading is highest, at a radial position of ~15 cm.

^bThe fractional contribution is computed as $(\nu\Sigma_f)_j \phi_j / \sum_k (\nu\Sigma_f)_k \phi_k$,

where Σ_{fj} is the macroscopic fission cross section in group No. j, and ϕ_j is the group-j neutron flux as printed out following the REX multigroup computation.

^cThe sum of the fractional contributions is slightly larger than unity because of round-off. Although these fractions are listed to three decimal places as computed, it must be understood that the spectrum of fission events is sensitive to the particular model employed in computing group cross sections.

4. Heat Transfer

a. Analytical Studies. A study has been initiated to investigate the effect on heat transfer of attaching 0.040 x 0.040-in. spacers of varying lengths in the fuel assemblies. These effects will be reflected in the core thermal performance. The spacers, desirable for holding channel dimensions to tolerance during manufacture and operation, are assumed bonded

to the heat transfer surface of a fuel plate such that the spacer and channel axes are parallel. The study, made on the CDC-3600, employed the General Electric THTB Code.¹⁶ Three geometric representations are under investigation: a fuel plate with central spacer, a fuel plate with edge spacer, and a one-quarter section of an AARR heat transfer test section. Two-dimensional calculations of temperatures within the three shapes have been made for specific boundary conditions.

A study is continuing to permit selection of an appropriate heat transfer coefficient at the metal-fluid interface near the spacer. Proper selection of the heat transfer coefficient will permit calculation of the upper and lower bounds of the internal temperature in the fuel plate.

The inclusion of the AARR heat transfer test section into the calculations will permit a comparison of computed results with those obtained by experiment. Such a comparison may lead to better selection of heat transfer boundary conditions, thereby affording more accurate appraisal of the effect of a spacer attached to a fuel plate.

b. Experimental. The steady-state heat transfer test program will evaluate the maximum steady-state power level in a test section simulating AARR Mark-I design. Three new test sections have been designed and are nearing completion of fabrication. These sections contain the following innovations:

(i) The section is formed by flattening a thin-walled tube over a mandrel. Previous sections were made by milling a channel out of a blank and then welding on a cover plate. The new method is less expensive in fabrication, holds a closer tolerance on channel spacing, and eliminates troublesome welding.

(ii) The edges are sufficiently rounded to drop the corner heat flux below the circumferential average.

(iii) Sheathed, ungrounded, 0.020-in. thermocouples with an accurately located junction will be installed in the flow path downstream from the heated section.

(iv) Small grooves have been milled in the backup assembly to allow a relatively metal-free path for the gamma rays used to measure steam volume fraction at the exit. Calibration blocks for void measurement by use of γ rays have been built.

(v) Taps for additional pressure measurements have been installed.

(vi) The three sections have different channel spacings: 0.030, 0.040, and 0.050 in.

¹⁶Applied Mathematics Division Program RE 322 (Modification of the General Electric THTB Code for the CDC-3600 Computer).

Response speed of data acquisition has been improved by changing over to an optical recording system. This system offers greater chart width and an increase in the number of data channels which can be handled, and will also be used for the transient heat transfer test program. Its use will provide a smoother transition from steady-state to transient testing.

5. Hydraulic Tests of Fuel Assemblies

Hydraulic testing of the AARR fuel assemblies requires a device to measure fuel-plate distortion or vibration, and pitot-static tubes to measure the flow distribution within the assembly.

The investigation will be conducted with a simulated fuel assembly installed in a test-section holder designed to permit variation of the dimension of the flow channel between an outer plate and the wall of the holder. The frequency and amplitude of any movement of this plate will be measured by a variable-impedance transducer. Pitot tubes will be used in each flow channel to determine flow distribution and to indicate impending plate collapse.

A variable impedance transducer for low-temperature application and an oscillator-demodulator have been adjusted, and the amplitude calibration was verified in a fixture designed and constructed for that purpose. Both the fixture and equipment combination perform satisfactorily.

A previously constructed apparatus was adapted for use in calibrating the pitot tubes. This apparatus was used in conjunction with an existing hydraulic loop to provide water speeds of ~ 60 ft/sec in a rectangular channel measuring from 0.050 to 0.035 in. x 2.75 in. in cross section. The channel is 5 in. in length. Total flow measurement is made by means of a turbine flowmeter, and the pitot tube output is measured with the aid of differential pressure transducers.

