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Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

August 1965

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT,

August 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:

February 1965	ANL-7017
March 1965	ANL-7028
April 1965	ANL-7045
May 1965	ANL-7046
June 1965	ANL-7071
July 1965	ANL-7082

Reactor Development Program
Highlights of Project Activities for August 1965

EBR-II

The reactor has been operated to 45 MWt and about 1.1 a/o maximum burnup. The methods of fission product monitoring appear to be satisfactory. The anomalous swelling behavior of the special surveillance samples may be due to poor quality of sodium bonds.

FARET

The A-E Title II design is essentially complete. All design packages, except V and IX, have been ready for signature since early 1965, but approval is being withheld pending completion of AEC review.

Bechtel's proposal for construction of two working models is being evaluated by the Laboratory.

FARET criticals were completed. Final experiments were a series of material substitutions in various fuel zones.

ZPPR

A contract for coring of the Gravel Gertie structure was awarded to Eberline and Associates, Santa Fe, New Mexico.

General Fast Reactor Fuels Development

Six U-15 w/o Pu-10 w/o Fs samples in V-20 w/o Ti tubing were irradiated to 5 a/o burnup before the first evidence of failure occurred; these capsules had been designed for 3.1 a/o burnup. Samples of the U-Pu-Ti and U-Pu-Zr alloys have successfully gone beyond 4 a/o burnup.

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I. LIQUID-METAL-COOLED REACTORS

A. EBR-II

1. Operations

The reactor was operated at power levels to 45 MWt until August 12, when it was shut down after attainment of about 1.1 a/o maximum burnup. Thereafter, the reactor was operated at increments of power up to 45 MWt for physics measurements until August 16. During the remainder of the month, the plant was maintained in standby condition while awaiting the results of the inspection of surveillance fuel elements. During this time, work continued on the replacement of a control rod drive mechanism and on the rotating plug seals (see Section I.A.2).

At shutdown, the cumulative production of thermal energy for the reactor was approximately 2900 MWd. During power operation, detailed observation and surveillance of operation of the fission product monitor was performed (see Section I.A.4.b).

2. Systems and Components

a. Rotating Plug-seal Troughs. Investigation continued of the large plug-seal trough (see Progress Report for July 1965, ANL-7082, pp. 1-2) after completion of fuel handling on August 20. The distribution of oxides or oxide-metal mixtures seems to be affected by previous plug rotation and the length of time the alloy is molten.

A piece of steel tubing attached to a vacuum system was inserted into the "window hole" of the large plug dip ring and used to remove oxide from the top surface of the molten alloy. Approximately 2 lb of black powdery oxide, nodules, and metal were recovered by this procedure. Samples of these substances are being analyzed chemically to determine their composition.

b. Control Rod Drives. A previously used control drive shaft, which had been in storage as a spare, was disassembled for examination of the gripper shaft bellows. A small hole was found in the lower bellows, probably caused by local overheating during the cleaning process for removal of residual sodium. New replacement bellows have been ordered.

Because of malfunction of the sensing rod in control drive No. 9 (see Progress Report for July 1965, ANL-7082, p. 3), the drive was removed from the primary tank on August 23. It will be examined to ascertain the cause of the sensing rod malfunction. A replacement drive, which had been previously reworked and leak-tested to verify bellows integrity, was installed on August 25.

c. Sodium and Argon Systems. The primary sodium plugging temperature remained less than 220°F. The secondary sodium plugging temperature was kept below 300°F by intermittent use of the cold trap.

The composition of the secondary argon remained relatively constant at 8 ppm hydrogen and 1800 ppm nitrogen. The composition of the primary argon cover gas also remained relatively constant, showing less than 10 ppm hydrogen and 1.5% nitrogen.

Special sodium samples were taken and sent to Argonne, Illinois to determine hydrogen by an "isotopic dilution" technique. Results indicate that the primary sodium contains 4 ppm hydrogen and the secondary sodium 2.5 ppm.

3. Fuel Handling

As part of the fuel-surveillance program, a safety rod and fuel sub-assembly from row No. 3 were replaced with fresh subassemblies and stored in the basket. Replacement of the new safety rod ran into difficulty because of an error in procedure and the misalignment of the rod top adaptor. The rod was removed and replaced with a satisfactory one. Subsequent tests indicate that no damage was done to the fuel-handling gripper.

Both subassemblies which had been removed from the reactor were transferred to the Fuel Cycle Facility for surveillance measurements during the week of August 23 after having been allowed to cool in the basket. Fuel elements in these subassemblies have a maximum of 1.1% burnup.

4. Reactor Physics

a. Reactor Stability Monitoring. During reactor operation to 1.1 a/o maximum burnup, measurements relative to reactivity monitoring were made as follows:

(i) Rod Calibration--Control rods No. 3 and No. 10 were recalibrated. The worths of these rods were consistent with previous calibrations.

(ii) Rod Drop--The trace of power versus time was obtained during and after the dropping of special control rod No. 8. The results were consistent with data taken during previous power runs which showed approximately 16% of the total feedback recovered in 0.5 to 1.0 sec.

(iii) Trapezoidal Tests--The reactor was oscillated, by means of the trapezoidal equipment, at 40 MW and again at zero power. Data were taken at drive times of 11.6, 5.8, and 1.0 sec. The results of 11.6- and 5.8-sec data are given in Table I. The 1.0-sec data are being analyzed. The ratios $\Delta n/n$ of the 40-MW to zero-power data show a strong negative feedback component at each of the measured frequencies.

TABLE I. Trapezoidal Data

Wait Time (sec)	11.6-sec drive $\Delta n/n$			5.8-sec drive $\Delta n/n$		
	(a) Zero Power	(b) 40 MW	Ratio (b/a)	(a) Zero Power	(b) 40 MW	Ratio (b/a)
3	0.079	0.070	0.89	0.035	0.031	0.89
20	0.137	0.108	0.79	0.070	0.054	0.77

(iv) Power Coefficient--The power coefficient as a function of power was measured during the increase in power at the beginning of this run and during the decrease in power at the end of the run. The overall reactivity results gave 77 and 74 lh, respectively. The results of these reactivity-monitoring techniques confirm that the value of the overall shutdown coefficient has not been affected by the present condition of the fuel.

b. Fission Product Monitoring. Because of the increased emphasis on reliable fuel rupture detection, the Fuel Element Rupture Detector (FERD) system was carefully observed during power operation this month. By suitably adjusting the discriminator on counting circuit Channel B (BF_3 and B-coated counters), it was possible to reduce the equilibrium sodium background level to less than 5 cps without significantly affecting the neutron sensitivity. Present measurements indicate that the normal background neutron level is approximately 0.5 cps/MW. Channel A (fission counters) by comparison is less sensitive to neutrons by a factor of 12.

Routine periodic sampling of the cover gas for fission products was instituted to establish the background level for Xe^{133} and Xe^{135} as a function of power operation.

c. Buildup of Plutonium Reactivity. The reactor was maintained at zero power for three days after the high-power run to evaluate any long-term reactivity effects, which may be due to buildup of plutonium. A total increase of 5 lh was noted, which is approximately as calculated.

5. Digital Computer Control for EBR-II

A study was initiated to determine if EBR-II could be controlled by a digital computer. The plant can be completely automated only if its state is known as a function of time and space. It is, therefore, necessary to understand the transient behavior of the complex system from its dynamic model.

For accurate control of the plant, the distributed-parameter approach is preferable to a lumped-parameter model because it results in a more accurate design. Accordingly, this investigation was divided into

the following four phases: (a) derivation of transfer matrices for the heat-exchange processes; (b) rational approximations of the transcendental transfer functions and analog computer simulation; (c) optimization studies; and (d) implementation of results.

(a) Derivation of the Transfer Matrices for the Heat-exchange Processes. The EBR-II power plant is a distributed-parameter system and is described by partial differential equations with respect to time and space coordinates. The partial differential equations, in turn, are concerned with the derivation of transfer matrices relating the reactor power level, inlet and outlet temperatures, and flow rates for the reactor, the primary heat exchanger, and the secondary heat exchanger.

(b) Transfer Function Approximation and Analog Computer Simulation. In the design of systems with transcendental transfer functions, mathematical difficulties are encountered in the determination of the system response and in analog-computer simulation. The transcendental transfer functions are, therefore, approximated in a suitable manner. The standard technique is to expand the functions in power series and retain the first few terms of these series. A serious disadvantage of these shortened approximations is that in certain cases instability of the system is implied where it does not actually exist. Oldenburger and Goodson employed infinite products instead of infinite series in order to eliminate this difficulty of instabilities and have shown that it is sufficient to retain only the first few terms of the infinite product expansion to represent adequately the transcendental transfer function, which is thereby replaced by a rational approximation. In the investigation of EBR-II digital control, the transcendental transfer functions will be approximated by employing Oldenburger and Goodson techniques, which permits simulation of the system on the analog computer by using conventional components. By the on-line use of a digital computer, a sampled-data system must be employed. Garvey and D'Souza have developed a technique of forming a representative sampled-data system for a given distributed-parameter system. A fictitious sample-and-hold circuit is inserted in the system and the Z-transform of the resulting representative sampled-data system is obtained. The system can then be analyzed and synthesized by employing sampled-data theory and a digital computer.

(c) Optimization Studies. It is economically desirable to control the variables in a suitably defined optimal fashion. The purpose of this portion of the study is to generate optimal control laws from which the input variables to the system can be determined such that a given performance index is extremized. The control laws are functions of the distributed state variables of the system.

Power reactors are ordinarily taken from one power level to another over a substantial period of time in order to permit the reactor and its subsystems to reach nuclear and thermal equilibrium between

increments of power. A suitable performance index for optimization appears to be a determination of the minimum time for change from one power level to another while considering the physical constraints and minimum effort so as to prolong the life of the fuel. Furthermore, the digital computer must command the nuclear reactor to provide power optimally at the levels required by the load from time to time. At the same time, the relationship between reactor power level and flow rates must be established and maintained. Among the variables that may be manipulated are the position of the control rods (the reactivity removal is a nonlinear function of the position), pump speeds and valve positions which change the primary and secondary sodium flow rates, drum water level, and steam flow rate.

(d) Implementation of Results. The proposed control system should be a combined digital computer-analog control hybrid system. Suitable inputs from the different sensors will be fed to the digital computer by analog-to-digital converter channels. From the preprogrammed instructions, the computer will synthesize the optimum control laws and supply control signals to the analog controllers. The startup and shutdown operations will thus be completely automated.

6. Fuel Cycle Facility

a. Argon and Air Cells. After replacing bearings and substituting a lighter lubricant on the south fan for the gas-circulation system of the Argon Cell, it was reinstalled and is now operating satisfactorily. The fan bearing temperature is about 145°F, and the radiation field in this area is several R/hr.

A failed grip drive unit on an Argon Cell manipulator was replaced remotely. A Model A manipulator slave unit in the Argon Cell developed operating difficulties and was replaced with a spare. Preparations are being made for its removal to a glovebox for repair.

A limit switch on the Air Cell crane was replaced after withdrawing the crane through the roof hatch. Radiation exposure was minor.

b. Fuel Processing and Fabrication. One core-type subassembly (of 1.06 a/o burnup) has been dismantled in preparation for examination and processing. Nine melt-refining runs and corresponding slag-oxidation runs were completed.

A pin in the T-bar handle of the oxide dumper broke, and this was repaired by remote means.

An examination of the data for the pouring yields obtained after nine melt-refining runs with irradiated fuel (no recycle material), for which yields ranged from 92.7 to 94.5%, indicated a decrease in yield of

about 0.24% for each 0.1% maximum atom percent burnup. This corresponds roughly with estimates of quantities of fission products lost to the slag.

In fuel-fabrication operations, six injection-casting runs were completed and 410 pins were inspected to yield 245 acceptable pins. Assembly of 316 elements was accomplished, and 388 were leak tested (5 were rejected). Bonding and testing yielded 363 acceptable elements, and assembly of four new reactor assemblies was completed.

c. Fuel Surveillance. Postirradiation measurements are being made at the Lemont site of a group of special fuel rods as part of the fuel-surveillance program (see Progress Report for July 1965, ANL-7082, pp. 5-7). These rods had been given careful preirradiation measurements. They were intended to provide a comparison between information obtained from prior capsule irradiations of the EBR-II fuel in thermal reactors and the behavior of the fuel in EBR-II itself. More recently, detailed postirradiation examinations have been started on regular production fuel to supplement surveillance data being obtained at the Idaho site.

The data obtained to date indicate that the volumes of the special fuel pins are increasing more rapidly with increasing burnup than those of the regular fuel pins. In fuel subassembly C-138, which achieved a calculated maximum burnup of 1.0 a/o, six special fuel pins showed volume increases ranging from 4.50 to 8.68%. All measurements were on bare fuel pins, and volumes were obtained from immersion densities. A single regular pin from the same subassembly showed a volume increase of 3.86%, assuming that the fuel had the average preirradiation density of the special fuel pins. Observations of sodium levels at the Idaho site have shown consistently higher levels in the special rods as compared to the regular rods. The higher sodium levels in the special pins appear to be the result of greater changes of fuel volume rather than due to higher initial levels of sodium.

The reason for the greater rate of volume increase in the special fuel rods has not yet been established. The fuel pins in the special rods were taken from production runs, so that there is no evident compositional difference. Bond-quality tests of irradiated fuel rods at Idaho have shown that the sodium bond in the special rods is generally poorer than that in the regular rods. This could lead to increased thermal resistance between the fuel and coolant, which in turn could cause higher fuel temperatures and increased swelling rates. At the moment this appears to be the most logical explanation for the different behavior of the special fuel rods.

In a separate experiment a regular clad fuel rod with 1.0 a/o burnup from subassembly C-138 was heated to temperatures of 550, 600, 645, 700, 750, and 775°C. At each temperature dimensional and volume changes were measured. No significant change was noted until after the final heat treatment at 775°C. A maximum increase in diameter of 0.0045 in.

occurred in the lower fuel section, accompanied by a volume increase of 0.36%. Leak testing showed that the cladding was still intact. The cladding was then removed from the fuel pin, which showed a volume increase of 11.6%.

The annealing results give considerable encouragement that the present EBR-II fuel rod can tolerate, without clad failure, substantially greater fuel-volume increases than may have yet occurred in the reactor. Nevertheless, in such a large number of fuel rods as are present in the EBR-II core, normal statistical variations would indicate that early random failures can be expected that would not necessarily be indicated from tests on small lots. For this reason, occasional releases of fission products into the EBR-II coolant as fuel burnups proceed to higher and higher levels should not be entirely unexpected.

B. FARET

Note: Please note that Figs. 2 and 7, together with their titles, of Progress Report for July 1965, ANL-7082, on pages 9 and 13, have been interchanged.

1. General

There has been no change in the status of the Architect-Engineer's portion of the Title II design during the previous month. The design is essentially complete and all design packages have been signed and issued, except Packages V and IX. The later package drawings and specifications have been ready for signature since early June. Approval is being withheld until completion of AEC FARET Project reviews and the lack of a firm date for start of construction.

Pending authorization to proceed with Title III, work at the Architect-Engineer offices is at a very low level, requiring approximately 3 to 4 men full time. The efforts of these people has been directed for the following:

a. The Architect-Engineer Title II cost estimate for design Packages V and IX has been completed and transmitted to the AEC. This completes the estimates for all of the Title II design.

b. The final updated design criteria have been completed by Bechtel and transmitted to the Laboratory for review and comments.

c. Minor revisions of design Packages III, Liquid-metal Heat Exchangers, and IV, Liquid-metal Pumps, is essentially complete, and the Laboratory has been advised that the material will be available for ANL and AEC signatures by the end of August.

d. Design specification VI is being rewritten to include only liquid-metal valves. The special stainless steel piping formally covered by this specification is now being included in another specification which will cover all project piping. The reason for separation of the special stainless steel piping from the rest of the piping specifications was to permit UE&C to expedite early procurement by making such specifications available prior to the expected availability of the general piping specifications. However, AEC approval to proceed in this manner has not materialized; as a result, the need for special treatment of the long-lead piping procurement no longer exists. Consequently, it is now more advantageous for ultimate project procurement activities to direct Bechtel to make the required specification changes. The extra costs included are considered significant, but not excessive.

e. Bechtel has submitted a proposal to the Laboratory for construction of two working models. One model, on a 3/8-in.-to-the-foot scale, will include the cell. The second model, on a 3/4-in.-to-the-foot scale, will include the cavity and vault structure and internals. This proposal is now being evaluated by the Laboratory.

The Laboratory has participated in an extensive series of meetings with the AEC to review the project status. The entire scope of project activities including the Bechtel Title II design, the ANL Title II design, the current schedule, and cost estimate were reviewed. The purpose of these discussions was to provide Commission personnel with further understanding of the technical and management aspects of the project.

The current delay in authorizing the initiation of Title III continues to multiply schedule and cost-estimating problems. The liquid-metal heat exchanger procurement, Package III, and the liquid-metal pumps procurement, Package IV, are now becoming dominating factors in establishing the construction schedule. In addition, costs continue to increase as a consequence of changing market conditions.

The Laboratory has received from UE&C drafts of bid invitations for a significant portion of the UE&C procurements. This material covers several million dollars worth of procurement. ANL comments have been returned to UE&C and incorporated in final drafts which appear ready for release to qualified vendors. The invitation packages include the liquid-metal pumps, liquid-metal heat exchangers, instrumentation cell vault and cavity liners, and personnel air locks.

2. Fuel Assembly Sodium Flow Test Loop

Installation of the thermal insulation and pipe-line heaters is continuing.

The loop pump is essentially ready for final assembly at the Byron Jackson plant. The pump will be tested at the plant prior to shipment to ANL, which is scheduled for early September. The temporary pump now being used with this loop (100 ft at 230-gpm rating) will be replaced by the new Byron Jackson pump, with a rating of 320 ft at 800 gpm. This large pump is essential for completion of the test program intended for the loop.

3. Static Test Facility

Chemical-analysis data for the three small pressure vessels required for this facility have been submitted to ANL. It has been verified from these data that one section of the 4-in.-dia vessel was fabricated using a fitting of Type 304L stainless steel and is therefore unacceptable to the Laboratory. Negotiations are now in progress with the fabricator to correct this situation.

The pit in Building 206 to accommodate the FARET static test facility has been relocated (see Progress Report for July 1965, ANL-7082, p. 10). New bids will be solicited on or about September 1.

4. Cell-annex Bellows

The design of the FARET cell and its annex provides for a large vertical bellows, extending from the floor of the cell up to the underside of the annex floor, to provide isolation between the cell atmosphere and the annex atmosphere. It will then be possible to operate the facility with a significantly different atmosphere in the annex than in the cell. Such operating conditions are expected to provide increased access to the control rod drives and an inherently more favorable environment to promote increased operational reliability of the drives themselves.

a. Performance Tests. A 1/4-scale test bellows, 33 in. across flats on the outside and 27 in. across flats on the inside, is now being fabricated. The bellows will consist of three complete convolutions and a 2-in.-high collar on each end to attach the bellows to appropriate flanges. The test bellows will be constructed of a double layer of 0.018-in.-thick Hypolon neoprene-coated nylon fabric. Aluminum slats, 0.050 in. thick, will be bonded between the two layers of fabric.

A testing device for these sample bellows has been designed and will be fabricated shortly. With its use, the sample bellows section will be evaluated for the effects of differential pressure, flexing, and characteristics of oxygen and moisture transmission.

b. Material Tests. These tests will be conducted on the same material as specified for the test bellows series as described above. They will be devoted entirely to investigating the properties of the basic materials without regard to application. The tests will cover differential pressures, flexing, and transmission of oxygen and moisture under simulated conditions of FARET cell operations.

5. Control-rod Drives

a. Test of Bellows Seal. The tests of the control-rod drive-shaft bellows seal is continuing successfully. At this time the bellows seal has successfully accumulated 8885 cycles.

b. Control-rod Electrical Components. A test fixture for the control drive electrical cable connector penetration has been designed and is presently being fabricated. The test fixture simulates the proposed penetration in the top of the external tank for the control drive cables. The test fixture consists of a steel housing and two flanged and gasketed electrical cable connectors.

It is planned to test the fixture at 30 psig for structural strength and leaktightness, which are FARET operating conditions.

6. Fuel Cut-off Machine

The window support stand, the personnel platform, and the General Mills Manipulator which are to be used for the FARET cell mock-up are being installed in Building 208. The experimental instrument-lead cutters and an associated test stand are being fabricated. These units will be installed in the mock-up cell to test operation, viewing, and handling features of the designs. They will also be used to test durability of the cutter blades when functional tests are completed.

7. Shielding Windows

A series of tests has recently been completed at the Laboratory in connection with the proposed shielding windows for the FARET cell. Conclusions from previous tests (see Progress Report for February 1965, ANL-7017, p. 14) are as follows:

a. It is feasible to use a heavy glass slab, approximately 36 x 36 x 6 in. thick, for a gas-tight sealing member. The window should be able to support a differential pressure of at least 60 psig across the window when using simple flat gaskets.

b. In order to withstand potential missile damage originating from within the cell, the design concept of three slabs in series, with the most protected slab being used as the primary gas seal, is feasible.

c. The design concept using three slabs in series is also feasible for withstanding transient high-temperature conditions, even if the high transient temperature exposure is as long as one hour.

The above conclusions were substantiated in recent tests in which two glass slabs, $29 \times 44 \times 8\frac{1}{2}$ in. thick, together with a $1/2$ -in. thick tempered glass cover plate, 30×44 in., were used.

Each heavy glass assembly had its own steel frame. The $1/2$ -in. cover plate was supported by aluminum holddown bars at top and bottom only. Koroseal gaskets were used throughout for cushioning of glass against metal. The heavy steel test fixture in which the three glasses were mounted was lifted into an erect position after the glass slabs and frame had been assembled. A vertical bank of radiant heaters (Nichrome elements within quartz tubes) was positioned 10 in. from the surface of the cover plate. Bridging the gap at the top between the heater bank and test fixture bottom and sides were aluminum reflectors, backed by transite sheets. These completed the heat trap enclosure on the heated side.

With this arrangement, several trial runs at low heater power produced no noticeable effects on the glass. Following these trial runs, a 38-min test was made starting the heaters at 13.2 kW. At the end of the first 20 min heater power was raised to 17.3 kW. The front and back surfaces of the $1/2$ -in. slab were 49°F and 430°F , respectively, at the end of this test. The front and back surfaces of the middle thick slab were 260°F and 120°F , respectively. The surfaces of the outer heavy slab only reached 110°F and 90°F under the conditions of this test. There appeared to be no ill effects to either glass or gaskets. It is estimated from the observed rate of temperature rise that the heat-absorption rate was approximately 500 Btu/hr-ft², whereas it had been expected to be approximately 3000 Btu/hr-ft².

A second test was performed in which the $1/2$ -in. glass cover plate was removed, permitting exposure of the center slab of $8\frac{1}{2}$ -in. thick glass directly to the heaters. The surface of this slab reached 340°F in 31 min. (During this test the Koroseal gasket cushion on the hot side of the first slab had softened and started to drip at about 290°F .) When the temperature subsequently reached 342°F , the first glass slab cracked, but remained in place in the frame. Neither the back surface temperature of the cracked slab nor any temperatures of the uncracked slab changed perceptively in the next half-hour. It may be concluded that there was no significant heat transfer through the first slab which resulted solely from the crack. Inspection of the outer slab of glass, which was not cracked, indicated no visible damage as a result of this test.

8. FARET Criticals

The program of experimental studies based on the proposed design of the FARET reactor was completed. During the month, traverses of reactivity samples and fission counters were run radially through Core 46F, a mock-up of a multituel-zoned FARET core (see Progress Report for July 1965, ANL-7082, pp. 14-17). The final experiments were a series of material substitutions in the various fuel zones of Core 46F to determine reactivity coefficients.

Core 46F contains an inner zone of an oxide-fuel simulation ($\text{PuO}_2\text{-UO}_2$) surrounded by annuli of carbide, oxide, and a mixed oxide and alloy simulation fuel zones. Traverses of fission chambers were made radially at the midplane of the reactor to determine the variations of fission rates and power densities in the core zones. The various chambers used were about 1/2 in. in diameter by 2 in. long, plated with enriched uranium, depleted uranium, and plutonium. A small-diameter BF_3 proportional counter was also traversed. The results obtained with the U^{235} and U^{238} fission chambers and the BF_3 counter are plotted in Fig. 1.

Traverses were also run radially with small reactivity samples of Pu, U^{235} , U^{238} , and B^{10} to measure the reactivity coefficients of these materials as a function of radius. Data from these experiments are undergoing analysis.

A series of material substitutions was made in Core 46F to find the average reactivity coefficients in the four different fuel zones. Sodium coefficients were determined also in two axial regions of the zones. These coefficients are necessary to correct for differences in composition between the mock-up fuel simulations and the actual FARET specifications. Table II lists the results of the experiments, expressing the material worths as reactivity per kg of the material replacing void.

TABLE II. Average Worths of Materials in Core Zones of ZPR-3 Assembly 46F

Material	Reactivity Coefficients, lh/kg				
	Radial Zone I (Oxide)	Radial Zone II (Carbide)	Radial Zone III (Oxide)	Radial Zone IV (Alternate Oxide and Alloy)	
				Oxide Drawers	Alloy Drawers
Na, 0 to 4 in., axially	25	48	63	66 ^a	66 ^a
Na, 4 to 10 in., axially	52	59	48	48 ^a	48 ^a
SS, Type 304	2.4	5.2	9.8	13.6	10.7
Depleted U	3.3	6.2	4.9	5.6 ^a	5.6 ^a
U_3O_8 (depl. U)	12.6	-	15.6	17.6	-
Graphite	105	119	109	-	-
Oxygen ^b	65	-	76	84	-
Zirconium	-	-	-	-	15.7

^aAverage worths in oxide and alloy drawers together.

^bDerived from depleted uranium and U_3O_8 coefficients.

Fig. 1. Radial Traverses of Neutron Detectors through ZPR-3 Assembly 46F

C. General Fast Reactor Physics

1. ZPR-6

a. Doppler Measurements. Doppler measurements with Assembly No. 4Z (see Progress Report for May 1965, ANL-7046, p. 14) continued. Measurements were made with samples containing appreciable fractions of U^{235} in U^{238} , with U^{235} alone, and some repeat measurements were done with U^{238} to check reproducibility.

In the U^{235} measurements three different element types: two types of oxide element (one allowing no axial expansion, the other allowing free expansion), and a freely expanding U^{235} metal element, were used. The nonexpandable $U^{235}O_2$ element yielded a reactivity effect per gram of U^{235} about one-third the magnitude of U^{238} , but positive. The ratio of the U^{235}/U^{238} effects varied with temperature and decreased as the temperature increased. The freely expanding $U^{235}O_2$ element gave results that were also positive, but somewhat smaller in magnitude. Indicating a small negative expansion effect in the oxide samples.

The U^{235} metal element was heated through the α - β phase change in order to provide a calibration point for the expansion effect in this sample. The temperature-reactivity curve for this metal sample was oddly shaped, going negative at high temperatures. However, the experimental curve may be matched very well through the use of published data on U^{235} metal expansion, the phase transition reactivity calibration of the expansion effect, and the positive Doppler effect for U^{235} found in the oxide samples.

A measurement done with the same boron filter that reduced the U^{238} Doppler effect to 20% of its previous value reduced the U^{235} Doppler effect by a much smaller amount.

The basic measurements with $U^{238}O_2$ were repeated this month, some ten weeks after they were done initially. In the intervening period many changes had been made in the core and driver loadings, but after returning the reactor to the original configuration, the results checked precisely with those taken previously.

b. Calculations of Fat-man Effect for ZPR-6 Assembly No. 5. The Fat-man effect for the proposed ZPR-6 Assembly No. 5 has been calculated for a large number of cases, starting with a bare half (for which $k_{eff} = 0.8410$) and then calculating the effect of covering the interface with different materials of varying thicknesses. Assembly No. 5 has the composition of a carbide-fueled reactor (critical mass of 1525 kg U^{235}); has a U^{238}/U^{235} volume ratio of 6.94 and a cylindrical core of 2554 liters; has a radius of 75.60 cm and a height of 142.24 cm ($L/D = 0.94$). The radial and axial

TABLE III. Volume Fractions in ZPR-6 Assembly No. 5

Material	Volume Fractions	
	Core	Blanket
U^{235}	0.0319	0.00169
U^{238}	0.2213	0.8319
Na	0.4208	-
C	0.1539	-
SS (Considered as Fe)	0.1483	0.0708

blankets are made up of depleted uranium, 30 cm thick. Table III gives the composition of core and blanket.

The moderator slab (Fat Man), covering the half, was assumed to consist of ordinary water or Benelex. The composition of Benelex is given in Table IV.

The absorbers considered for introduction between the Fat Man and the core are either iron or Boral (or a combination of the two). Boral has the average composition shown in Table V.

Figure 2 gives the calculated effects of slabs of Benelex, water, or iron located adjacent to the interface. The reactivity increases with the thickness of homogeneous materials (Benelex or water) rapidly at first and then levels off after about 20 cm, when the condition of an infinite reflector is approached. A similar trend is obtained when iron is used, but the leveling off is less pronounced.

TABLE IV. Composition of Benelex

Element	N (10^{24} atoms/cm ³)
H	0.055
C	0.034
O	0.024

TABLE V. Composition of Boral

Element	N (10^{24} atoms/cm ³)
Al	0.0460
B	0.0259
C	0.0064

Interposing sheets of Boral between the half assembly and the Benelex slabs (kept constant at either 20 or 50 cm) reduces reactivity (see Fig. 3). It is interesting to note that upon going from 1/4-in. to 1/2-in. Boral, there is a slight increase of reactivity. It is not yet clear if this result is a genuine effect or whether it derives from inaccuracies in the calculations.

As the Benelex used for measurements of the Fat-man effect may contain up to a maximum of 100 ppm of boron, some of the calculations (see Table VI, Cases No. 3 and 5) were performed with this quantity being added to the Benelex composition. The changes in k_{eff} were insignificant.

Figure 4 (Curve G) shows the effect of interposing iron between the half assembly and 1/4-in. Boral plus 20-cm Benelex. In this case, the second absorber causes a slight increase in k_{eff} .

Fig. 2
Fat-man Effect--The Influence of Benelex, Water, or Iron on the Reactivity of a Bare Half of the ZPR-6 No. 5 Assembly

Fig. 3
Fat-man Effect--Influence of Boral on Reactivity When Placed between the Bare Half and Either 20- or 50-cm Benelex on ZPR-6

TABLE VI. Calculated Fat-man Effect

Item No.	Geometrical Configuration	k_{eff}
1	Bare Half	0.8410
2	Bare Half + 10-cm Benelex	0.8950
3	Bare Half + 20-cm Benelex	0.9079
		(0.8965)*
4	Bare Half + 50-cm Benelex	0.9085
5	Bare Half + 1/4-in. Boral + 20-cm Benelex	0.8497
		(0.8497)*
6	Bare Half + 1/2-in. Boral + 20-cm Benelex	0.8510
7	Bare Half + 1/4-in. Boral + 50-cm Benelex	0.8500
8	Bare Half + 1/2-in. Boral + 50-cm Benelex	0.8510
9	Bare Half + 1/2-in. Iron + 1/4-in. Boral + 20-cm Benelex	0.8531
10	Bare Half + 1-in. Iron + 1/4-in. Boral + 20-cm Benelex	0.8564
11	Bare Half + 1/2-in. Iron + 1/4-in. Boral + 50-cm Benelex	0.8531
12	Bare Half + 1-in. Iron + 1/4-in. Boral + 50-cm Benelex	0.8564
13	Bare Half + 1/2-in. Iron + 20-cm Benelex	0.8719
14	Bare Half + 1-in. Iron + 20-cm Benelex	0.8628
15	Bare Half + 1/2-in. Iron + 50-cm Benelex	0.8720
16	Bare Half + 1-in. Iron + 50-cm Benelex	0.8628
17	Bare Half + 10-cm Iron	0.8683
18	Bare Half + 20-cm Iron	0.8815
19	Bare Half + 50-cm Iron	0.8902
20	Bare Half + 10-cm Water	0.8887
21	Bare Half + 20-cm Water	0.8983
22	Bare Half + 50-cm Water	0.8987

*This calculation was made assuming that the Benelex contains 100 ppm of boron.

Fig. 4
Fat-man Effect--Influence of Iron on
Reactivity When Placed between Core
and Moderator

For the evaluation of the Fat-man effect in the various cases described here, use was made of the MACH-1 one-dimension diffusion program and of the ANL 16-group cross-section set No. 201. A few cases which involve no strong moderator have also been calculated with use of the ANL 22-group cross-section set No. 224, which extends only to a low energy limit of 29 eV. A comparison of the results is offered in Table VII.

TABLE VII. Comparison of Calculations Run with Cross-section Sets 201 and 224

Geometrical Configuration	k_{eff}		Item in Table VI
	#201	#224	
Bare Half	0.8410	0.8445	1
Bare Half + 10-cm Iron	0.8683	0.8945	17
Bare Half + 20-cm Iron	0.8815	0.9091	18
Bare Half + 50-cm Iron	0.8902	0.9171	19

The results of the present calculations indicate that, for the assembly here considered, sufficient reduction in k_{eff} can be achieved by shielding the interface of the half assembly with rather modest thicknesses of absorbing materials. In such conditions, work between the halves of Assembly No. 5 will be possible, provided adequate protection is used.

The case of closing ZPR-6 with only one half loaded (Item Nos. 17, 18, 19 of Table VI and Curve C of Fig. 2) also does not present particular problems.

c. Instrument Development. A Li^6 semiconductor detector fast-neutron spectrometer has been the object of considerable experimentation at AFSR. Experiments were conducted with this device in ZPR-6 Assembly 4Z to provide a direct comparison of the Li^6 device with measurements with an experimental proton-recoil proportional counter and with the calculated spectra in this assembly.

The Li^6 fast-neutron spectrometer consists of a pair of semiconductor surface barrier diodes mounted with their sensitive surfaces facing each other and with a thin film of Li^6 suspended between them. The total energy from the $\text{Li}^6(n, \alpha)\text{H}^3$ reaction is measured for each event when the spectrometer is exposed to the in-core flux. The result is a pulse-height distribution which can be processed to give data on the neutron flux per unit energy as a linear function of neutron energy.

Runs were made with the spectrometer located near the core midplane just above the core center line (position 23 horizontal and 22 vertical), in the depleted uranium blanket (position 11 horizontal and 23 vertical),

and in the driver zone (position 16 horizontal and 18 vertical). Useful data were obtained in the core-center position and in the blanket position.

Fully satisfactory methods of extracting the neutron spectrum from the experimental data have not yet been found. To compare the results of these experiments with the other data, expected pulse-height distributions were calculated from neutron spectra obtained by other means and the known response function of the semiconductor fast-neutron spectrometer.

In both regions of the ZPR-6 assembly, the semiconductor detector neutron spectrometer observed fewer low-energy (below 100 keV) neutrons than were observed with the proton-recoil spectrometer.

The comparison between the semiconductor spectrometer data and the calculated spectra is quite good. The calculations were based on a 22-group, MAIM-6 computer code, using homogenized cross sections (Set 801) ELMOE-averaged over the constituents.

2. ZPR-9

After completion of the studies with Assembly No. 6A, Assembly No. 7 was constructed; the core was dismantled, and the core and reflector region were loaded with drawers filled with aluminum oxide. Core drawers containing fuel and diluent materials were then added a few at a time, and the displaced aluminum oxide drawers were moved to the outer edges of the reflector. Multiplication measurements during this method of loading predicted a critical mass of 278 kg of U^{235} . Initial criticality was achieved at 278.5 kg of U^{235} and an excess reactivity of ~ 29 lh. In subsequent loading changes both the core and the reflector were cylinderized. The addition of 102 drawers of aluminum oxide to cylinderize the reflector added considerably more reactivity than was predicted analytically. Subsequent studies were directed toward the change of critical mass as a function of the reflector thickness. A number of 16-group, diffusion-theory calculations in spherical geometry were made in which the critical mass was determined as a function of reflector thickness. These results are shown by the solid line in Fig. 5. The corresponding experimental points are shown as the circles in the figure. The excess reactivity was converted to mass through the measured edge fuel worth.