The channel is formed by two flat plates that deflect slightly under pressure; an effect accounted for in system calibration. The loop is used to study the effect of variations such as lateral placement in the channel, yaw, rotation, pitot-tube design, and water temperature. The results of preliminary tests were utilized in improving technique and establishing the requirements of a channel gauge and measuring probe. Data have been obtained for straight pitot tubes with two and with five static holes. The test data do not indicate a significant advantage for either design. The data justify procurement of larger quantities of tubes.

The final design uses two static holes. The periphery of the outer tube is oval, with the major axis serving also as the centerline of the static holes. The placement of the tubes will be such that the plane of the

major axis is parallel to the plane of the major dimension of the flow-channel cross section, i.e., parallel to the heat transfer surfaces.

6. Core Package Design

A reference design for the Mark-I Core Package has been developed. This package includes the fuel-element assemblies, control and safety rods, core-support grid, upper holddown grid, and inner and outer shrouds. The design has progressed sufficiently to determine the feasibility of incorporating several special functions within the package. As it is planned to preassemble and test each core prior to installation in the reactor vessel, the core package has been designed to provide positive locks on each control and safety rod. These locks can be actuated remotely after installation to retain the rods within the core during all handling operations.

An absorber section surrounding the ITC has been designed as a part of the core package. This absorber can be inserted and removed remotely, and will provide additional reactivity holddown available after shutdown and prior to disassembly and removal of the experimenters' hardware in the ITC.

The core-support grid, the main structural element of the core package, supports the inner beryllium reflector, as well as all other elements, of the core package. Elements of the reflector are removed during core change in order to provide clearance around the core package and access to the bolts holding the core-support grid to its mounting plate.

7. Control Rod Drive Mechanism

A simulated drive system has been attached to the magnetic clutch. The mechanism, which consists of an extension shaft, miter gears, pinion, rack, and a 70-lb dead-weight, represents system inertia. The measured delay (the time between power interruption and rack movement) was 12 msec. It was found that the addition of a 0.08- μ fd capacitor, connected in parallel with the clutch coil, reduced the delay to 2 msec, but raised the attendant discharge voltage to 3200 V. Experimentation with a parallel circuit of a 10,000-ohm resistor in series with a 0.47- μ fd capacitor resulted in a delay time of 4 msec while producing a discharge voltage of only 900 V. The investigation is continuing.

8. Development of Experimental Facilities

Tests to determine the effect of heat flux on the corrosion of 6061 aluminum (see Progress Report for November 1965, ANL-7122, pp. 76-77) have continued for the purpose of evaluating the aluminum beam tube thimbles under reactor conditions of heat generation and jacket-cooling water temperature and quality.

An aluminum specimen that had a heat flux of about 800,000 Btu/hr-ft² across its surface for 2000 hr has been examined metallographically. Metallographic examination of the surface verified the previous observation that many small areas of the corrosion product had spalled from the surface before the run was terminated, leaving an irregular oxide surface. A series of measurements across the center of the specimen 1 in. from the inlet end showed an average oxide thickness of 1.65 mils, with individual thicknesses varying from 0.2 to 3.25 mils. Similar measurements at the middle of the specimen gave an average thickness of 2.19 mils, with local thickness variations of 0.65 to 2.95 mils. One inch from the outlet end (hottest region), the average thickness was 2.50 mils, with thicknesses ranging from 1.75 to 3.65 mils. In all cases, the metal was attacked uniformly; there was no evidence of localized attack. The oxide was removed from one axial half of the specimen, and the metal loss at several locations was determined as the difference between the initial metal thickness and the thickness after the test. Metal losses increased uniformly from about 2 mils at the cooler inlet end to 4 mils at the outlet end.