Although estimates of edge fuel worth had been made previously, they were not consistent with later measurements. In fact, the worth of fuel at the top of the core¹ was notably different than that at the bottom. In order to resolve this discrepancy, at least in part, the worth of a column

¹ZPR-9 assemblies are built as cylinders with the axis horizontal. Here "top" and "bottom" refer to locations on the radial boundary.

of B^{10} (2 in. wide, 1/8 in. thick, and the full length of the core) was determined at four peripheral locations 90° apart. A 12% difference was noted between top and bottom worths, and a 10% asymmetry between right and left side worths. The right-left asymmetry can be accounted for by the geometry of the fuel plates within the drawers. A top-bottom asymmetry can only be ascribed to reflections from the table bed and from the floor. A further check was made by measuring the worth of the same column of B^{10} some 10 cm outside the Al_2O_3 reflector, both at the top and bottom of the core. Again an asymmetry was observed, with the worth at the bottom of the core 4 times that at the top. In order to isolate the assembly from its surroundings, a 10-cm-thick layer of aluminum metal was added to the outer periphery of the Al_2O_3 reflector. The critical mass of the isolated core is shown as the square data point in Fig. 5 at 57 cm.

Fig. 5. Critical Mass of ZPR-9 Assembly for Various Reflector Thicknesses

Although isolating the assembly with an additional 10 cm of aluminum metal produced symmetry in the boron worths on the four sides of the core, fuel worth at the bottom of the core remained about 10% higher than that at the top. For the purposes of reporting critical mass, an average value of 17.61 lh/kg was used as the edge fuel worth. The final assembly, which was considered reasonably well isolated, had a 47-cm-thick aluminum oxide reflector and a 10-cm-thick aluminum ring around the aluminum oxide. The critical composition of the isolated core is shown in Table VIII.

TABLE VIII. Critical Composition of ZPR-9
Assembly No. 7 Core

Core length (including central gap)	51.0 cm
Radius ^a	26.94 cm
Volume ^a	116.28 liters
Edge fuel worth	17.61 lh/kg of U ²³⁵
Critical mass ^a	204.51 kg U ²³⁵

Composition		
	Mass Density (g/cm ³)	Atom Density (10 ²⁴ atoms/cm ³)
U ²³⁵	1.759	0.00451
U ²³⁸	0.128	0.000323
Al	0.721	0.0161
W	6.527	0.0214
Fe	0.033	0.000355

^aCorrected for excess reactivity.

Central worth measurements were made for a large number of samples (see Table IX). Results for B¹⁰ were for fully enriched boron powder in the amorphous form; those for B(Cry) are for natural boron in the crystalline state. Excepting for U²³³, the errors are estimated to

TABLE IX. Central Reactivity Worths in ZPR-9 Assembly No. 7

Sample	Sample Weight, g	Measured Worth		Sample	Sample Weight, g	Measured Worth	
		lh	lh/kg			lh	lh/kg
U ²³³	291.92	85.02	291.2	Re	633.8	{ -59.54 -58.01	-92.78**
U ²³⁵	134.98	23.73	175.8	C	106.0	2.67	25.23
U ²³⁸	506.0	-1.35	-2.66	Au	1184.0	-23.66	-19.98
UO ₂	106.5	+0.52	+4.84	Be	1143.2	-0.265	-0.23
B ¹⁰	29.29	-72.03	-2460	Eu	324.93	-52.41	-161.25
B*	75.2	{ -37.46 -37.77	-500.2**	EuO	34.2	-2.41	-70.3
B ₄ C	59.6	-13.95	-234.1	Gd	397.93	-16.46	-41.36
W	1052.0	-11.60	-11.03	GdO	59.2	0.46	7.74
WO ₂	78.4	-0.66	-8.41	Y ₂ O ₃	81.5	2.95	36.20
				DyO	104.7	2.95	28.18
				H			3710

*Natural boron in crystalline form.

**Average of two measurements.

be ± 0.2 lh. In the case of the U^{233} sample, it was necessary to separate the tables and insert the sample into one of the fuel drawers. Day-to-day experience in the reproducibility of the critical rod position for a given configuration indicate that the error in this type of measurement is $\pm 1/2$ lh.

Central worth measurements of polyethylene samples ranging in weight from 5 to 60 g gave an average worth of 3.71 lh/g of hydrogen. No dependence on sample shape or weight was observed.

Unlike previously reported worths for boron hydrogen mixtures in the earlier assemblies, these same measurements in Assembly No. 7 are not at all consistent. Previous measurements, for example, gave the same results for compressed pellets as for mixed powders. In the present measurements, especially in terms of the boron enhancement factor, the pellets display a different behavior from that of the powders. For this reason, the data as taken are listed in Table X, and no conclusions are reached as to what fractional mixture would provide the best control material.

TABLE X. Central Worths of B-Lucite Mixtures in ZPR-9 Assembly No. 7

Form	Sample			Worth		Boron Enhancement Factor
	w/o Boron	w/o Lucite	Sample wt., g	Measured, lh	lh/g of Sample	
Block	0	100	72.97	25.777		
Powder	20	80	27.232	-1.150	-211.4	4.02
Pellet	25	75	19.275	-7.958	-530.5	0.98
Powder	40	60	31.307	-6.023	-481.1	2.10
Powder	50	50	28.893	-4.007	-276.3	0.93
Pellet	50	50	19.854	-5.88	-588	1.90
Powder	70	30	33.578	-10.69	-449.1	0.86
Pellet	75	25	19.939	-2.79	-558	3.96
Crystal Powder	100	0	75.2	-37.5	-500	

Measurements of the Rossi-alpha are in progress with Assembly No. 7. After this the worth of a boron ring in the reflector and completely surrounding the core will be determined.

3. ZPPR

Start of construction of ZPPR is contingent upon approval by the AEC. The Preliminary Safety Analysis Report was submitted to the Division of Reactor Licensing, AEC, in February 1965.

A contract for coring of the Gravel Gertie structure, as requested by the ACRS Subcommittee, was awarded to Eberline and Associates, Santa Fe, New Mexico. As of August 26, a 48-in.-dia core was driven into the gravel of the Gravel Gertie structure down to the cell floor. Preliminary analysis indicated an abundance of uranium dispersed in the lower levels of the gravel. Detailed analyses of the gravel samples obtained are in progress.

The following is a status report on reactor components:

a. Reactor Bed and Tables. The preliminary design drawings for the bed and tables have been received from Giddings and Lewis Machine Co., Fond du Lac, Wisc. Comments on these drawings have been sent to the vendor.

b. Matrix Drawers. The test tube assembly was completed by the vendor, Mechanical Products Mfg. Co., Seattle, Wash. The form dies for the $1\frac{1}{2}$ - and 2-in. channels and the perforating die were completed but not tried out. Tooling work on some smaller parts is also in progress.

c. Matrix Tubes. Materials for producing the four pilot tubes have been ordered. Necessary tooling and dies are being fabricated.

d. Poison Safety Rod Drives. Bids have been received on the poison safety rod drives. The procurement package was submitted to the Commission on August 11.

e. Reactor Knees. Bids for the reactor matrix knees have been received and are being reviewed.

D. General Fast Reactor Fuel Development

1. Irradiation of Uranium-Plutonium Alloys

a. CP-5 Irradiations. Five instrumented, temperature-controlled capsules containing uranium-plutonium fast reactor fuel alloys are presently being operated in the vertical thimbles of the CP-5 reactor.

The specimen loading and objectives of these experiments are as follows (listed by capsule No.):

CP-33. This capsule contains three specimens each of U-10 w/o Pu-10 w/o Fz and U-15 w/o Pu-10 w/o Fz alloy jacketed in Nb-1 w/o Zr tubing. The wall thickness of the jackets varies between 0.015 and 0.025 in. The design objective of the experiment is to determine the maximum achievable burnup without jacket failure. To date, three of the specimens have reached a burnup of 8.7 a/o without evidence of jacket failure. Also, none of the other three show evidence of failure.

CP-40. This capsule (not included in the above five) was removed from the reactor August 10 following temperature indications which suggested fuel release. The six specimens in the capsule had been designed to achieve a fuel burnup of 3.1 a/o before the calculated internal pressure would rupture the jacket. When the above burnup value had been reached without jacket failure, it was decided to continue irradiation until the

specimens were intentionally failed. The first evidence of jacket failure occurred when the specimens had attained 5 a/o burnup. Neutron radiographs have confirmed a deformation of four specimens and the capsule will therefore be destructively examined. The specimens were of U-15 w/o Pu-10 w/o Fs jacketed in V-20 w/o Ti tubing. The wall thickness of the tubing was 0.016 in. and the ID's varied from 0.157 to 0.169 in.

CP-41. In this capsule, six specimens are being irradiated which have preselected configurations calculated to result in jacket failures near 6 a/o burnup. The jacket material is V-20 w/o Ti tubing, having a wall thickness of 0.016 in. and ID dimensions between 0.157 and 0.165 in. The fuel pins are either U-10 w/o Pu-10 w/o Fz or U-15 w/o Pu-10 w/o Fz, of 0.144-in.-dia and 2.0 in. in length. The specimens are being irradiated at 610°C fuel temperature and have attained 4.0 a/o burnup without indication of failure.

CP-44. This capsule contains one fully enriched, 4.0-in.-long, U-15 w/o Pu-10 w/o Ti alloy fuel pin, jacketed in V-20 w/o Ti tubing. The ID of the tubing is 0.173 in. and the wall thickness 0.015 in. The specimen will be irradiated until evidence of jacket failure. The present irradiation temperatures are: 760°C for the central metal and 560°C maximum for the jacket. The specimen has been irradiated to a calculated 4.6 a/o burnup.

CP-45. This capsule contains one fully enriched, U-18.5 w/o Pu-14.1 w/o Zr pin jacketed in V-20 w/o Ti tubing. The fuel pin is 4.0-in. long. The jacket has an ID of 0.175 in. and a wall thickness of 0.015 in. The rod is being irradiated at relatively high central fuel temperatures of 820°C and jacket surface temperatures of 600°C. The present calculated attained burnup is 4.2 a/o without indication of failure.

CP-46. This capsule contains six specimens of vibratory compacted U-15 w/o Pu-10 w/o Fz metal powders. The mesh sizes of the powders are either -60 +120, -120 +200, or -200 +325 and are contained in Nb-1 w/o Zr tubing. The powders are impregnated with sodium to improve the heat-transfer characteristics of the compact. The inside dimension of the tubing is 0.156 in. and the wall thickness 0.009 in. One specimen is instrumented for direct measurement of central fuel temperature. The specimens have been irradiated at maximum fuel temperatures of 690°C for a calculated 1.2 a/o burnup.

The postirradiation examination will include fission-gas recovery and analysis. The resulting data will be compared with calculated results on the probability of fission-recoil escape given by the equation²

²White, D. W., Beard, A. P., and Willis, A. H., Irradiation Behavior of Dispersion Fuels, KAPL 1909 (1957).

$$\bar{f} = 3/4(2R/D) - 1/16(2R/D)^3,$$

where $\bar{f}$ is the probability of escape of recoils, R the range of fission fragments, and D the diameter of particle.

b. MTR Irradiations. Capsule ANL-55-7, containing three full-length EBR-II-type fuel rods was put into the MTR on February 2, 1964 and removed on July 6, 1965. The calculated burnup of the three fuel pins is 2.8 a/o. A gas annulus in the capsule was used for temperature control. The maximum surface temperature and the maximum fuel center temperature as calculated from thermocouple data were 555°C and 600°C, respectively.

In all three rods the fuel was U-15 w/o Pu-10 w/o Fs, the uranium being 7.7% enriched. All fuel pins had a diameter of 0.142 in.

On August 19, 1965 the capsule was opened and the three fuel rods were removed without difficulty. A preliminary examination revealed no evidence of any damage. All jackets are intact. The general appearance of the fuel rods corresponded very well with that before irradiation. Examination is proceeding in the Alpha-Gamma Hot Cell.

2. Corrosion of Fuel-cladding Materials

A new sodium sampler has been used which permits flushing of the sample tube with sodium from the bath prior to taking a sodium sample for oxygen analyses. First results using the new technique with a nickel sample tube were not encouraging. Sampling hot gettered sodium at 550°C gave values of 82 and 96 ppm oxygen, whereas sampling of the system at 155°C gave a value of 14 ppm.

3. Refractory Metal Alloys for Service in Oxygen-contaminated Sodium

Six vanadium-base alloys were corrosion tested for 7.0 days in 650°C sodium containing 45-50 ppm oxygen. Results are shown in Table XI.

TABLE XI. Corrosion of V-Ti Alloys

Alloy, w/o	External Loss, μ	Internal Penetration, μ	Total, μ	Alloy, w/o	External Loss, μ	Internal Penetration, μ	Total, μ
V-20 Ti Control	42	43-46	85-88	V-40 Ti Cold Worked	28	13-16	41-44
V-30 Ti Cold Worked	52	23-27	75-79	V-40 Ti Annealed	14	13-16	27-30
V-30 Ti Annealed	33	15-17	48-50	V-50 Ti Cold Worked	25	12-18	37-43

Cold Worked--As cold rolled.
Annealed--900°C--1 hr in vac.

The adverse effect of cold work on total metal penetration is quite evident. A range of heat treatments will be investigated to obtain the optimum corrosion resistance.

The high-velocity-pumped sodium loop has been delayed somewhat because of faulty welds but most of the difficulties have now been corrected.

E. General Fast Reactor Fuel Reprocessing Development

1. Skull Reclamation Process

The skull reclamation process is being developed for the specific application of recovering and purifying residual uranium left in crucibles after melt refining spent fuel pins from the EBR-II reactor. The chemistry and technology developed in this process will also be applicable to the processing of a variety of uranium-bearing fuels, such as metals, oxides, or carbides. Most of the pilot-plant work, which is now complete except for certain analytical data, has been carried out on a scale of 1.5 kg of uranium; the development of plant-scale (~4.25 kg uranium), remotely operated equipment is under way.

Seven pilot-plant runs were made by the modified flowsheet (see Progress Report for June 1965, ANL-7071, pp. 24-25). Chemical analyses of samples from four of the runs have shown that the noble metal extractions were identical with those of earlier runs. Good removals of ruthenium (about 80%) were accomplished; molybdenum removals continued to be somewhat poor. A program is currently under way to ascertain the reasons for the poor molybdenum decontamination. Increasing the settling time prior to solution transfer and decreasing the rate of solution transfer are possible, although not desirable, means of improving molybdenum removal.

The next step of the modified process, the reduction step, is accomplished by contacting the uranium oxide from the noble metal extraction step with a 60 w/o Mg-Zn solution. During this operation, the bulk of the uranium precipitates from solution; at the conclusion of this step, the salt phase and the Mg-Zn supernatant, which contain rare earths and zirconium, are transferred to waste. In one run at 800°C, uranium reduction was complete after 3/4 hr with an equilibrium amount of uranium (0.6% of the charge) remaining in the flux. In runs at 700 and 650°C, the reductions were slower; but, after 2 hr, only 1.1 and 1.6%, respectively, of the uranium remained in the flux. Increasing the agitation or the reaction time should decrease the uranium loss in the flux to less than 1% at the lower temperatures if the equilibrium values remain constant at the lower temperatures. Zirconium reduction was slower than that of uranium in these runs; some zirconium removal (about 1/3) was accomplished by the use of 60 w/o Mg-Zn solution.

Since the products have not been recovered by retorting, accurate analyses are not available. However, based on existing analyses, the approximate overall decontamination factors for representative fission elements in these pilot-plant runs were:

Cerium	14
Ruthenium	$7\frac{1}{2}$
Molybdenum	2
Zirconium	$2\frac{1}{2}$

Two additional plant-size skull-reclamation runs following the modified procedure have been completed in a furnace located in an inert-atmosphere enclosure. These runs are part of a testing program for the plant-scale equipment under simulated process plant conditions. All mechanical operations (charging, heating, mixing, sampling, and transferring) were accomplished without difficulty. Equipment for controlling the escape of fumes from the hot furnace during intermediate charging operations is being developed.

In these plant-scale runs, the transfer tube, which had been rebuilt, showed excellent performance. The new construction embodied a number of simplifications designed to increase the performance reliability of the transfer tube. Of these modifications, the principal change was the replacement of the seven heaters insulated with swaged magnesia, which had been used previously to heat the transfer tube, by three heaters insulated with braided silica. The wire used in the new heater cable is No. 14 braided Chromel P wire.

The use of silica-insulated Chromel P wire as a heater cable is unusual, and studies are under way to establish the suitability of the cable for extended use. In a preliminary test, a single length of the cable was wound around a section of stainless steel tubing to simulate the transfer-tube assembly. This assembly was heat-cycled in an argon atmosphere between room temperature and temperatures ranging from 750 to 975°C. After approximately 400 hr of operation at elevated temperatures, no evidence of deterioration of the cable was noted. During normal furnace operation the transfer line would be heated to about 750°C for 6 hr for each run.

2. Pyrochemical Processes

Compact pyrochemical processes are under development for processing fast breeder reactor fuels of the ceramic (e.g., oxide and carbide) or metallic types. Such processes utilize Cd-Mg, Cu-Mg, or Zn-Mg solvent alloys in contact with molten halide salts.

When CdCl_2 is used in combination with liquid cadmium in a process, formation of cadmium (I) chloride may be expected. It has been found that

the following reaction occurs in molten lithium chloride solvent at 650°C:

Assuming $[\text{Cd}_2[\text{CdCl}_4]] \ll ([\text{CdCl}_2] + [\text{LiCl}])$, the equilibrium constant

$$K = \frac{[\text{Cd}_2[\text{CdCl}_4]]}{[\text{CdCl}_2]^2} ([\text{CdCl}_2] + [\text{LiCl}])$$

determined from spectrophotometric measurements and chemical analysis was found to have an average value of 0.22 at 650°C.

Investigations are also being conducted to study the formation of the lower chlorides of zirconium and thorium in molten salt solutions. The lower chlorides appear to be formed when the salt solutions are contacted with liquid-metal alloys containing magnesium.

In connection with the use of zinc-magnesium alloys as process solvents, the solubility of plutonium in this system is of interest. Solubility data were obtained at 600, 700, and 800°C from analyses of filtered samples of liquid zinc-magnesium alloys saturated with plutonium. Although the liquidus lines and the phase diagram are still tentative, a liquid miscibility gap in the magnesium-rich composition region has been observed.

Engineering work related to the development of continuous molten metal-molten salt extraction processes (see Progress Report for July 1965, ANL-7082, p. 28) is under way. Initial experiments were designed to permit examination of metal-salt extraction kinetics for transient reactions in packed columns. In an experiment, a metal alloy of Cd-11 w/o Zn-5 w/o Mg containing uranium or cerium was passed through a packed quartz tube filled with static, molten salt (50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl). These transient tests have indicated that equilibrium stage heights of less than 2 ft can be expected in packed columns. The tests have also indicated the order of magnitude of typical mass-transfer rates and thus have demonstrated the feasibility of countercurrent extractions in the system. Future experiments will be performed in equipment that more closely simulates steady-state columns.

3. Materials Evaluation

a. Corrosion Resistance of Ferrous Alloys. One of the compact pyrochemical processes currently under development for processing fast breeder reactor fuels is based on the partition of uranium, plutonium, and fission products between a Cu-Mg alloy and different Zn-Mg alloys in mutual contact with a 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl salt.

In an effort to find an inexpensive, easily fabricated material of construction for this process, four ferrous alloys (304 SS, 405 SS, AISI 4130, and AISI 1020) were exposed to three copper-magnesium solutions

(50 w/o Cu-Mg, 70 w/o Cu-Mg, 90 w/o Cu-Mg) at 600 and 800°C for 200 hr in the presence of the halide salt. Little corrosion occurred in any combination at 600°C; at 800°C the corrosion increased significantly, the higher corrosion rates occurring with increased concentration of copper in the melt. However, even at 800°C corrosion is sufficiently low to warrant further testing.

b. Procurement of Tungsten Crucibles for Skull Reclamation Process. Two crucibles will be employed in the EBR-II skull reclamation process. The first crucible is fabricated of tungsten and is of relatively large size (15 in. in ID by 30 in. high). It will be used for the steps between oxidation of melt refining crucible skulls and retorting of uranium-magnesium-zinc solution for recovery of uranium product. The second crucible, which may be made of tungsten or beryllia (both are being considered), is of smaller size (7½ in. in ID by 17 in. high) and will be used in the retorting step. The tungsten crucibles used to date in the skull reclamation process have been made by pressing and sintering, and have given good performance. However, because the method of their fabrication results in crucibles with thick walls, the crucibles are heavy, unwieldy, and expensive.

A method of fabrication which should overcome the disadvantages of pressed-and-sintered tungsten crucibles is shear-forming. A crucible formed in this way should be light in weight, close to theoretical density, and less subject to failure by shock because of the lower brittle-to-ductile transition temperature.

Metallwerk Plansee, the Austrian affiliate of the National Research Corporation, has announced that they have succeeded, after at least 15 failures, in fabricating an 11-in.-OD by 18-in.-high tungsten crucible by shear-forming. They feel that the difficulties encountered were largely a result of developing the art, and that with their new equipment, developed for this job, they should be able to fabricate similar crucibles with little difficulty.

c. Tungsten-0.1 w/o Nickel Alloy as Crucible Material. It has been reported³ that the addition of 0.1 w/o nickel to pressed-and-sintered tungsten reduces the porosity of a finished tungsten shape. Such a modification in composition could be helpful in fabricating tungsten crucibles that are impervious to metal/salt solutions. Accordingly, a sample of pressed-and-sintered tungsten containing 0.1 w/o Ni was mounted as an agitator blade and exposed to Zn-47.5 m/o MgCl₂-47.5 m/o LiCl-5 m/o MgF₂ at 800°C for 200 hr. No evidence of deterioration of the sample was noted. More testing will be done before accepting this additive in our plant-scale crucibles.

³Oustalet, J. A., Oak Ridge National Laboratory, private communication.

II. GENERAL REACTOR TECHNOLOGY

A. Theoretical Reactor Physics

1. Development of Reactor Computing Codes

The space-dependent multigroup thermalization code THERMOS⁴ is being modified to facilitate coupling with the space-independent multi-group constants code MC² (see Progress Report for May 1965, ANL-7046, p. 39). The resulting coupled code will permit calculation of constants for both the thermal- and fast-energy ranges.

The validity of THERMOS as a "homogeneous" thermalization code is questionable for very large systems, e.g., if the buckling $B^2 < 10^{-4}$, because of the numerical approximations involved.

Various group cross sections were calculated by THERMOS for a BeO-U²³⁵ system with a moderator-fuel ratio of 100 and B^2 varying from 1.0 to 10^{-6} . The results for $B^2 > 10^{-4}$ differed by 3-8% from homogeneous thermalization code calculations by means of TEMPEST⁵ with the heavy-gas moderator model. These differences can be attributed in a large part to errors in the heavy-gas kernel as opposed to the more exact free-gas kernel used in THERMOS.

For values of $B^2 < 10^{-4}$ THERMOS calculations diverged for the BeO-U²³⁵ problem. For this reason a lower limit on B^2 , e.g., $B_{\min}^2 = 10^{-4}$, will be used when coupling MC² and THERMOS. This $B_{\min}^2$ will be chosen so that numerical approximations used in THERMOS will remain valid and yield consistent group constants.

2. Convergence Acceleration by Nonlinear Sequence Transformations

The value of a set of typical nonlinear sequence transformations, the Shanks e_k transformations, has been tested for estimating the eigenvalues of several iterative reactor problems. The examples included problems in 1-dimensional diffusion theory (REX), 1-dimensional transport theory (SNARG), and 2-dimensional transport theory (DDF). When the convergence of the initial sequence of eigenvalues was slow, these transformations extrapolated to a steady value with about half as many iterations. Even when the convergence of the original problem was relatively fast, the transformations improved convergence, although the improvement was not as dramatic.

⁴Toppel, B. J. and Baksys, I., The Argonne Revised THERMOS Code, ANL-7023, to be published.

⁵Shudde, R. H. and Dyer, J., TEMPEST-II, AMTD-111 (June 1962).

The e_k transformations were devised by Shanks⁶ to determine the constant B in sequence $\{T_n\}$ of the approximate form

$$T_n = B + \sum_{i=1}^k \alpha_i \lambda_i^n, \quad (1)$$

where the parameters α_i and λ_i may be either real or complex, and may be of any magnitude. For any particular k , the e_k transformation is successful even if $|\lambda_i| > 1$, so that the original sequence diverges. For computation, an Algol algorithm published some time ago⁷ was transcribed into a FORTRAN subroutine for the CDC 160A computer, and was applied to sequences of eigenvalues extracted from the output of standard codes. The transformed sequences usually required only about 50-60% as much data as the original sequences. The exception to this occurred when the latter converged so rapidly that the desired accuracy was attained before sufficient data had been gathered to apply the transformation.

Results for a slowly convergent problem are shown in Table XII. The accelerated values on each line were obtained only from the original eigenvalues on that line and the line above it. Thus, if, as appears likely, the eigenvalue was converging to 0.978 ± 0.001 , three-decimal accuracy in the eigenvalue is obtained by applying any of the acceleration procedures except e_1 to the results of the first 9 iterations, and the result of applying the e_1 transformation is only slightly worse. Even after 14 iterations, the original sequence is in error by 0.015.

TABLE XII. Acceleration of Eigenvalue Convergence for Two-dimensional Transport Problem (DDF)

Iter. No.	Eigenvalue	Results of Applying Acceleration Procedures to the Original Eigenvalue Sequence						
		e_1	e_1^2	e_2	e_1^3	e_3	e_1^4	e_2^2
0	0.0000000							
1	1.336236							
2	1.249784	1.255037						
3	1.186745	1.017014						
4	1.139592	0.9996321	0.9982628	0.9996607				
5	1.104051	0.9952702	0.9938089	0.9939847				
6	1.076884	0.9887485	1.008441	1.009763	0.9972234	1.009989		
7	1.055863	0.9839656	0.9708109	0.9732258	0.9979054	0.9637813		
8	1.039463	0.9812592	0.9777314	0.9781234	0.9766564	0.9792092	0.9972446	0.9982026
9	1.026603	0.9798855	0.9784698	0.9785703	0.9785580	0.9786604	0.9784018	0.9783691
10	1.016483	0.9791054	0.9780800	0.9781331	0.9782147	0.9783661	0.9782672	0.9782395
11	1.008500	0.9786787	0.9781636	0.9781819	0.9781488	0.9781781	0.9781332	0.9781970
12	1.002189	0.9783680	0.9775357	0.9775693	0.9780898	0.9781388	0.9775818	0.9784429
13	0.9971914	0.9781752	0.9778598	0.9778682	0.9777495	0.9778146	0.9781612	0.9468592
14	0.9932273	0.9780224	0.9774401	0.9774620	0.9776769	0.9777401	0.9776573	0.9775970

⁶Shanks, D., Nonlinear Transformations of Divergent and Slowly Convergent Sequences, J. Math. Phys. **34**, 1-42 (1955).

⁷Thacher, H. C., Jr., Algorithm 215, Shanks, Comm. ACM **6**, 662 (1963).

It can be seen from Eq. (1) that the e_1 transformation is equivalent to the Aitken δ^2 extrapolation, which is one of the devices used in the EXTERMINATOR code.⁸ Transformations of higher order, however, allow for the possibility that the sequence has two or more significant λ_i , without seriously reducing efficiency if only one is present; this is aside from the need for $2nk + 1$ elements of the sequence to apply the e_k^n transformation.

Aside from the examples described here, the Shanks transformations have been shown to be effective in accelerating or inducing convergence of a large variety of slowly convergent or divergent sequences, including partial sums of series, successive iterates in the solution of nonlinear equations, and so forth. The FORTRAN program may thus be applicable in a variety of problems. When it can be shown that successive elements of the sequence do indeed satisfy Eq. (1), success of the method is assured, except for the effect of roundoff error. If this is not the case, the success of the transformation should be confirmed by a final check of the accelerated value.

3. Effects of Randomness on Group Cross Sections

When isotopes have unresolved resonances, effective cross sections are usually calculated by assuming the statistical properties (average spacing, capture width, reduced neutron width, etc.) to be those of the resolved levels or, lacking such information, those estimated by nuclear theory. The computation of average cross sections for such isotopes may be in error when other isotopes with competing resonances are present.⁹ In addition, there may be fluctuations from the average behavior.

To estimate the possible size of these effects on group cross sections and on Doppler coefficients, effective group cross sections have been calculated for a number of energy groups for Pu^{239} in a typical $\text{UO}_2\text{-PuO}_2\text{-Fe-Na}$ mixture. The unresolved resonances were replaced by randomly generated "resolved" resonances, that is, in regions where there are no resolved resonances, parameters were generated by a random process such that they would have the average behavior usually ascribed to the unresolved region. This process is repeated a number of times to get an average cross section and its standard deviation.

Results to date show that the behavior of the Doppler coefficient for an infinite homogeneous medium is about as expected when the effects of competition are included: the Pu^{239} component is small and negative at

⁸Fowler, T. B., Tobias, M. L., and Vondy, D. R., EXTERMINATOR - A Multigroup Code for Solving Neutron Diffusion Equations in One and Two Dimensions, ORNL-TM-842 (Feb 1965).

⁹Codd, J., and Collins, P., Some Calculations Concerning the Influence of Resonance Overlapping on the Doppler Effect in a Dilute Fast Reactor, Proc. Conf. on Breeding, Economics, and Safety in Large Fast Power Reactors, ANL-6792, pp. 711-726 (1963).

low energy, gradually rises with energy to small positive values, then decreases again. The probable error, on the other hand, shows a relatively constant behavior with energy. Thus the probable error may be expected to be less dependent on reactor spectrum than the coefficient itself.

Future work will include application to a realistic problem using perturbation theory. Also, an attempt will be made to differentiate between effective cross-section fluctuations from competition between isotopes and those inherent in the random process.

4. Resonance Interference

One situation in which resonance interference effects should be noticeable is that when a fuel rod contains a mixture having both U^{238} and Th^{232} . Foell and Connolly¹⁰ have measured the resonance integrals of mixtures of $Th^{232}O_2$ and $U^{238}O_2$ with PbO_2 present as a diluent contained in aluminum cylinders, 5 in. long and one inch in diameter, in the Advanced Reactivity Measurement Facility at the National Reactor Testing Station. They compared the resonance integrals of the mixtures with the sum of the suitably weighted resonance integrals for the individual absorbers in a mixture with lead dioxide and found the resonance integrals of the mixtures to be 2 to 4% lower. This reduction in the resonance integral can be attributed to the effects of interference between the resonances of the two nuclides.

The RIFF RAFF code,¹¹ which uses space- and lethargy-dependent sources, was used to compute the resonance integrals of four UO_2 - ThO_2 - PbO_2 mixtures and the resonance integrals of the corresponding mixtures of UO_2 - PbO_2 and ThO_2 - PbO_2 for the energy range 1-1000 eV for a temperature of 300°K. Given in Table XIII are the experimental and the computed resonance integrals for the mixtures and their deviation ΔI from the sum of the weighted resonance integrals for the individual absorbers. The calculated values of ΔI are 85 to 90% of the measured values. When we consider that the calculations did not include the unresolved energy range

TABLE XIII. Resonance Integrals of $Th^{232}O_2$ - $U^{238}O_2$ - PbO_2 Mixtures

$(N_u + N_{th}) \times 10^{-24}$, atoms/cm ³	N_u/N_{th}	$N_{PbO_2} \times 10^{-24}$, molecules/cm ³	I_{exp} , b	ΔI_{exp} , b	I_{calc} , b	ΔI_{calc} , b
0.00524	3/1	0.00668	32.90	-0.91	27.69	-0.76
0.0121	3/1	0.0	23.70	-0.91	17.83	-0.79
0.00604	1/3	0.00644	30.45	-0.84	23.07	-0.76
0.00928	1/3	0.00286	24.82	-0.83	18.20	-0.72

¹⁰Foell, W. K., and Connolly, T. J., Measurement of Resonance Absorption in Thorium-232 Dioxide/Uranium-238 Dioxide Mixtures, Trans. Am. Nucl. Soc. 7, No. 1, 84 (1964).

¹¹Kier, P. H., RIFF-RAFF, A Program for Computation of Resonance Integrals in a Two-region Cell, ANL-7033 (to be published).

and the effects of the aluminum can, and that experimental error should introduce an uncertainty of at least 10% in the experimental ΔI , the agreement between calculation and experiment appears to be good.

5. Numerical Evaluation of Fourier Integrals

In many applications, one must evaluate Fourier transforms of the form

$$f(\omega) = \int_0^{\omega} f(t) \cos(\omega t) dt. \quad (1)$$

It should be noted that customary quadrature techniques, such as the Laguerre-Gauss or the Hermite-Gauss method,¹² cannot be applied to Fourier integrals. Due to the oscillatory term in the integrand [i.e., $\cos(\omega t)$], the interpolation formulas used in these methods do not provide a close fit for the behavior of the integrand. Consequently, different techniques must be employed in the efficient evaluation of Fourier integrals.

In the present approach, the range of integration is partitioned into two regions: $0 \leq t \leq t_m$ and $t_m \leq t < \infty$, the latter designated as the asymptotic region. In terms of this partitioning, Eq. (1) reduces to

$$f(\omega) = \int_0^{t_m} f(t) \cos(\omega t) dt + J(\omega, t_m), \quad (2)$$

where the expression $J(\omega, t_m)$ represents the asymptotic contribution. The appropriate range of values for t_m depends only upon the behavior of $f(t)$. If t_m is chosen large enough, then a simple asymptotic form of $f(t)$ is valid for all $t > t_m$. Thus

$$f(t) \sim f_a(t), \quad t \geq t_m. \quad (3)$$

Consequently, the approximate asymptotic contribution reduces to

$$J_a(\omega, t_m) = \int_{t_m}^{\infty} f_a(t) \cos(\omega t) dt. \quad (4)$$

The expression given above may also be evaluated in an asymptotic series, since t_m has been chosen sufficiently large. Hence, the contribution used in Eq. (2) is the asymptotic form of the integral given in Eq. (4). This expansion can usually be found utilizing integration by parts. One can retain as many terms as is desired.

¹²Hildebrand, F. B., Introduction to Numerical Analysis, McGraw-Hill Book Co., New York (1956).

There remains the evaluation of the integral in the region $0 \leq t \leq t_m$ in Eq. (2). This term can be determined accurately if $f(t)$ is a slowly varying function. Let $\{t_i\}$, $i = 1, 2 \dots m$, by a proper net (subdivision) on the interval $[0, t_m]$; then the contribution to the integral which arises between t_i and t_{i+1} is

$$I_i = \int_{t_i}^{t_{i+1}} f(t) \cos(\omega t) dt. \quad (5)$$

Applying the mean value theorem to Eq. (5), one has

$$I_i = f(\bar{t}) \int_{t_i}^{t_{i+1}} \cos(\omega t) dt = \frac{f(\bar{t})}{\omega} [\sin(\omega t_{i+1}) - \sin(\omega t_i)], \quad (6)$$

where $\bar{t}$ is some value of t in the interval $t_i \leq t \leq t_{i+1}$. To ensure the applicability of the mean value theorem, the function $\cos(\omega t)$ must not change sign in the interval $t_i \leq t \leq t_{i+1}$. This can be accomplished by including the zeros of $\cos(\omega t)$ as a subset of the set of net points. Moreover, for slowly varying $f(t)$, one can use

$$f(\bar{t}) \approx \frac{f(t_{i+1}) + f(t_i)}{2}. \quad (7)$$

By means of Eqs. (6) and (7), the Fourier transform becomes

$$f(\omega) = \frac{1}{2\omega} \sum_{i=1}^{m-1} [f(t_{i+1}) + f(t_i)] [\sin(\omega t_{i+1}) - \sin(\omega t_i)] + J_a(\omega, t_m). \quad (8)$$

This form is easily adapted for computer evaluation.

The accuracy of this method can be estimated by utilizing the fact that the Fourier transform $f(\omega)$ should be independent of the choice of t_m (provided t_m is large enough to ensure that the asymptotic expansions are valid). By employing a number of different values of t_m and examining the change which occurs, the accuracy of this method can be ascertained for any specific case of interest.

The method has been employed to evaluate the following Fourier integral, which arises for a special case of pileup:¹³

$$f(\omega) = \frac{2e^{-\alpha}}{\pi} \int_0^\infty \cos[(\omega - 1)t] e^{-\beta^2 t^2} \exp[\alpha e^{-\beta^2 t^2} \cos t] \cos[\alpha e^{-\beta^2 t^2} \sin t] dt. \quad (9)$$

¹³Goed, R., Theoretical Estimates of Pile-up Distortion, ANL-6949 (March 1965).

Here, one has

$$f(t) = \frac{2e^{-\alpha}}{\pi} e^{-\beta^2 t^2} \exp [\alpha e^{-\beta^2 t^2} \cos t] \cos [\alpha e^{-\beta^2 t^2} \sin t], \quad (10)$$

where α and β are parameters.