A second heat flux specimen has now been in test for 807 hr at a heat flux of 400,000 Btu/hr-ft² under the following conditions:

Distance from Inlet End, (in.)	Temperature at Water Interface (°F)	Bulk Water Temperature (°F)	Fluid Film Coefficient (Btu/hr-ft ² -°F)
1.00	190	125	6150
1.25	190	126	6250
2.25	190	132	6900
3.25	193	137	7140
3.25	196	137	6780
4.25	197	143	7410
5.25	202	148	7410
5.50	201	149	7690

The maximum observed temperature increase, which reflects the formation of a layer of corrosion products, has been 9°F at a distance of 5.50 in. from the inlet. If 1.3 Btu/hr-ft-°F be the thermal conductivity of the corrosion product, this ΔT corresponds to a corrosion product thickness of 0.35 mil. This test will be continued for a total time of 2000 hr.

The isothermal test to determine the effect of exposed aluminum surface area on the corrosion of beryllium is still in progress.

The conceptual design drawings of two types of gas-actuated rabbit facilities have been completed. These facilities provide for samples

traveling nonstop through the core, for very short irradiations, and for samples to be stopped in the high-flux zone where cooling is required. Both concepts have been integrated with the present design of the internal thermal column package.

9. Nuclear Instrumentation

The arrangement of nuclear instrumentation has been modified and coordinated with the design requirements of the beam facility. Three criteria were used as the basis for the modified design: (1) to reduce the number of decades (range) over which the instruments must respond, (2) to remove the necessity for making penetrations through the reactor vessel for instrumentation, and (3) to increase the thermal-neutron flux "seen" by nuclear instruments, in order to assure fast and accurate response in high-flux and period measurements. The need for the latter has been indicated by the results of transient and heat transfer studies.

In order to achieve these ends, it was necessary to overcome two fundamental difficulties::

a. The core, although operating at great power, is relatively small, making physical distances to the outside of the reactor vessel relatively large. Neutron attenuation is, therefore, correspondingly large. (Competition for space in the vicinity of the core is intensified by this factor.)

b. The range of the instrumentation is fundamentally dependent on the ratio of the maximum power to the minimum, or source-level, power.

An aluminum window was selected as a means of allowing increased neutron transmission to the outside of the reactor vessel. Theoretical calculations (see Progress Report for October 1965, ANL-7115, pp. 64-70) and critical experiments (see Progress Report for September 1965, ANL-7105, pp. 59-60) indicate that the neutron intensity is raised sufficiently to permit external placement of all nuclear instrumentation provided that a source level of approximately $1/4$ W or greater is established, implying a required instrument range of 9 decades or less. The resulting design offers some freedom in the exact placement of neutron detectors and corresponding thimble size, and provides some isolation from local reactivity perturbations within the core and reflector.

10. PERT/Cost Experiment

The PERT/Cost experiment is continuing. Approximately 40% of the resource estimates for research and development activities have been

completed and are being incorporated into the computer program. The work-breakdown structure has been issued in preliminary form, but will not be ready for distribution until the PERT/Cost computer data for the research and development program have been validated.

B. Energy-conversion Systems.

1. Regenerative Emf Cells

a. Bimetallic Cells

(i) Engineering Material Study. Corrosion studies, both static and dynamic, have been carried out to determine the suitability of materials for construction of thermally regenerative emf cell systems. The results of previous dynamic tests were reported in the Progress Report for August 1965, ANL-7090, p. 67. The static tests reported here were conducted to determine the corrosion of various materials by liquid bismuth and tin at temperatures of about 700 to 1000°C.

For each static test, the material under study (usually in the form of a coupon, $1/2$ by 1 by $1/8$ in.), together with tin or bismuth, was contained in a crucible fabricated from molybdenum-30 w/o tungsten, an alloy which is considered to be nearly inert to attack by bismuth and tin. The loaded crucible was placed in a stainless steel vessel located in a resistance-heated furnace and was held at the test temperature for a predetermined period of time.

The results of all the tests conducted to date are presented in Table XVII. Common materials, including low carbon steel, pure iron, and stainless steels, show a fair degree of resistance in liquid bismuth at temperatures up to 800°C. These materials, however, were not tested in liquid tin because of their high solubility in tin. The refractory metals, except for niobium and its alloys, show promise as containment materials for both bismuth and tin. Alumina was tested for possible use as an electrical insulating material and appears to be a satisfactory material for this purpose.