The asymptotic contribution is, to third order,

$$J_a(\omega, t_m) = \left(\frac{2e^{-\alpha}}{\pi} \right) e^{-\beta^2 t_m^2} \left\{ \frac{\cos [(\omega - 1)t_m]}{2\beta^2 t_m} - \frac{(\omega - 1) \sin [(\omega - 1)t_m]}{4\beta^4 t_m^2} - \frac{\cos (\omega - 1)t_m}{4\beta^4 t_m^3} \right\}. \quad (11)$$

The accuracy obtained for this Fourier transform was better than 1-2% of $f(\omega)$ for values of $f(\omega)$ as low as 10^{-7} . A graph of the Fourier transform, $f(\omega)$, for the parameters $\alpha = 0.01$ and $\beta = 0.06$, is given in Fig. 6.

Fig. 6

The Fourier Transform of

$$f(t) = \frac{2e^{-\alpha}}{\pi} e^{-\beta^2 t^2} \exp [\alpha e^{-\beta^2 t^2} \cos t] \cos [\alpha e^{-\beta^2 t^2} \sin t] \text{ for } \alpha = 0.01 \text{ and } \beta = 0.06 \text{ in the region } \omega \geq 1.6$$

6. General Shield Analysis

The bulk shielding code "MAC"¹⁴ was originally written for a slab core with slab shield regions. The code is being modified to incorporate options of spherical and cylindrical geometry for the neutron calculations. Gamma-ray results will still be calculated by means of slab geometry.

The code, compiled for use with the CDC-3600 machine, provides a method for calculating radiation distributions in preliminary shield-design studies. Fluxes and dose rates are determined in one computer problem calculation for 31 neutron energy groups and 7 gamma-ray energy groups.

Total dose rates for a MAC calculation compared favorably with the values secured by using a combination energy-independent removal plus diffusion method.

However, at deep penetrations (greater than about one foot into the outside concrete region) the combination method gave much higher values. In studying radiation distributions in deep penetrations, other calculations can be made to check the MAC results.

¹⁴Peterson, E. G., MAC - A Bulk Shielding Code, HW-73381 (April 1962).

B. High-temperature Materials

1. Ceramic Fuel Materials

a. Mechanical Properties of Uranium Compounds. Further strain rate data for uranium sulfide (see Progress Report for June 1965, ANL-7071, pp. 33-4) were obtained with freshly prepared material having a density of about 88% of theoretical, at a medium strain rate. Fracture tests with these specimens gave results shown in Table XIV.

TABLE XIV. Mechanical Test Data for Fast Strain Rate of Uranium Sulfide

Number of Samples	Density, g/cc		Maximum Stress, kg/cm ²		Mean Load	Mean Strain	Mean Total	Temp, °C
	Mean	Standard Deviation	Mean	Standard Deviation	Rate,	Rate,	Strain,	
					g/sec	mm/sec x 10 ⁴	mm	
Higher-density Specimens--Fast Strain Rate								
3	10.28	0.005	567	19.5	1275.7	55.4	0.15	25
3	10.26	0.005	720	64.0	758.2	33.3	0.20	1000
4	10.27	0.04	1457	126.8	95.3	24.1	2.54	1250
4	10.36	0.01	626	16.9	49.9	28.8	3.04	1500
2	10.435	0.015	113	23.2	86.3	15.3	0.13	1615
2	10.335	0.005	68	13.6	-	-	(0.13)	1750
Lower-density Specimens--Medium Strain Rate								
3	9.59	0.02	515	25.1	322.3	16.3	0.20	25
3	9.54	0.01	804	74.4	145.3	8.9	0.37	1000
3	9.61	0.04	730	28.3	23.2	10.7	2.54	1250
3	9.55	0.02	383	43.9	14.1	12.0	2.85	1500
2	9.64	0.02	222	7.7	10.7	11.3	1.70	1600
2	9.70	0.06	72	7.2	-	-	(0.13)	1750

At the fast strain rate the strength increased up to 1250°C and then decreased. At the medium strain rate the maximum was at 1000°C, both in previously tested specimens and in these low-density specimens. The mean load rate at the fast strain rate was about four times the value obtained at the medium strain rate. The mean strain rate, likewise, was greater by a factor of about three.

The ductility increased slightly to 1000°C, in both cases, then very rapidly to 1250°C, at which the material was highly plastic. The specimens were easily strained to the limit of the equipment without cracking (2.54 mm). At 1600°C the fast strain rate fractured the specimens at low load rates and strain rates with very little deformation. The material cracked by intergranular separation and pulled apart. At 1750°C there was no measurable load or deformation, the material merely separated as rapidly as the load was applied. Based on these tests uranium sulfide has essentially zero strength above 1600°C, especially low-density specimens or when tested at the fast strain rate.

b. Anelasticity of Some Uranium Compounds. A probable cause of the large decrease of the Young's modulus in the first stages of oxidation

TABLE XV. Effect of Oxidation and Reduction on the Young's Modulus of Uranium Oxides

Ratio of Oxygen to Uranium	Young's Modulus	
	$E_f^{(a)}$	$E_e^{(b)}$
	kilobars ^(c)	kilobars
2.005	2069	2136
2.02	1898	1971
2.05	1417	1474
2.06	1221	1274
2.07	1149	1195
2.005(d)	2005	2072

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

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

(c) One kilobar is equal to 10^9 dynes/cm².

(d) Specimen was reduced in hydrogen from an O/U of 2.07 to an O/U of 2.005.

of uranium oxides might have been due, in a large degree, to the cracks originated by the stresses developed by the diffusion of oxygen into the body (see Progress Report for July 1965, ANL-7082, p. 35). In order to prove whether that or the excess of oxygen itself is the reason of this change in the elastic modulus, one sample was reduced in hydrogen. The oxidation and reduction history of the specimen is shown in Table XV. A metallographic examination was performed after the reduction. As expected, the cracks observed when there was an excess of oxygen remained, but there was no evidence of the second phase detected in the oxidized sample. These observations and the results obtained for the Young's modulus seems to indicate that the decrease of the elastic modulus is due mainly to the excess of oxygen itself and not to the cracks.

c. Uranium-mixed-anion Systems. The liquidus temperatures for compositions in the system UP-US have been determined under two atmospheres of helium (see Table XVI). The melting point of US corresponds closely to that found by other investigators. The melting point of UP is somewhat lower than the value (2610°C) found for material sealed in a tungsten crucible. Undoubtedly, some dissociation of UP occurred which resulted in formation of metallic uranium and somewhat depressed the overall melting temperature. The maximum melting temperature occurred at the composition 70 UP-30 US. The data show that incorporation of only 10% of UP in US has a marked effect on the liquidus temperature.

TABLE XVI. Melting Points of UP-US Mixtures

Composition, %	Liquidus Temp, °C	Composition, %	Liquidus Temp, °C
100 UP	2575	40 UP-60 US	2638
90 UP-10 US	2634	30 UP-70 US	2625
80 UP-20 US	2656	20 UP-80 US	2598
70 UP-30 US	2666	10 UP-90 US	2560
60 UP-40 US	2658	100 US	2465
50 UP-50 US	2654		

d. Development of Thermal Diffusivity Apparatus. The thermal diffusivity characteristics of a pressed and sintered specimen of plutonium

Fig. 7. Thermal Diffusivity of Plutonium Monosulfide

monosulfide were investigated as a function of temperature. After firing, the 12.5-mm-diameter disc was lapped to 3-mm thickness. The first set of values, designated as Run 1, are shown in Fig. 7. After the specimen was lapped down to 2 mm, Run 2 was carried out. A confidence band 12% wide was obtained near room temperature; at 600°C this band width increased to 20%.

2. Liquid-metal Corrosion

a. Dissolution Kinetics. In conjunction with dissolution experiments in the Ta-Sn system, measurements of the self-diffusion coefficient of Sn^{113} and Ta^{182} in liquid tin are in progress. The self-diffusion coefficient of tin has been determined by the capillary-reservoir technique over the temperature range 250-900°C. The low-temperature data are in good agreement with the results obtained by Careri¹⁵ and Ma.¹⁶ The considerable deviation from linearity on a $\log D$ versus $1/T$ plot reported by Ma at the higher temperatures was not observed in this work. This point will be further confirmed by increasing the temperature range another 200°C upon completion of the tantalum solute diffusion measurements.

b. Lithium Corrosion Studies at Elevated Temperatures. The three-heat-zone assembly and power supply for determining lithium corrosion of refractory metals and alloys were delivered and installed in the ultrahigh vacuum chamber. As a result of bake-out, leaking developed in two of the three heat zones. The equipment has been returned to the vendor for repair.

3. Irradiation Testing

a. Hydrogen Embrittlement in Irradiated Steels. Pressure vessel steels are tough and ductile, and in general their yield strength is too low for hydrogen-induced delayed failure to be observed. Pressure vessel steel A212B will be irradiated and embrittled by neutrons, then hydrogenated and tested under static load conditions. If hydrogen embrittlement occurs, catastrophic failure will be observed in a matter of hours under stresses less than half the nominal yield stress.

¹⁵Careri, G., Paoletti, A., and Vicentini, M., *Nuovo Cimento, Series X*, **10**, 1088-99 (1958).

¹⁶Ma, C. H., and Swalin, R. A., *J. Chem. Phys.* **36**, 3014-18 (1962).

A stainless steel capsule containing twenty-one notched stress-rupture specimens of A212 B, seven similar specimens of hardened 4340 steel, and two shaped tensile specimens of 4340 was inserted into a vertical thimble of the CP-5 reactor on August 11, 1965. It will receive about one month's irradiation, which should be enough to produce shifts in the Charpy transition temperature of up to 170°C. No Charpy specimens are included, but iron and nickel flux wires will provide enough data to compute the exposure and compare it with published data.

Hardened 4340 steel, unlike A212 B, is susceptible to hydrogen embrittlement. We will hydrogenate the irradiated steels electrolytically and determine the effect of the neutron bombardment on their behavior under static load.

A vacuum analysis system has been built to measure the hydrogen content of electrolytically charged specimens. The system is now complete and ready for testing.

C. Other Reactor Fuels and Materials Development

1. Nondestructive Testing

a. Determination of Elastic Moduli at High Temperatures by Ultrasonics. The low-temperature (that is, from room temperature to 900°C) measurements with a tungsten-rod sample have been completed. First-order calculations (i.e., uncorrected for thermal expansion) of longitudinal wave velocity, transverse wave velocity, Young's modulus, shear modulus, and Poisson's ratio have been calculated to 900°C. As soon as it is possible to use the Brew furnace, measurements at higher temperatures will be made.

It has been suggested by others that the elastic moduli of γ -uranium should be determined to help clarify some results of the measurement of the diffusion coefficient in this material. With this purpose in mind, a threaded bar of uranium was obtained and mounted into the vacuum furnace in the usual way. Pt + Pt-10% Rh thermocouples were used. From room temperature to about 550°C all echoes and trailing pulses were visible; at 600°C the trailing pulses disappeared. At the $\alpha \rightarrow \beta$ transformation temperature and all the way up to the $\beta \rightarrow \gamma$ temperature the echoes from the shoulder and end of the bar were visible, but no trailing pulses were seen. After passing through the $\beta \rightarrow \gamma$ transformation, no more echoes of any kind were seen.

After cooling the rod to room temperature, still no echoes were visible. The grains in the heated end of the rod evidently grew so that they were of the same order of magnitude of the wavelength of ultrasound in the rod, which was ~ 0.7 mm. Thus no data on the moduli of uranium at high temperatures were obtained.

b. Ultrasonic Instrument and Transducer Development. The investigation to determine the feasibility of sintered stainless steel products as backing members in ultrasonic transducer probes has continued. Stainless steel discs with various porosities and particle sizes have been received from the vendor. Measurements of density and longitudinal wave velocity are being taken of each sample. The resultant acoustic impedance values (product of the density ρ and velocity c) can then be compared to that of lead zirconate titanate which has been used predominantly as the piezoelectric material in ANL-fabricated probes. Ideally, the acoustic impedances of the piezoelectric material and the backing member should be equal.

Work on the Schlieren systems which will be used to evaluate ultrasonic transducer probes has continued. Electronic circuitry for the pulsed system is nearly complete. An oscillator for the continuous wave system has been ordered.

c. Development of a Neutron-image Intensification System. The neutron-image-intensifier equipment has been returned to Juggernaut reactor upon completion of the radioactive neutron source tests at the Chemistry Division Hot Laboratory. Observation tests of the output phosphor of the intensifier tube employing dark adapted observers have been completed. Although measurements of output phosphor-screen brightness were not obtained, visual indications were that the output phosphor brightness continues to drop in a regular fashion for decreasing neutron intensity striking the tube, for neutron intensities less than the previously measured value of 10^4 n/cm²-sec. Reliable visual indications of thermal-neutron intensities as low as 150 n/cm²-sec were obtained. With that neutron intensity, a visual decrease in output phosphor brightness could be readily observed when a 2.5-cm-thick Masonite absorber was placed in front of the tube. This would indicate that the tube response at that point was actually caused by neutrons in the beam. Some visual response at the output phosphor was observed for an incoming thermal-neutron intensity as low as 30 n/cm²-sec, but, at that low level, much of the response observed could have been caused by gamma radiation in the beam.

An analysis of these data would indicate that the plot of the linear extension of the intensifier tube output phosphor brightness versus neutron intensity is reasonably accurate.¹⁷ That extension would yield an output phosphor screen brightness of about 10^{-4} ft-Lambert for a thermal-neutron intensity of 150 n/cm²-sec. That brightness is at the lower end of the brightness range normally used in medical X-ray fluoroscopy and would be expected to be detected by dark-adapted observers.

d. Infrared Systems. A thermal testing system utilizing a thermistor as the thermal detector was constructed. The system is a near-surface direct-current thermal method that is not significantly affected by emissivity changes. A Victory Engineering Corporation 41A401C bead thermistor was used. The circuit in the system is a simple dc bridge. The amplified signal from the bridge is applied to a strip chart recorder, providing a permanent record of the inspection. To evaluate the system, test samples of flat stainless steel plate with discontinuities machined in the back (unscanned) surface are now being fabricated.

Work on the infrared heat source has continued. A lens, 82 mm in diameter and having an 89-mm focal length, was installed. The projected size of the filament is approximately 1 x 1 cm. The aberrations in the optical system produce considerable scattering of the light around the image of the filament on the test sample. Efforts to reduce these aberrations are now under way.

¹⁷Berger, H., Niklas, W. F., and Schmidt, A., An Operational Thermal Neutron Image Intensifier, J. Appl. Phys. 36, 2093 (1965).

e. Noise Reduction in Electromagnetic Systems. The problem of noise in general is the chief factor limiting the application of electromagnetic test systems to many problems in nondestructive testing which a superficial consideration would indicate could best be solved by an electromagnetic method. In broadband test systems, noise generated within the test system itself can be a limitation on its useful sensitivity because the noise power available from many sources is proportional to the system bandwidth. Work in this program has so far been concentrated on a study of this type of noise rather than noise originating in the test specimen.

Noise generated in the test system preamplifier has been an important contributor to the overall system signal-to-noise ratio (see Progress Report for June 1965, ANL-7071, pp. 39-41). The performance of these preamps has been improved to a point at which their noise contribution is now less than the noise generated in the pickup due to environmental electrical disturbances. Two preamplifiers, one using type 7586 Nuvisitors, and the other using type 2N3822 N-channel field-effect transistors, have been designed and constructed. These provide substantially lower noise performance than the low-noise type 2N2613 transistor preamps used up to this time. In addition, the new circuits provide considerably lower input capacitance, which is an advantage since it raises the resonant frequency of the parallel resonant circuit formed by the pickup inductance and the preamplifier input capacitance to beyond the amplifier cutoff frequency. This permits the preamplifiers to provide a more faithful representation of the time derivative of the magnetic field outside the apertures.

An analysis of the effect of a pulsed plane wave impinging upon a plane conductor has been undertaken in an attempt to obtain a means for predicting the optimum pulse lengths for particular problems in pulsed electromagnetic testing. An idealized defect consisting of a certain reduction in metal thickness was assumed. The nominal depth of penetration was defined for the square pulses used in the analysis. As expected, the difference between the incident wave and the reflected wave at the metal surface is much different for the idealized plane-wave case and the relatively localized curved field provided by the mask-aperture assemblies. However, a comparison between the calculated field transmitted through a given thickness of metal and some experimental results shows that the analysis may provide useful information about optimum pulse lengths despite the disparity between the idealized and actual reflections at the metal surface.

f. Thermal Conductivity of Irradiated Fuel versus 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 will thereby 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.

One trial run with several standard conductivity specimens and with five uranium-fissium alloys has been completed in the cave. Values of the diffusivity and conductivity measured during this run are in agreement with those reported previously (see Progress Report for May 1965, ANL-7046, Table XVII, p. 53). This run also indicated that several changes are necessary in the physical setup to facilitate operations. These changes are being incorporated into the system, and another trial run will be made before the cave is closed and irradiated material introduced into it.

D. Engineering Development

1. Two-phase Flow

a. Void Fraction--Pressure-drop Facility. Repairs to the electromagnetic pump and pressure-control system are complete. The system is now back in operation and two boiling runs have been obtained in the stable regime above 8 psia. Previous void-fraction data will be checked before terminating the experiment.

2. 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 and a pressure of approximately 8 atm. 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.

The loop was received at ANL on July 26 after shipment from Pratt & Whitney (CANEL). Careful inspection and a helium leak test showed no damage or leaks; inspection of the loop welds by X rays is underway.

Construction of the storage and assembly room for the loop is complete. The loop-support structure is being assembled in this room, and a plywood form is being constructed for handling of the Nb-1% Zr loop during its attachment to the support structure.

Instrumentation assembly is underway. The chromel-alumel, Nb-sheathed thermocouple assemblies will be received by the end of August. Vendor fabrication of the W-W 3% Re assemblies will be started in early September. The pressure transducers are being calibrated in

conjunction with the data-acquisition system recently received from Hewlett-Packard. The flowmeter-cooling system has been completed and tested; it will remove 3400 Btu/hr at a maximum NaK temperature of 600°F. A General Electric partial pressure analyzer has been received and will be installed in the vacuum chamber.

The vacuum-chamber facility is essentially complete; a comprehensive operating and maintenance manual has been prepared and distributed to personnel who are potential users of the facility.

b. Heater Experiments

(i) Electron Bombardment Heater Experiment. The electron bombardment sodium pool boiler has operated satisfactorily; the new power leads have proven adequate. Replacement of a defective Al_2O_3 insulator has caused a slight delay. Higher-purity Al_2O_3 insulators have been ordered for replacements in case future breakdowns occur. Since this experiment will determine the design basis for the large Nb-1% Zr electron boiler, the final design is still pending results of the smaller test.