TABLE XVII. Static Corrosion of Refractory Metals, Steels, and Alumina by Bismuth and Tin

Test Materials	Bismuth			Tin		
	Temp (°C)	Time (hr)	Results	Temp (°C)	Time (hr)	Results
Molybdenum	1000	97	No corrosion.	1000	26	Penetration, <15 μ depth.
Niobium	1000	64	Dissolution, 30 μ thickness. Intergranular, <50 μ .	1000	24	Slight dissolution. Intermetallic layer with dendrites, <20 μ thickness.
Tantalum	1000	95	No apparent attack.	1000	96	Penetration, <20 μ depth.
Tungsten	1000	24	No apparent attack.	1000	96	Penetration, <5 μ .
Mo-30 w/o W	1000	97	No corrosion.	1000	67	No corrosion.
	~1000	>1100	No visible attack. Intermetallic layer of ~100 μ thickness.	~1000	>850	No visible deterioration. Intermetallic layer of ~100 μ thickness.
Mo-50 w/o Re	1000	91	No apparent attack. Intermetallic layer of >12 μ thickness.	1000	63.5	No apparent attack.
Nb-1 w/o Zr	1000	167	Severe intergranular attack, <250 μ depth. Specimen became brittle.	1000	160	Intergranular attack, <100 μ depth.
Nb-10 w/o W-2.5 w/o Zr	1000	71.5	Slight dissolution and pittings.	1000	87.5	Slight dissolution and pittings.
Armco Iron	850	24	Slight attack.	-	-	Not tested.
	1000	50	Complete dissolution.	-	-	-
1040 Steel	1000	22	Intermetallic layer of <80 μ thickness. Cr preferentially segregated.	-	-	Not tested.
Type 304 SS	850	97	Intergranular, <100 μ depth. No attacks found in vapor phase.	-	-	Not tested.
Type 304 SS	700	97	Slight dissolution. Decarburization, <5 μ depth including vapor phase.	-	-	Not tested.
High-purity Alumina	600	99	No attack. No wetting.	600	77.5	No attack. No wetting.
	850	52	Partly wetted and grayed.	850	29	Slightly wetted.
	1000	72	Partly wetted and grayed, not electrically conducting.	1000	74	Completely wetted. Slightly conducting.

IV. NUCLEAR SAFETY

A. Reactor Kinetics

1. Fast Reactor Safety

a. Coolant (Water) Expulsion Studies. A new series of tests have been completed in which the initial temperature of the water was varied from 25 to 100°C. It was found that the initial pressure pulse increased with increasing temperature. These initial pressure pulses, which provide the initial water acceleration, are very small, in no case greater than about 2 atm (for energy inputs of about 3 kW-secs). However, the water-hammer pressure (caused by the two-phase, water-steam mixture falling back into the tube) has exceeded 10 atm.

b. Superheat Experiments. It is desirable to know the degree of liquid superheat required to initiate nucleate boiling in sodium under conditions simulating a reactor environment. Construction of a test apparatus and containment vessel is underway, and the design of a gas-purification system is complete.

Two high-temperature, fast-response piezoelectric pressure transducers to measure pressure transients during sudden void formation have been ordered. Also, an oscilloscope and associated equipment have been ordered to measure accurately the high-frequency signals which the crystal transducer is capable of producing.

The existing X-ray generator system has been set up to determine the feasibility of using this existing X-ray equipment to measure transient void formation in sodium by means of X-ray attenuation. Initial tests simulating a sodium system with water are underway.

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

The design and construction of experiments to study critical flow of different fluids are continuing. Final design of the sodium loop and the water loop to be used in the low-pressure region (0-15 psia) have been completed.

The experimental facility for studying the fluid behavior at the point of choking has been completed. A circular test section (ID = 0.25 in., L/D = 150) has been installed, and about fifty runs have been made in the quality range from 0.1 to 2%, including measurements of flow, axial temperature, and pressure profiles. Some of the preliminary findings are:

(1) the obtained flowrates appear considerably higher than those predicted on the basis of previous investigations; (2) the critical flowrate is not found to exhibit a maximum as a function of quality as predicted by the Levy and Moody models; (3) the assumption of thermal equilibrium appears not to be valid.

A rectangular test section (1 x 1/3 in. and L/D = 150) has been designed and is being built.