3. Slave Television for Visual Feedback in Manipulator Systems

Recent experiments have shown that the usefulness of TV for the visual link in master-slave manipulator systems can be greatly increased if the camera and monitor are slaved to follow the "pan and tilt" motions of the manipulator operator's head.

It is well-known that a specific TV system provides a limited number of elements to form the picture. The number of elements is the same whether the camera is equipped with a wide-angle or narrow-angle lens. To get reasonably good angular resolution it is necessary to limit the angle of view of the TV camera. The real advantage of head-controlled TV is that this restricted angle of view can be quickly and naturally moved about to view a much greater area. For the experiments, camera viewing angles from 15 to 30° were used, but manipulation tests are carried out throughout an angle of more than 90°.

Figure 8 illustrates the system used in experiments with head-controlled TV in conjunction with a pair of Electric Master-Slave Manipulators, ANL Model E2. The TV monitor was supported by a mechanical arm and linkages arranged to keep the monitor facing the operator. The 14-in. monitor used was spaced about 23 in. from the operator's eyes. The pan and tilt motions of the monitor system were driven by nearly conventional servos. The operator was free to move his head in any direction or angle, but only the "pan and tilt" head motions controlled movements of the monitor and camera in their corresponding pan and tilt motions.

The camera was mounted near the manipulator slave arms (about 40 in. away) and was driven by conventional servos which caused it to rotate about the two intersecting axes shown. A simple, lightweight headpiece was connected to mechanical linkages which in turn were referenced to "ground." Small synchros mounted in the linkages provided command signals for the pan and tilt motions. The headpiece and attendant linkages had low friction and inertia; consequently the operator felt no restraining forces on his head.

Fig. 8. An Experimental Head-controlled TV System for Use with Electric Master-Slave Manipulators

When the camera and monitor were made to follow closely and accurately the "pan and tilt" motions of the operator's head, there was a surprising amount of annoying camera and monitor motion due to the inadvertent motions of the operator's head while carrying out manipulations. Considerable improvement of the system was achieved by providing a dead zone of between 7 and 12° in which the operator could move his head without causing the monitor or camera to move. We think that the inadvertent motions are annoying when there is no dead zone because there is some blurring of the picture when the camera is moving in pan or tilt faster than about 0.05 radian/sec. Another source of annoyance to the operator was the constant motion of the monitor.

Although the area viewed by the camera at any one time is much smaller than that covered by the manipulators, operators can easily carry out manipulations over an angle of 90° or more. They also remember the position of objects with respect to each other. We believe that this is because the camera, the monitor, and the operator's head all have to go through the same general angular displacement in gaining a new view.

Persons experienced in manipulator operation can perform tasks quite satisfactorily with this new viewing system after a learning period of 30 to 90 min. With this amount of practice, they can operate the system in a fairly natural way with only moderate attention given to changing the direction of view. Of course, we expect considerable improvement in proficiency with greater experience.

Experiments were also made using conventional switch-controlled TV. The same camera, monitor, and manipulators were used in making these tests. The pan and tilt motions of the camera were controlled by switches. The monitor remained in one position. With this system, it was much harder to gain new views quickly. Consequently, the overall rate of performing manipulations was considerably slower.

These experiments are only a step in improving TV for use in master-slave manipulator systems. Near-future experiments will be made with systems which provide head control in 2 angular and 3 translational motions.

E. Chemical Separations

1. Fluidization and Volatility Separations Processes

a. Recovery of Uranium from Low-enrichment Fuels

(i) Laboratory Support Work. The reactions of bromine pentafluoride (BrF_5) with various uranium compounds and mixtures of uranium and plutonium compounds are being studied to obtain data relevant to the possible use of BrF_5 as a fluorinating agent in a modified fluid-bed fluoride volatility process. In this conceptual process, the uranium in uranium-plutonium-bearing fuel, after decladding, is fluorinated by BrF_5 to UF_6 , which is volatilized from the reaction vessel and collected in a condenser along with excess BrF_5 , the reaction product BrF_3 , and volatile fission product fluorides. During the conversion of uranium to UF_6 , plutonium reacts to form nonvolatile PuF_4 and remains in the reaction vessel. The UF_6 , the interhalogens, and volatile fission product fluorides are separated by fractional distillation, resulting in the production of decontaminated uranium hexafluoride. The BrF_3 in the mixture of bromine fluorides is refluorinated to BrF_5 and the BrF_5 recycled. The fission products are disposed of as waste.

In a second fluorination step, the plutonium compound PuF_4 , which remains in the reaction vessel, is fluorinated with fluorine gas to PuF_6 . The PuF_6 is then volatilized from the vessel and collected in separate cold traps. After collection, the PuF_6 is revaporized and passed into a heated vessel, where it is decomposed thermally to nonvolatile PuF_4 product.

The capability of BrF_5 to act as a selective fluorination agent was demonstrated previously (see Progress Report for July 1965, ANL-7082, p. 43). Continuing this work, a determination was made of the effect of BrF_5 partial pressure on the rate constant for the reaction of BrF_5 with U_3O_8 at 250°C . These experiments were performed to evaluate the feasibility of using a lower temperature (250°C) for the fluorination of uranium by BrF_5 . Bromine pentafluoride concentrations in the gas phase of 26, 46, and 66 v/o (remainder nitrogen) corresponded to partial pressures (total pressure of 760 mm) of 198, 350, and 502 mm Hg, respectively. These experiments resulted in values of the rate constants of 0.0051, 0.0131, and 0.0235 min^{-1} at 26, 46, and 66 v/o BrF_5 , respectively. The values of the rate constants obtained with 46 and 66 v/o BrF_5 approached the values obtained with 26 v/o BrF_5 at 300 and 350°C , respectively. Further, extrapolation of the data shows that to obtain a value of the rate constant equal to that obtained at 400°C using 26 v/o BrF_5 (0.0395 min^{-1}) would require greater than 86 v/o BrF_5 at 250°C . Additional work investigating the effect of BrF_5 concentration on rate constants is under way.

(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 and differential thermal analysis. Melting points were determined for UF_6 - PuF_6 mixtures containing 0.132, 0.244, 0.376, 0.388, 0.623, 0.677, 0.714, 0.751, 0.795, and 0.946 mole fraction UF_6 . The melting points were found to be between the melting points of the two pure compounds, thereby indicating that no eutectic is formed. Based on this observation and the shape of the thermal analysis curves, it is concluded that the solid phases involved in solid-liquid phase equilibria of UF_6 - PuF_6 mixtures are solid solutions. Melting-point data for mixtures containing 0.4 to 0.6 mole fraction UF_6 are to be obtained.

(iii) Engineering-scale Alpha Facility. An engineering-scale alpha facility has been installed to permit demonstration of the major steps of the fluid-bed fluoride volatility process for the recovery of uranium and plutonium from ceramic fuels. Equipment units for fluorinating batches of UO_2 - PuO_2 pellets to hexafluorides and for demonstrating the conversion of hexafluorides to oxides have been installed. The hexafluoride-to-oxide converter will also be used in studies on the separation of plutonium as the tetrafluoride from UF_6 - PuF_6 mixtures by thermal decomposition.

Preparations continued in the alpha facility toward the initiation of the experimental program with plutonium materials. The 102 windows in the two alpha-boxes were sealed, and the boxes were leak-tested under several inches of positive water pressure. The smaller of the two boxes has a leakage rate of 0.07%/hr of box volume (volume, ~500 cu ft). Leak-testing of the larger box (volume, ~1600 cu ft) is near completion. The normal air flow through these boxes will be 150 and 600 cfm, respectively. The fluorinator and the converter (the latter to be operated as a fluid-bed thermal decomposer) are undergoing a final shakedown run with uranium. Plutonium is to be introduced in the succeeding run after a final safety review of the facility.

Installation work on the distillation unit, which has been erected in a mock-up area for preliminary testing, was continued. The distillation unit will be used to study purification of UF_6 and separation of UF_6 from PuF_6 . Work has progressed to the point where the heated enclosure, the final major mechanical item, can be installed. Actual distillation tests await completion of electrical, heat, and flow tests.

(iv) Decladding and Fluorination. Studies of alternative procedures for processing UO_2 pellets were continued in a 3-in.-dia fluid-bed system. Oxidative pulverization followed by fluorination is being tested. A current objective of these studies is to determine whether samples taken after pulverization are representative of the charge and thereby offer a solution to the overall problem of plutonium accountability.

Uranium dioxide pellets of two enrichments were employed in the latest tests, the U^{235} fraction serving as a standin for the plutonium fraction. A 6-in.-deep bed of UO_2 pellets, consisting of alternate 1-in.-thick layers of depleted (0.2198% U^{235}) and normal (0.7218% U^{235}) UO_2 pellets, was charged to the reactor. Equal quantities of the two enrichment types were used, the total being 4.4 kg. Sintered alumina, -100 mesh size with an average particle diameter of 84 μ , was used as the fluid-bed material. The fluidizing gas, 13 v/o oxygen in nitrogen, was fed at a superficial velocity of 1.5 ft/sec. The oxidation was conducted at fluid-bed temperatures of 450 to 500°C.

A series of radiographs taken during the oxidation period are shown in Fig. 9. The overall white appearance of the bed after $5\frac{1}{2}$ hr shows that oxidation was essentially complete.

Bed samples taken at the end of the oxidation from sampling points 8, 28, and 38 in. above the bottom of the fluid bed contained 0.4645, 0.4650, and 0.4714% U^{235} , respectively, thus indicating bed homogeneity with respect to U^{235} . These values differ by only about 1% from the average input U^{235} concentration of 0.4708%. Total uranium contents of samples taken at these three levels were 28.3, 31.5, and 32.3 w/o, as compared with 32.2 w/o based on the uranium recovered from all process streams and with 33.6 w/o

Initial Charge

1 hour

2 hours

$2\frac{1}{2}$ hours

3 hours

$3\frac{3}{4}$ hours

5 hours

$5\frac{1}{2}$ hours

Fig. 9. Gamma-ray Pictures of Oxidation of Uranium Dioxide Pellets

Temperature: 450°C

Initial Charge: 4.4 kg of UO_2 ; 6-in.-deep bed

Oxygen in Feed: 13 v/o

based on the uranium estimated to be in the charge. These results suggest that a fluid bed of alumina containing large quantities of fines can be sampled representatively for accountability purposes.

Modifications to the bench-scale unit are being made to accommodate BrF_3 as the fluorinating agent for uranium immediately following the pulverization step.

b. Recovery of Uranium from High-enrichment Fuels

(i) Studies with Irradiated Fuels. Studies with irradiated alloy and oxide fuel materials are being conducted in a bench-scale fluid-bed unit installed in the Chemical Engineering Division senior cave. The facility consists of a $1\frac{1}{2}$ -in.-dia fluid-bed reactor and a packed-bed filter with associated equipment for the containment of volatile chlorides and uranium hexafluoride produced in the process (see Progress Report for May 1964, ANL-6904, pp. 89-90). Studies made in this facility of the hydrochlorination and fluorination of uranium-Zircaloy-2 alloy fuels were reported previously (see Progress Report for March 1965, ANL-7028, pp. 64-65). Similar studies with uranium-aluminum alloy (U-Al) fuels are presented below.

Five runs were made in which the uranium in the U-Al fuels was separated from the aluminum by hydrochlorination in fluid beds of alumina maintained in the temperature range from 220 to 245°C. The uranium, thus separated, was reacted with elemental fluorine in the temperature range from 260 to 520°C for recovery as UF_6 . Most of the operating conditions were similar to those used in the previous bench-scale and pilot-scale studies with nonirradiated fuels. The characteristics of the fuels used in the five runs are given in Table XVII. The results of the five experiments with U-Al fuels show:

1. Uranium removal from the reactor was greater than 99% in all cases, indicating that fission products and the presence of a radiation field does not affect the recovery of uranium from irradiated U-Al fuels.

2. For the elements cerium, strontium, cesium, ruthenium, and antimony, the decontamination factors ranged from 1.2×10^3 to 7.2×10^4 when the uranium hexafluoride product was collected in a sodium fluoride trap. When an additional separation step (desorption from the sodium fluoride trap and collection in cold traps) was employed, the decontamination factors increased, ranging from 5.4×10^3 to 4.6×10^6 . The lowest decontamination was that obtained for ruthenium.

3. For the experiments with short-cooled U-Al fuels, decontamination factors for gamma and beta emitters of 2.4×10^7 and

1.4×10^6 were obtained. Iodine-131 was found to be removed almost entirely during the hydrochlorination step and collected nearly completely in a caustic scrubber. Trace amounts of I^{131} appeared in the stack off-gas. Tellurium activity was quantitatively collected by an activated alumina trap.

Table XVII. Uranium-Aluminum Alloy Fuel Charges

Run No.	Fuel Charge, g	Uranium in the Charge, g	Original Enrichment, %	Burnup, %	Cooling Period, months	Fission Products in Fuel Charge, g
1	100	4.1	0.71	-	-	-
2	107	6.15	93	46.5	7	5.85
3	108.4	6.2	93	46.5	7	5.9
4	{ 40.3 60.0	{ 2.5 2.2	{ 93 0.71	{ 47 -	{ 3 -	{ 2.3 -
5	{ 34.8 60.4	{ 2.2 2.2	{ 93 0.71	{ 47 -	{ 3 -	{ 1.9 -

F. Sodium Coolant Chemistry

1. Chemistry of Carbon in Liquid Sodium

a. Nature of Carbon in Sodium. Studies are continuing on the characterization and identification of carbon contaminants present in sodium. Such information is being sought by experiments in which sodium is hydrolyzed and the carbonaceous products are determined. The information thus obtained is indirect since the original carbon species may be altered during hydrolysis. Nevertheless, it is believed that the reaction products can give some insight as to the nature of the original carbon species. Gas chromatographic analysis will be used to identify carbon compounds in the gaseous products. Experimental conditions have been ascertained for resolving and identifying the following hydrocarbons by gas chromatography: methane, ethane, ethylene, and acetylene. Air, oxygen, nitrogen, hydrogen, carbon monoxide, and carbon dioxide can also be resolved by this method.

b. Reactions of Carbon Compounds with Sodium. An equilibration apparatus for use up to 800°C is being constructed to determine whether various carbon compounds react with or dissolve in liquid sodium. Appropriate chemical analyses will be performed on the filtered solutions and residues. To facilitate these analyses, carbon-14-labeled compounds will be used.

c. Ultrapurification of Sodium. In addition to distillation and filtration, two other techniques are being investigated for the ultrapurification of sodium from carbon. Previous studies (see Progress Report for February 1965, ANL-7017, p. 70) have shown that a considerable fraction of the carbon in sodium is present as submicron-sized particles. An effective means of ridding sodium of such carbon might be one whereby sodium atoms or ions are made to migrate through an electrolyte. Accordingly, thermal diffusion and electromigration of sodium through fused salts are being tested as processes for the removal of carbon. Two experiments employing the electromigration technique have been performed. The results of the experiments are being evaluated.

2. Control of Sodium Oxide Impurity

Two new oxygen cells from United Nuclear Electrochemical have replaced those damaged in tests during July (see Progress Report for July 1965, ANL-7082, p. 50).

Two sample tubes of sodium for oxygen analyses were taken from a bypass section of the loop and analyzed by the Chemistry Division. The time period between samples was 12 days. The results were as follows:

<u>Date</u>	<u>ppm Oxygen</u>
7-15	30, 32
7-27	45, 30, 19--Average 31

Assuming the initial oxygen content was 30 ppm, the voltage reading of the electrochemical cells was approximately 1.2 V. This base voltage versus chemical analysis is being correlated further.

G. Plutonium Recycle Reactors

1. Reactor Vessel

The mechanisms of cladding cracking (see Progress Report for July 1965, ANL-7082, pp. 50-51) lead to successive propagations of unsurfaced cracks. Accordingly, repair weld overlays over the bare SA212B shell steel were deferred to permit further studies of cladding-crack propagation and corrosion of the SA212B under reactor environmental conditions.

Vessel regions from which "boat" samples were removed for metallographic investigations were ground to remove the slitting-saw notches. Measurements showed that the maximum thickness reduction of the vessel wall was approximately 3/16 in. This reduction of the vessel wall does not affect the strength of the vessel for continued operations at

600 psig because the actual wall thickness in the local thin spots is still 9/16 in. thicker than that required by the ASME Code for "Power Boilers."

Two regions, one each in the steam and water zones, are being prepared for contour relief mapping for the corrosion studies. Two of the four upper shock-shield sections are being equipped with removable windows to permit visual observations of corrosion and the measurement of the rate.

The internal steam collector duct, previously removed for cladding observations, was replaced.

2. Reactor System Piping

Repairs of primary-system piping welds continued. Approximately 85% of those required for the cold critical experiments were completed and radiographed. A few new welds required repairs.

All of the primary-system piping welds required for the high-power reactor operations were radiographed. Corrective repair was concentrated on welds inside the containment shell to meet the time schedule for core loading. The remaining welds, together with the tunnel and reboiler building, will be corrected and radiographically reexamined, at a later time.

Radiographic examinations of the primary-system terminal valves (i.e., the first valve subject to reactor pressure) were started. Thirty-one of 34 valves examined thus far met the ASTM E-71 standards for Class II castings. Faults, and dispositions, of the three valves of poorer quality were:

- a. 1-3-in. feedwater stop valve: Class III -- blow hole near surface and repairable to Class II.
- b. Two $1\frac{1}{2}$ -in. water column angle valves: below Class V -- gross blow hole at midwall in acute angle adjacent to valve seats. Replacement of valve if welding repairs are unsuccessful.

Reinforcing rings (see Progress Report for May 1965, ANL-7046, p. 64) were welded to Filters No. 3 and No. 4 to reduce stresses in the body flanges of the flanged joint. The upper and lower welds joining the ring to the flange will be magnafluxed prior to the reinstallation of the filters.

III. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Critical Experiments

Experimentation continued with the 810 fuel-foil core. Measurements were made to determine the worth of five solid aluminum rods distributed in the internal thermal column (ITC). The rods displaced H_2O , producing voids which increased reactivity. By comparison with nonabsorbing Teflon rods of similar size, the reactivity effect of neutron absorption in the aluminum was determined, yielding an average of -0.015% reactivity for each percent volume of the flux trap occupied. The combined effect of voiding and absorption was to increase reactivity because the void was three times more effective, as shown in Table XVIII. Both effects were somewhat depressed near the periphery of the ITC, but azimuthal variations seem to be displaced 30° between the void and absorption coefficients. The minimum in void worth was in an ITC corner. The least effective location for absorber was midway along a side, i.e., coincided with the most effective azimuthal location for void.

TABLE XVIII. Reactivity Effects of Aluminum and Teflon Rods in ITC (810 Fuel-foil Loading)

Radius, cm	Teflon Worth*	Aluminum Worth**	Aluminum Absorption Coefficient [†]
0	0.046	0.031	0.015
1.59	0.046	0.030	0.016
3.18	0.048	0.031	0.017
4.58 (side)	0.048	0.039	0.009
5.33 (corner)	0.031	0.017	0.014

*Reactivity increase, in percent, for each percent of ITC volume voided by displacement, using a 1.27-cm-dia, solid Teflon rod at the indicated location. Each rod had the equivalent of 0.787% of the ITC volume.

**Reactivity increase, in percent, for each percent of ITC water displaced by a 1.27-cm-dia solid aluminum rod.

[†]Difference attributed to neutron absorption in aluminum, in percent reactivity for each percent of ITC volume occupied by aluminum.

Installation of a graphite thermal column (42 x 52 x 84 cm) outside the beryllium radial reflector permits rough measurement of the increase

in neutron flux obtainable at the vessel wall (of the critical facility) by displacing water from the region between the beryllium reflector and the wall. This information may be helpful in determining whether AARR operational flux detectors require thimbles penetrating the pressure vessel wall. An increase to at least 300 times the reference flux level was obtained at the vessel wall (5-ft radius in the critical facility), but large variations resulted from small changes in geometry, procedures, etc. Difficulty was found in making foil-activation measurements because of post-shutdown irradiation (after H₂O dump) by neutrons from fission product (γ, n) reactions with the beryllium reflector.

2. Analyses of Transients

An analog computer program was prepared to study the effect of changes in relevant reactor parameters upon predicted transient behavior of AARR. The changes studied fall into the three general categories of reactor properties which determine its response to reactivity perturbations, including the following:

a. Neutronic Properties

- (i) Mean prompt-neutron lifetime
- (ii) Void coefficient of reactivity
- (iii) Water temperature coefficient of reactivity
- (iv) Doppler effect on reactivity

b. Thermal Properties

- (i) Metal heat capacity
- (ii) Metal thermal conductance
- (iii) Convective film heat transfer coefficient

c. Design Conditions

- (i) System pressure
- (ii) Coolant Flow rate

Variations were made singly in each of the above quantities, and the transient responses to 33-ms ramp reactivity insertions of 0.75, 1.00, and 1.50 dollars were determined. Comparisons were made against results obtained for design center or reference values of all quantities. All results were obtained for a simulated initial reactor power level of 100 MW.

The changes in value of thermal properties and of void and water temperature reactivity coefficients were selected in the range of $\pm 25\%$ of reference values; gross changes in the reference values for design

conditions and the neutron lifetime and Doppler effect were studied. The variations selected for study do not indicate actual uncertainties or anticipated changes in the AARR design, but rather have been arbitrarily chosen to illustrate the relative importance of reactor parameters on transient calculations.

Table XIX presents data obtained for those variations which yield moderate changes in transient behavior and which are therefore presented for only the most severe transient, due to \$1.50 reactivity insertion, and for the largest variations in each quantity studied. Transient results determined for large changes in mean prompt-neutron lifetime and magnitude of the Doppler effect are displayed on Figs. 10 and 11, respectively.

TABLE XIX. Effect of Various Reactor Properties on Transient Behavior of AARR

Item Varied	Variation Magnitude, %	Peak Power Variation, %	Integrated Energy Variation, %
Metal Heat	+20	+25.3	-22.1
Capacity	-20	-24.1	-22.1
Metal Thermal	+20	-4.7	-4.2
Conductance	-20	+3.7	+4.2
Film	+20	-3.2	-2.5
Coefficient	-20	+4.7	+3.9
Coolant	+22.2	-2.3	-1.6
Flow Rate	-22.2	+0.8	+0.7
System			
Pressure	-76.9	0	-9.6
Void Coefficient	+25	-13.9	12.1
of Reactivity	-25	+18.6	+17.0
Water Temperature	+25	-1.4	-4.0
Coefficient of Reactivity	-25	+1.7	+1.6

- Notes: 1. Reference value of peak power, 3700 MW.
 2. Reference value of integrated energy, 43.4 MW-sec.
 3. Estimated accuracy of computed table entries, values given $\pm 1\%$.

The values of integrated energy were derived by two different methods of calculation. For the data shown in Table XIX and Fig. 11, the time of occurrence of the transient power pulse is practically unaffected by the

variation imposed on the reactor. Hence, integrated energy values were determined for 60 msec following initiation of a transient which includes nearly the entire power pulse for all cases studied.

Fig. 10. Prompt-neutron Lifetime Effect on AARR Transient Behavior

Fig. 11. Doppler Effect on AARR Transient Behavior

Changing the prompt-neutron lifetime, however, essentially changes the time scale of the transients. To illustrate this effect, the values of integrated energy presented in Fig. 10 are those obtained following invitation of a transient to the time of peak power for a prompt-neutron lifetime of $10 \mu\text{sec}$. The times required to attain to peak power for these transients are also shown in that figure.

The major conclusion from the data in Table XIX is that the transient response of AARR to rapid rates of reactivity insertion is appreciably sensitive to only those changes which directly affect prompt reactivity compensation mechanisms, e.g., metal heat capacity and the effect of void coefficient on the reactivity worth of metal expansion. The

importance of prompt feedback mechanisms for rapid, super-prompt critical transients is further illustrated by the results presented in Fig. 11 for gross changes in the Doppler effect.

Although the results exhibit insensitivity to changes in the design conditions of coolant flow rate and pressure, a reduction in these quantities should be viewed as detrimental to the transient performance of AARR because of the accompanying reduction in margin against core burnout. Specific core-damage estimates have not been included in these calculations.

The beneficial effect of extending the mean prompt-neutron lifetime, as should occur with core burnup, is clearly illustrated in Fig. 10. Peak power is reduced, the time to peak power is increased, and hence the safety-system action can be significantly more effective.

3. Heat Transfer

Calculations have been performed by means of the CHIC-KIN computer code¹⁸ as part of an analytical program for determining the behavior of AARR hot channels during reactivity-induced power transients. The case studied was that of a one-dollar step in reactivity starting at a steady power level of 100 MW. The power versus time behavior of the hot channel with skewed axial heat generation was available from results of previously reported analog computer studies (see Progress Report for December 1964, ANL-6997, pp. 53 and 54) and was used as input for the CHIC-KIN computations. Other input conditions and physical properties were chosen so as to agree with values used in previous work, with the exception that the initial coolant velocity was a nominal 45 ft/sec and did not contain a 1.08 hot channel factor as in the analog studies. The fuel region was divided into ten axial and eight radial sections.

The calculations constituted a parametric study involving three quantities, all of which must be supplied as constant input quantities for a given run:

- Φ_{DNB} Maximum surfact heat flux which can be maintained with nucleate boiling;
- f_r Fraction of heat going into voids;
- τ Bubble collapse (or removal) time.

These quantities are essential to the physical description of transient boiling employed in CHIC-KIN and their variation can have a profound effect on the course of hot channel behavior.

¹⁸Redfield, J. A., CHIC-KIN - A Fortran Program for Intermediate and Fast Transients in a Water-moderated Reactor, WAPD-TM-479 (Jan 1965).

The important features of the calculative runs are summarized in Table XX. For most runs, printouts of plate temperatures, surface heat fluxes, and fluid conditions at 1-ms intervals for times up to 30 ms after initiation of the transient and at 2-ms intervals from 30 to 100 ms were available. Run 5 was extended to 180 ms, and Runs 10 and 12 to 200 ms.

TABLE XX. Summary of Computed CHIC-KIN Results for AARR Hot, Skewed Channel at 100-MW, One-dollar Steps

Run No.	$\Phi_{\text{DNB}} \times 10^{-6}$, Btu/hr-ft ²	f_r	τ , ms	Comments
1*	7.1	0.002	100	No melting. $\Phi_{\text{Surf}} < \Phi_{\text{DNB}}$.
2	6.25	0.002	100	No melting. Transition boiling.
3	5.0	0.002	100	Melting indicated >100 ms during steady-state runout. (Film boiling)
4	4.38	0.002	100	Blind spot in code. Subcooled DNB.
5	6.25	0.01	100	No melting or pressure surge indicated >180 ms during steady-state runout.**
6	6.25	0.05	100	Small pressure surge (~18 psi) at time of initial vaporization (~20 ms), followed by large surge (~2000 psi) at 40 ms and fluid expulsion. Computation ceases.
7	6.25	0.1	100	Pressure surge of ~250 psi at time of initial vaporization (18 ms) followed by large surge (~2000 psi) at 28 ms and fluid expulsion. Computation ceases.
8	6.25	0.1	10	Pressure surge of ~250 psi at time of initial vaporization (18 ms). Pressure loss (~200 psi) at 38 ms following appearance of vapor at $J = 7$. Negative internal pressures at 43 ms indicating large surge. Computation ceases.
9	6.25	0.1	1.65	Slight pressure oscillation (+3 to -12 psi) noted at time of initial vaporization (~17 to 23 ms). No subsequent pressure pulse or melting indicated.
10	5.0	0.002	10	Melting indicated >200 ms during steady-state runout. (Film boiling)
11	5.0	0.002	1.65	No melting indicated >100 ms (Film boiling). Temp in plate will reach 1400 to 1750°F before steady (2.3 times initial) heat generation is accommodated.
12	5.0	0.01	1.65	Melting indicated >200 ms. (Film boiling)

*System pressure for Run 1 = 700 psia; all other runs at 650 psia.

**Conclusions about no large pressure surges during "runout" assume a final power level 2.3 times initial.

The parametric studies indicate that a wide variety of effects may be predicted, depending upon which values of the transient boiling parameters Φ_{DNB} , f_r , and τ are chosen. At present, measurements of these parameters do not appear to be available for AARR hot channel transients. Furthermore, the model of transient void formation in CHIC-KIN is an oversimplification of an extremely complicated process.

If the post-critical heat-flux model described by the CHIC-KIN program is acceptable, the possibility exists that under some conditions no melting of fuel will occur beyond DNB during certain power transients. This was illustrated by Runs 2 and 11. In the former run, the plate temperatures peaked and receded without departing from the transition to the film-boiling region because a relatively high value of Φ_{DNB} was specified. In the latter run, although film boiling was indicated, the relatively low value of τ chosen resulted in a very small void fraction and relatively large vapor conduction film-boiling coefficients. Thus, the current computations are more optimistic than those of the analog studies referred to above, in which complete insulation past DNB is assumed.

The CHIC-KIN code predicts that sizable pressure surges (as high as 250 psi for the case studied) may occur upon initiation of subcooled local boiling during a transient if the rate of vapor formation is rapid enough. Very large local pressures ($>2,000$ psia) are also predicted during the subsequent steady-state runout when a sizable amount of vapor is present in the fuel channel. In the latter case, the fluid is at, or near, saturation conditions at the channel exit, and expulsion is predicted from both ends of the channel. Whether, in fact, such pressure surges occur as predicted remains to be verified. Their magnitudes are questionable because the CHIC-KIN code assumes a constant reference pressure and neglects sonic-velocity effects. The number of axial sections chosen in the computation also affects the pressure surges.

For the one-dollar step at 100 MW, it is believed that values of f_r no less than 0.01-0.05 and values of τ no less than 10 ms would constitute reasonable guesses as to values of these parameters which are conservative from a safety viewpoint. Values of f_r as high as 0.05 or 0.1 cannot be ruled out. DNB fluxes computed from the Zenkevich-Subbotin critical heat flux correlation should be conservative during the power burst, but might not be overly so during the quasi-steady-state runout. Thus, it must be concluded from the results of the present series of computations based upon current knowledge or lack thereof, that a one-dollar step at 100 MW will lead either to melting of fuel and/or pressure surges with fluid expulsion at hot channel locations. This conclusion supports previous estimates of the analog studies referred to above.

The modifications of the STDY-3¹⁹ code previously reported (see Progress Report for June 1965, ANL-7071, pp. 56 and 57) have been completed. The changes permit use of a film coefficient based on average film temperature, the use of the Zenkevich-Subbotin critical heat flux correlation, and provides the solution of conditions for flow instability.

4. Fuel and Materials Development

Contracts have been completed and work is in progress in the following areas:

- a. investigation of the problems associated with the fabrication of 6 w/o UO_2 -SS cermets;
- b. ZrB_2 burnable-poison addition and attestation of dispersion in cermets;
- c. UO_2 strength and characterization studies;

¹⁹Pyle, R. S., STDY-3, A Program for the Thermal Analysis of a Pressurized Water Nuclear Reactor during Steady-state Operation, WAPD-TM-213 (June 1960).

- d. AARR fuel-production quality-control techniques;
- e. material-compatibility studies.

Investigation of fuel-assembly brazing techniques has been completed by Pyromet Company (see Progress Report for March 1965, ANL-7028, p. 71). The studies terminated with the delivery of a complete, all-brazed, dummy fuel assembly.

5. Primary System and Components

The testing of two small beryllium corrosion specimens (see Progress Report for July 1965, ANL-7082, p. 59) was discontinued after exposure for 1150 hr to deionized water at 200°F flowing at 44 fps. Final results are as follows:

	<u>Specimen No. 1</u>		<u>Specimen No. 2</u>	
<u>Total Time, hr.</u>	<u>Weight Loss, mg/cm²</u>	<u>Corrosion Rate, mpy</u>	<u>Weight Loss, mg/cm²</u>	<u>Corrosion Rate, mpy</u>
97	0.19	3.6	0.24	4.6
478	0.70	2.7	0.80	3.1
1150	1.68	2.7	1.74	2.8

The single pit (previously reported) noted on the edge of one specimen after 478 hr did not change during the final 672 hr. Neither did the thin, tarnish-like film, which appeared during the first 97 hr of the test.

The plot of average weight loss versus time yields a straight line whose slope corresponds to a corrosion rate of 2.8 mpy. Past tests under similar conditions, some of which lasted as long as two years, have shown that beryllium corrodes at a constant rate. It is concluded, therefore, that under the test conditions, 2.8 mpy represents an accurate corrosion rate which would remain constant for long periods.

A test designed to simulate the conditions which will exist at the tips of aluminum beam tubes in the AARR was initiated (see Progress Report for July 1965, ANL-7082, p. 59). In this test, heat generated in a 6.5-in.-long, 6061-T6 aluminum specimen is removed by the flow of cooling water through a 0.5 x 0.05-in. channel. The specimen is heated by passage of 60-cycle AC through it. Eight thermocouples were spot welded to the outside of the specimen, which was then insulated so that all of the heat is removed from the inside water-cooled surface. Coolant velocity is fixed at 10 fps, and the bulk cooling water temperature at the axial midpoint of the specimen is

controlled at 150°F. The heat input to the specimen is increased until the temperature at the aluminum-water interface at the axial midpoint reaches 260°F. A heat flux of 820,000 Btu/hr-ft² is required to achieve this condition.

The water interfacial temperatures along the specimen at the start of the test, as calculated from the thermocouple readings and from the inlet and outlet water temperatures, are shown below. The fluid heat transfer film coefficients calculated from these data are also shown.

Distance from Inlet End, in.	Temperature at Water Interface, °F	Bulk Water Temperature, °F	Fluid Film Coefficient, Btu/hr-ft ² °F
1.00	248	130	6950
1.25	257	132	6560
2.25	256	143	7260
3.25	256	154	8040
3.25	261	154	7660
4.25	267	165	8040
5.25	274	176	8370
5.50	265	178	9430

Although the above data are not completely self-consistent, they are reasonably so, and generally bracket the conditions of interest.

Since the operating conditions are held constant, any temperature increase indicated by the thermocouples is the result of corrosion product formation on the water-cooled surface. The test has been in progress 137 hr. During this time, the midpoint temperature of the specimen increased approximately 20°F. Under the conditions of test and assuming a thermal conductivity of the corrosion product layer of 1.3 Btu/hr-ft²-°F, this 20°F temperature rise indicates an oxide thickness of 0.38 mil.

B. Magnetohydrodynamics

Extensive cycle studies have established the potential of the liquid-metal MHD cycle both for space and central station applications. In comparison with plasma MHD cycles, the liquid-metal concept had the advantage of high efficiency (50%) at a relatively low temperature.

The purpose of this study is to determine the feasibility of using a liquid-metal MHD generator as a topping unit for a fast reactor central power station. An experimental program is underway to measure and optimize the efficiencies of two major components of the cycle: the pump which converts thermal energy from the reactor to kinetic energy in the working fluid, and the MHD generator itself.

1. MHD Condensing Injector

Efforts have been directed toward the design, construction, and testing of a condensing injector (see Fig. 12) associated with MHD power cycle studies. In order to obtain adequate stagnation head at the

Fig. 12. Condensing Injector

injector exit, previous cycle studies have indicated that the required high-velocity vapor entering the mixing section will be supersonic and that it is necessary that the area contraction at 2 approach maximum limits. The maximum area contraction is naturally limited by the area of the liquid nozzle exit at 1.

Freon-11 has been chosen as the fluid for study because of the following considerations. Existing facilities in Building 11 are available for Freon-11. The low value of latent heat under operating conditions (compare F-11 ~ 70 Btu/lb_m, Hg ~ 120 Btu/lb_m, Cs ~ 200 Btu/lb_m, and water ~ 850 Btu/lb_m) is not too dissimilar to the latent heats of some liquid metals, implying, of course, a reduction in heating requirements. Moreover, maximum area contraction can be achieved without constructing an excessively large injector.

Three-hundred-kilowatt service is being extended to the area adjacent to the existing Freon-11 loop. This should be sufficient to allow complete testing of a condensing injector with 1/2-in.-dia at Section 2. (Approximately 2100 kW would be required to test a steam-water injector of similar size.) Previously used test sections and Calrod heaters have been removed in preparation for necessary modifications, and arrangements are being made to bring 25-gpm water service to the loop for cooling requirements.

2. Liquid-metal Generator Studies

The NaK-nitrogen loop has been modified to accommodate experimental investigations of liquid-metal MHD generators. The loop is schematically illustrated in Fig. 13 and photographically in Fig. 14. Referring to Fig. 13, the NaK is circulated by a specially designed canned rotor pump to the mixer where the nitrogen is injected. The two-phase mixture is then passed through the nozzle where the NaK is accelerated by the nitrogen. The high-velocity effluent then passes through the MHD generator and into the separator. The NaK is returned by gravity feed to the supply tank, and the nitrogen is discharged through a series of filters and a holdup tank to the atmosphere.