2. Equation of State for Fast Reactor Accidents

Reactor fuels are of less than the theoretical density due to pores and, after some burnup, due to swelling produced by fission products. The effect of this porosity on the equation of state is thus of interest for fast reactor safety analyses. Any mechanism which reduces the core pressure, and therefore the disassembly rate, in the early stages of a hypothetical fast reactor accident tends to increase the total energy release of the accident. Porosity is such a mechanism.

a. Effect of Pressure on Elastic and Plastic Constants. The propagation of elastic-plastic waves in solids is an important shutdown mechanism for accidents involving the smaller reactivity insertions. The mechanical equation of state is represented by the dependence of the bulk and shear elastic moduli on temperature and pressure. Within the framework of finite strain theory, changes in the bulk modulus may be evaluated from experimental data on shock velocities by use of the Mie-Grüneisen equation of state.¹⁷

It can be shown that the change of the bulk modulus K with pressure P is given by

$$\frac{\partial K}{\partial P} = 4S - 1, \quad (1)$$

where S is the constant in the empirical relation (see Progress Report for September 1965, ANL-7105, p. 72) relating shock velocity u_s and particle velocity u_p :

$$u_s = C + Su_p \quad (2)$$

where C is the velocity of sound. The modulus of rigidity G is related to the bulk modulus by the equation

$$G = \frac{3}{2} \frac{1 - 2\nu}{1 + \nu} K, \quad (3)$$

¹⁷Jones, A. H., WL-TR-64-103 (Nov 1964).

where ν is the Poisson's ratio. Examination of experimental results has shown that the assumption of constant Poisson ratio is a good approximation. Extension of this theory to porous materials is under investigation.

The effect of density can be shown, to first order, using Eq. (2), with a constant S , and C proportional to the square of density. Calculations were made and compared against the experimental results of Skidmore and Morris¹⁸ on solid and porous uranium. Agreement was good. Estimates are being made for other materials.

It can be shown that for a given ratio of final density to initial density a shock wave will create a higher pressure in the porous material than in the solid. This follows from the laws of conservation of mass and momentum across the shock front, when combined with the experimental data on uranium shock velocity and particle velocity. However, this analysis shows that, on the basis of pressure versus internal energy the comparison is different. With a given specific internal energy input, in ergs/gram the pressure is higher in the solid material. Theoretical investigations on the behavior of porous material are continuing, in order to better identify the relationships between physical quantities and the significance to reactor accident studies.

3. Transient-pressure Response of Transducers and Mark-I Integral Loops

Evaluation of the transient response of pressure transducers and the associated equipment is necessary for analysis of transient-pressure data as well as for design of future loops and instrumentation. Several sequences of transient tests have been performed to survey this type of behavior. The tests have included shock-tube tests on a stagnant liquid container similar in size to the integral loop, performed on "bare" transducers, and on transducers in NaK-filled stand-off subassemblies (with "bare" transducers as monitors), and run both with and without water (whose physical properties are similar to those of sodium) inside the container. Tests were also run directly on a loop, at room temperature and filled with silicone fluid, using a quick-opening solenoid-operated gas valve, and with both bare transducers and transducers in NaK-filled stand-off subassemblies.

Tests in the container with both bare transducers and transducers in stand-off subassemblies showed the passage of the shock wave and reflections from walls. In the case of bare transducers, with d.c. excitation, the resonance of the transducer diaphragm (about 15,000 cps) was clearly discernible. An additional resonance, detected with both types of transducer mountings and of the order of several percent of full-scale transducer output, was found with a frequency of a few hundreds cps. On the basis of direct calculation of frequencies and the change of frequency with

¹⁸ Skidmore, I. C., and Morris, E., "Experimental Equation of State Data for Uranium and Its Interpretation in the Critical Region," Thermodynamics of Nuclear Materials, Intern. Atomic Energy Agency, Vienna (1962), p. 173.

liquid level when liquid was present, this resonance was assigned to resonance in the inert gas inside the cylindrical container. Except for the small 5,000-cps ripple existing when the transducers were excited with a 5-kc carrier signal, the response and degree of signal contamination were quite satisfactory for the stand-off-mounted transducers.

Loop tests (including in-pile data associated with the pressure surge accompanying sample failure) showed the presence of a resonance of the order of 100 cps, which was clearly not a result of 60-cycle electrical pickup, and was different, from the 120-cps true pressure signal arising from the operation of the a.c. electromagnetic pump. Other frequencies could be found, but the 100-cps signal was dominant.

Varying silicone fluid level from that corresponding to loop operation caused a change in the dominant frequency; a greater fluid mass caused a lower frequency, indicating that oscillations were in the fluid-filled part of the loop. Removal of the test section made only a small change in the excited signals. Total loop mass was also a factor: removal of the loop transformer caused an increase in frequency.

In addition, the tests have shown that the upper mounting of the loop test section and the flow deflector at the test-section outlet effectively mask off the pressure transducer at the test-section outlet from transient pressure signals (static measurements are accurate).

These tests have demonstrated that the stand-off technique is adequate for loop meltdown experiments, at least as long as a 5-kc carrier signal is used for transducer excitation and as long as oscillograph galvanometers with approximately 5-kc response are used. In addition, the information on loop response has given data to be included in the design of the Mark-II integral loop.

4. Meltdown Experiments with EBR-II Pin Clusters

Loop meltdown experiments in TREAT have to date been performed on single EBR-II pins, inside a ring of six dummy elements, all on an EBR-II pitch, and with coolant cross section mocking up the coolant cross section in an actual EBR-II subassembly. The next step toward greater simulation of fast-reactor core conditions with the available equipment is use of a cluster of active EBR-II pins. In order to plan such experiments, it is necessary to obtain typical transient-temperature profiles inside such a cluster, and to compare these results with those of similar calculations for a fast-reactor subassembly.

Calculations describing the time- and space-dependent temperature distribution for a seven-fuel-pin cluster in the integral loop was completed. On the basis of equal power generation in each of the pins (because of thermal-neutron flux depressions in TREAT, the central pin must be of higher enrichment than the surrounding pins for this to be the case), the

system was divided into thermally symmetric regions, each of which is amenable to description in terms of axisymmetric concentric cylinders, appropriate for ARGUS input.¹⁹ The principle of superposition was used to combine the results from separate regions, yielding a representation of the temperature distribution for the entire system. Similar calculations were also made for equal power per pin inside an EBR-II subassembly, for both central and peripheral pins (which have different coolant cross-section areas, as well as generally different boundary conditions).

A transient power pulse typical of TREAT and a nominal coolant velocity of 400 cm/sec, realistic conditions for integral loop experiments, were used to obtain temperature distributions. Comparison of the loop results with the subassembly results showed good agreement between central pins and between peripheral pins. Significant differences were found between central and peripheral pins, as has been found previously in other studies.²⁰ Similar results were found in a limiting constant-power case.

Although temperature simulation appears to be good, care must be taken in interpreting meltdown results obtained with a seven-pin cluster and applying the results to a full subassembly. The integral loop has one internal pin for six peripheral pins, whereas the 91-pin EBR-II subassembly has 24 internal pins adjacent to 30 peripheral pins, and there are an additional 37 internal pins forming a "core" inside those outer two rings. This indicates that threshold of failure data should be representative, but that failure propagation effects and gross postfailure and coolant fuel movements will not be weighted in a representative fashion.

B. TREAT

1. Operations

Metal-water reaction capsule CEN-217 was irradiated in room-temperature water. This capsule contained three Zr-clad uranium dioxide fuel rods suspended above the water to simulate a loss-of-coolant accident in which molten fuel falls into a vessel partially filled with water.

Two samples of High Flux Isotope Reactor fuel were irradiated in transparent capsules to obtain photographic data on the physical behavior of the samples while the reaction is in progress.

Three temperature-limited transients were run without samples in the reactor to compare calculated and actual reactor temperature distributions.

¹⁹Schoeberle, D. F., Hustand, J., and Miller, L. B., ANL-6654 (1963).

²⁰Gopinath, D. V., and Dickerman, C. E., ANL-6844 (1964).

2. Large TREAT Loop

Installation of the blanket gas system is in progress. One main valve has again been returned to the manufacturer for repair, and completion of the system is dependent upon the delivery of this valve.

The Zircaloy liner for the in-pile region has been received from the vendor. Dimensions are being checked, and the graphite filler blocks are being fitted to the liner.

The inner flow tube for the test section has required additional work since the original assembly resulted in mismatching of positional dimensions. The crossover bellows assembly is nearing completion.

The design-analysis report has been completed.

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

1. Metal-Water Reactions

a. Short-period Transient Irradiation. Additional results have become available for the metal-water experiments that were conducted during the Kiwi-TNT destructive test in January 1965 in Nevada. This test afforded an opportunity to obtain metal-water reaction data during a short-period (0.6-msec) transient. (Previous transient experiments have been performed in TREAT with reactor periods of 50 to 500 msec.) The Kiwi-TNT transient gave nearly adiabatic heating, thus simulating a very "fast" reactor excursion.

Prior to the Kiwi-TNT power excursion, capsules containing typical reactor fuel materials were placed at various distances from the reactor. A description of the capsules and a summary of most of the results were given in the Reactor Development Program Progress Report for March 1965, ANL-7028, p. 82. However, the data on the extent of uranium-water reaction for the unclad uranium alloy (5 w/o Zr, 1.5 w/o Nb) pins have only recently become available; these are given in Table XVIII. The extent of reaction of uranium with water was determined by analyses of the meltdown residues, rather than by direct hydrogen measurement, the method used in all previous in-pile metal-water reaction experiments. This was necessary because the hydrogen-sampling valves had been damaged during the Kiwi-TNT destructive test.

Three of the samples were analyzed by the following method. The unreacted uranium (metal) was dissolved in bromine-ethyl acetate; the uranium oxide residue was then dissolved in nitric acid. The uranium and UO_2 contents were determined by a colorimetric uranium analysis of each portion of the sample.

TABLE XVIII. Metal-Water Reaction Data for Uranium Alloy^a Fuel
Subjected to Kiwi-TNT Short-period Transient

No.	Distance ^b (ft)	Estimated Fission-energy Input (cal/g)	Estimated Peak Adiabatic Core Temperature ^c (°C)	Condition of Fuel after Transient	Metal-Water Reaction (%)	d _{sv} ^d (mils)
1	2/3	420	3800 (L+V)	Fragments and fine particles	77	6
2	2	200	3800 (L+V)	Completely melted into fine particles	76	3
3	3	125	2500 (L)	Fragments and fine particles	52	6
4	3.5	100	2200 (L)	Melted into globules and fines	65	21
5	5	63	1133 (L)	Melted into globules and fines	21	19
6	6	50	1133 (S+L)	Partially melted	3.4	98

^a93.5 w/o U, 5 w/o Zr, 1.5 w/o Nb, 20% enriched, 0.2-in. dia by 0.5 in. long.

^bDistance between autoclave and reactor vessel.

^cPhysical state of sample at that temperature indicated by V (vapor), L (liquid), and S (solid).

^dMean volume-to-surface particle size of fragmented sample.

For two samples an alternative procedure was used. The entire sample was first hydrided, that is, the uranium metal was converted to the hydride while the oxide remained as UO₂. Then, the sample was dehydrided by heating, and the hydrogen evolved was measured with a gas chromatograph, thus determining the amount of uranium metal present in the original sample. Following this, the entire sample was dissolved in nitric acid, and the total uranium content was found colorimetrically.

The data show a clear increase in the amount of chemical reaction with peak uranium temperatures. In addition, the Kiwi-TNT results are generally consistent with two other, quite independent, sets of data on high temperature uranium-water reactions. These are TREAT meltdown experiments²¹ and condenser-discharge tests.²²

²¹Chemical Engineering Division Summary Report, July, August, September, 1962, ANL-6596, p. 191.

²²Chemical Engineering Division Summary Report, April, May, June, 1960, ANL-6183, p. 141.

V. PUBLICATIONS

PapersSURFACE TEMPERATURES OF INFINITE PLATES SUBJECTED TO
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T. R. Bump

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Symp. on Two-Phase Flow, University of Exeter, June 21-22,
1965. Dept. Chem. Eng., University of Exeter, Exeter,
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H. K. Fauske

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- ANL-7086 THE EFFECT OF INTERPARTICLE FORCES ON THE
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- ANL-7092 CATALOG OF NUCLEAR REACTOR CONCEPTS.
Part I. Homogeneous and Quasi-homogeneous Reactors.
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Charles E. Teeter, James A. Lecky, and
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