Fig. 13. Schematic of MHD NaK-N₂ Loop

Fig. 14a. North View of NaK Generator Loop

Fig. 14b. South View of NaK Generator Loop

Fig. 14c. View of MHD Generator between Magnet Poles

Tasks completed this past month are:

1. installation of the electromagnetic flow meter;
2. installation of the pump cooling system; this system was integrated into the magnet-cooling system and uses kerosene as a coolant;
3. installation of liquid-level indicators and alarms in the supply and calibration tanks.

In addition, the final electrical hookup of the generator and other instrumentation was initiated.

C. Energy-conversion Systems

1. Regenerative Emf Cells

a. Bimetallic Cells. The sodium-bismuth cell (without a thermal regenerator) previously described (see Reactor Development Program Progress Reports for January and February 1965, ANL-7003, p. 74, and ANL-7017, p. 77) has now been in operation as a battery for over 250 days at temperatures between 540 and 630°C. The cell has been discharged and electrically recharged over 100 times with no observable decrease in cell performance.

b. Engineering Material Study. Dynamic corrosion studies of low-carbon steel and Type 316 stainless steel by liquid bismuth and sodium-bismuth alloys have been continued. These tests are being conducted to find suitable materials of construction for the sodium-bismuth regenerative cell system. The tests are carried out in thermal convection loops. Previous tests have been discussed in Reactor Development Program Progress Reports for October and November 1964 and January 1965, ANL-6955, p. 90, ANL-6977, p. 80, and ANL-7003, p. 75.

Mild steel and Type 316 stainless steel loops containing bismuth or sodium-bismuth alloys, which were operated at hot leg temperatures of about 850°C and cold leg temperatures of 450°C, developed leaks after periods of operation ranging from 125 to 430 hr because of internal corrosion due to mass transfer. However, at hot leg temperatures of about 650°C and cold leg temperatures of 550°C, a Type 316 stainless steel loop has been in operation for over 3000 hr and a mild steel loop has been in operation over 1000 hr. No leaks have developed in either loop.

These dynamic corrosion tests indicate that low-carbon and stainless steel will not be suitable as materials of construction for operation with sodium-bismuth alloys at hot zone temperatures of 850°C or higher. However, they may be suitable for some operations at temperatures of 650°C or lower, such as those employed in the cell portion of a sodium-bismuth bimetallic system.

Loops of niobium, tantalum, molybdenum-30 w/o tungsten, and niobium-1 w/o zirconium have been fabricated for testing with liquid bismuth and tin.

IV. NUCLEAR SAFETY

A. Reactor Kinetics

1. Fast-neutron Hodoscope

The multichannel prompt-neutron camera (see Progress Report for July 1965, ANL-7082, p. 62), developed to detect sample fuel position as a function of time and space during TREAT meltdown experiments, using specimens inside opaque materials (as, for example, in the case of sodium loop tests), was set up at TREAT (see Fig. 15) and tested during reactor transient experiments. In these, transparent capsules were used in order to make direct comparison between optical and neutron camera representation. Although only 50 channels of electronics were used, the collimating matrix of slotted holes was full-sized, and the entire installation was made to duplicate a complete unit as far as possible.

In Fig. 15 the test fuel pin (or cluster) is shown in its opaque test assembly. Radiation from the sample passes through openings in special slotted core elements, and openings in the reactor shielding. Angled slots in the multichannel collimator are pointed toward the focal area in the reactor. Individual detector packages are located at the end of the collimator slots. Detector output signals are processed electronically to reduce the background, and transmitted to the data acquisition and readout unit (see Fig. 16). Signals are sent to an array of flashing fluorescent indicator tubes, arranged in the same pattern as the collimator slot matrix. Below the light array are test number and a real-time clock digital readout panel. The light output from indicators fed by collimator slots pointed at the fuel is higher than that from collimator slots pointed away: thus, a motion-picture film of the indicator light patterns constitutes a record of fuel position as a function of time and space.

2. Meltdown Experiments with Sodium

A 4.1% enriched EBR-II Mark I fuel pin, surrounded by a ring of six dummy pins consisting of empty EBR-II cladding tubes, run in an integral sodium loop with no-flow initial conditions, has been examined. Energy input was about 65% greater than that of the first integral loop sample, which was also run under no-flow initial conditions, and was carried just short of the failure threshold (see Progress Report for July 1964, ANL-6923, p. 87). Extensive failure resulted from this high energy input, as may be seen from Fig. 17 which is a view of the samples taken during the post-transient inspection. At the top is half of the test section, removed after the section was slit longitudinally. Immediately below is the other half, containing the test sample nesting in four of the dummy elements. Extensive failure is evident from one end of the fueled pin to the other. At the bottom of the photograph are the two dummy pins

Fig. 16. Electronic Processing and Photographic Readout Assembly

Fig. 17. Samples Run in Integral Sodium Loop under No-flow Conditions

removed from the cluster to show the sample pin. The general appearance of the failed pin indicated pressure failures, with vigorous expulsion of fuel. Essentially no fuel was left inside the cladding. Fuel remains were found near the regions of failure, with little attack of the adjacent, hollow steel dummy pins, and no evidence of appreciable secondary fuel movement after the initial expulsion. The transient recording of output from the test-section inlet pressure transducer showed small pressure surges (fractions of an atm) superimposed on the smooth response of the transducer to reactor energy release, with a sharp pressure spike of about 3 atm occurring at 1.86 sec. This sharp spike could be a recording of sample failure. This analysis is consistent with calculations of transient temperature, which indicate that the vapor pressure of the sodium bond should equal the estimated bursting pressure of the cladding at 1.87 sec even though fuel temperatures were above the melting point at this time. Thus, pressure, and not dissolution of the cladding, should be the dominant mechanism of failure.

3. Fast Reactor Safety

a. Coolant (Water) Expulsion Studies. During the past month, the strain-gauge pressure transducers have been dynamically tested in order to determine fidelity in recording rapid pressure transients. The first tests consisted of pressurizing a small-volume (170 cc) container to about 20 atm and suddenly dropping the pressure to 1 atm by rupturing a diaphragm. Nitrogen and water were used as pressurizing fluids, neoprene and copper sheets were used as diaphragms, and the discharge orifice diameter was varied from 12.7 to 38.1 mm. The output signal from the pressure transducer was recorded on an oscilloscope, with a 6-V dry-cell battery as an energizing source, and compared to the signal recorded on the recording oscillograph which is to be used in the expulsion tests. The

length of time required for the pressure to drop from 20 atm to 1 atm varied from 1.6 to 87.5 ms. (which corresponds to frequencies of about 11 to 625 cps); in this range, the two signals were identical, including blips caused by the pressure shock waves (which exhibited frequencies of the order of 6000 to 7000 cps).

In the second series of tests a shock tube was used. In this situation, the pressure transducers were required to respond to a sudden increase in pressure caused by the rupturing of a diaphragm which separated high- and low-pressure chambers in a long tube. The pressure shock wave passed the face of the transducer in approximately 38 μ sec, corresponding to a frequency of approximately 26,000 cps. The transducer diaphragm vibrated with this high-frequency pressure shock. However, no differences were found in the traces from the oscilloscope and the oscillograph.

It is concluded that: (1) the pressure transducers will measure pressure pulses up to a frequency of about 6 to 7 kcps with good fidelity, and (2) the recording oscilloscope and associated instrumentation (amplifier, attenuating circuit, etc.) will faithfully reproduce signals at frequencies beyond 7 kcps. Since the expected transient signals will have frequencies of the order of several kcps, the existing instrumentation appears to be adequate. The actual expulsion experiments can now be undertaken.

b. Critical Flow Studies. Calculations of the progression of accidents in sodium-cooled reactors reveal that critical flow may exist at the discharge end of the reactor coolant channels.

When critical flow occurs, large pressure gradients exist at the discharge end of the reactor in addition to large pressure gradients within the core. These pressures must be known in order to calculate the magnitude of the sodium void fractions within the core.

As a first step in the study of this problem, it is planned to study the fundamentals of critical flow with a small steam-water system while concurrently designing a small-scale sodium experiment. Tests are planned with both the water and the sodium systems. At present, the initial water tests involve the measurement of void fractions (velocity ratios) under critical flow conditions with steam-water as the working mixture. Specifically, a detailed investigation of the fluid behavior at the point of choking in rectangular and circular constant-area ducts will be carried out. The data obtained from these experiments is required to evaluate accurately existing critical flow models.

A preliminary design of the experimental equipment has been made. Two sections will be made: one a 1/4-in. circular tube, and the other a 1/8 x 1-in. rectangular duct, each 3 ft long. The rectangular test section will be used to measure void fractions.

Construction of the test facility has been started. The small-scale loop in Building 11 is presently being modified and connected to the blow-down tank to serve as a heater for the working fluid, and equipment has been ordered for the two test sections.

4. Fast Reactor Control

a. Determination of Neutron-flux Levels through Measurements of Higher Moments. Neutron-flux level is determined by measuring the output current of an ion chamber placed in the radiation field. In the d.c. method, the averaged d.c. current, which is proportional to the neutron flux, is read, whereas in the a.c. method (Campbell system) the variance σ^2 or the second moment of the fluctuating current is read. Although the a.c. method results in a smaller signal strength, its ability to discriminate against background γ radiation is greater than in the d.c. method.

Determination of neutron level through the measurement of higher moments manifests a possibility of even greater discrimination against background radiation. The measurement of the third and fourth moment has been investigated, and it is shown that the highest moment which can be practically utilized is the third one. Flux measurements through the third moment has a very large discrimination ratio against γ radiation (10^6 and 10^3 times that of a conventional and Campbell system, respectively), thus making it possible to determine neutron-flux level in the presence of relatively high γ radiation.

A statistical analysis of the system reveals that the amplifier noise does not limit the accuracy of the measurement; it affects only the length of the measuring (averaging) time.

b. Space-Time Reactor Dynamics. A physically meaningful weighting function was derived by use of a variational principle. The functional chosen, the rate of change of the detector reading in the reactor, is made to be invariant with respect to arbitrarily small flux variations. The derivation is as follows: if we denote the detector reading as $N(t)$:

$$N(t) = \int_V W^T \phi(\vec{r}, t) dv, \quad (1)$$

where $W^T(\vec{r}, t)$ is the undetermined weighting function and $\phi(\vec{r}, t)$ is the group flux. Then

$$\frac{dN}{dt} = \int_V \left[\frac{\partial W^T}{\partial t} \phi + W^T \frac{\partial \phi}{\partial t} \right] dv. \quad (2)$$

If the reactor equations are written in operator form,

$$\frac{\partial \phi}{\partial t} = H\phi + S, \quad (3)$$

where H is the operator involving the space variables but not time, and S is the external source.

Define another operator H^* which satisfies

$$\int_v W^T H \phi \, dv = \int \phi^T H^* W \, dv. \quad (4)$$

Substitution of (3) and (4) into (2) yields

$$\frac{dN}{dt} = \int_v \left[\phi^T \frac{\partial W}{\partial t} + \phi^T H^* W + W^T S \right] dv. \quad (5)$$

Upon applying the variational principle with respect to $\delta \phi^T$ and using only the first and second term of the Taylor series expansion of $\phi^T H^*$ with respect to $\delta \phi$, the first-order variation of Eq. (5) is

$$\int_v \delta \phi^T \left[\frac{\partial W}{\partial t} + \frac{\partial(\phi^T H^*)}{\partial \phi^T} W \right] dv. \quad (6)$$

If the first-order variation is to vanish for arbitrarily small variation of $\delta \phi$, then

$$\frac{\partial W}{\partial t} + \frac{\partial(\phi^T H^*)}{\partial \phi^T} W = 0. \quad (7)$$

The solution of (7) presents no difficulty if ϕ is known. However, it is not necessary to have the exact form of ϕ since the functional (5) is not sensitive to small variations of ϕ . Thus, in the practical case, the unvaried ϕ can be used for the calculation.

B. TREAT

1. Operations

Metal-water reaction samples CEN 208 and 209 were irradiated. One consisted of a cluster of nine stainless steel-clad uranium oxide pins, and the other was a similar cluster of Zircaloy-clad pins.

Testing of the fast-neutron hodoscope (see Section IV.A.1) continued.

2. Large TREAT Loop

Three bellows assemblies for the Large TREAT Loop were completed in Central Shops and shipped to Idaho. One unit replaced the bellows damaged during installation, one replaced a defective unit, and the third unit will be stored as a spare. A spare bellows assembly (3-in. pipe size) was completed and is stored in Building 206 for possible future use. The dummy TREAT half-elements were finished in Central Shops and shipped to Building 206 for cleanup and assembly checkout.

A stress analysis of a "Conoseal" joint gasket was conducted. Numerical calculations indicated that compressive stresses at the upper inner edge and the lower inner edge of a stainless steel gasket (of 1.99-in. ID and 2.537-in. OD) reach 651,000 and 565,000 psi, respectively, showing that plastic flow does exist during and after assembling. Theoretical design characteristics for nonleaking gasket at elevated temperature have been checked. It is shown that in a 2.407-in. (coupling nominal diameter) stainless steel Conoseal joint, the gasket remains under radial compression at 1200°F. It is also shown that the width and thickness of the gasket and the inclination angle of bevel surface are the critical factors for successful sealing operation at high temperature.

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

1. Metal-Water Reactions

a. Studies in TREAT of HFIR Fuel. The reaction of water with aluminum-41.45 w/o U_3O_8 cermet core fuel plates clad with 6061 aluminum is being studied in experiments in which the plates, submerged in water, are exposed to TREAT transients. The fuel samples are small sections of a High Flux Isotope Reactor (HFIR) fuel plate. The objective of these studies is to obtain data about the extent of reaction and the physical behavior of the fuel material under conditions that simulate a power excursion or a run-away incident. Similar studies have been made with aluminum-uranium alloy fuel plates (see Progress Report for May 1964, ANL-6904, p. 106). The technique consists of subjecting an autoclave containing the sample

(weight, ~1.2 g), 45 cc of water, and 60 cc of free volume (helium filled) to a burst of neutrons in TREAT.

The extent of metal-water reaction is computed from the amount of hydrogen evolved on the assumption that the aluminum present reacts as follows:

Experiments have been performed with the system initially at 30°C (20 psia helium) and at 285°C (1000 psia H₂O vapor). Experimental results are summarized in Table XXI.

TABLE XXI. Summary of Results of TREAT Experiments with HFIR Fuel Samples^a

	CEN Run No.							
	198	200	199	201	204	205	206H	207H
<u>Reactor Characteristics</u>								
Integrated Power, MW-sec	532	134	168	170	258	425	165	263
Period, ms	152	148	109	109	109	68	106	104
Peak Power, MW	202	305	591	602	588	1460	590	588
<u>Experimental Data</u>								
Initial Wt, g	1.291	1.296	1.288	1.292	1.300	1.300	1.303	1.305
Water Temp, °C	30	30	30	30	30	30	285	285
Autoclave Pres, psia	20	20	20	20	20	20	1000	1000
Fission Energy, cal/g	133	335	420	425	645	1062	413	658
Metal-Water Reaction, %	0.2	1.2	1.4	2.5	74.6	28.4	11.8	91.3
Physical Appearance	intact	intact, bulged, edge ruptured	intact, center bulged	intact, bulged, badly ruptured	one fragment and gray powder	one large fragment, several small metallic fragments, gray powder	intact, bulged, ruptured	one large fragment, small amount of gray powder

^aSamples were 1.0 by 0.5 by 0.050-in. plates with 10 mils of 6061 Al cladding on two sides and a 30-mil thick Al-41.45 w/o U₃O₈ cermet core.

The HFIR samples retained their plate-like shape at fission energies as high as 425 cal/g, whereas the aluminum-uranium alloy fuel plates had fused into single globules at energies between ~200 and 530 cal/g. Large hemispherical cavities were apparent in the surface of the HFIR plates at energies above that sufficient to cause melting of the cladding (~200 cal/g). The appearance of the cavities suggested gas evolution.

At energies above 645 cal/g, the HFIR samples completely lost their shape. The HFIR cermet fuel samples at these energies formed one large globule that contained mostly uranium oxides, several smaller fragments of partially oxidized aluminum, and fine particulate that consisted mainly of aluminum oxide. (Previous analytical studies indicated that the aluminum oxide was mostly α -Al₂O₃.) On the other hand, the alloy plate

fuel at these energies was generally found, in previous studies, to form several small fragments of aluminum, uranium, and oxides, and fine particulate oxides. In 30°C water, the amount of fine particulate formed increased as the extent of reaction of either fuel increased. In high-temperature water each of the two types of fuel formed a single large globule that reacted extensively at high energies.

The extent of reaction is plotted as a function of fission energy input in Fig. 18. The extents of reaction of the HFIR fuel plates (as contrasted to the differences in physical behavior noted above) were quite similar to those of the alloy plates

Fig. 18. Results of TREAT Experiments with Samples of High Flux Isotope Reactor Fuel: Percent Metal Reacted vs Fission Energy

Core: 30 mil, Al-41.45 w/o U₃O₈
 Cladding: 10 mil, 6061 Al
 Sample Size: 1.0 in. by 0.5 in. by 0.05 in.

to those of the alloy plates (see ANL-6904). In 30°C water, at low energies, only a few tenths of a percent of a plate reacted when the cladding was only partially melted; and reaction did not exceed 2.5% when the cladding was fully melted, but still retained its shape (at 425 cal/g). At higher energies more extensive reactions occurred: at 645 cal/g, the extent of reaction was 74.6%; at 1062 cal/g, the extent of reaction was 28.4%.

In high-temperature (285°C) water, more extensive reaction occurred than in the runs in 30°C water. At 413 cal/g, 11.8% of a plate reacted, while in room temperature water only 2.5% reaction occurred at 425 cal/g. Similarly, at 658 cal/g, 91.3% of a plate reacted in hot water, while in 30°C water 74.6% reacted at an energy input of 645 cal/g.

It is postulated that at low energies, for which breakup does not occur or the plates do not lose their shape, the plates cannot react extensively, i.e., the nature of the reaction is to form a protective oxide film, and cooling rates are sufficient to prevent ignition. The subcooled water causes an even greater suppression of the reaction as a result of a more rapid cooling rate than in the heated water. Above a threshold energy (previously determined to be 530 cal/g for the alloy plate fuel and somewhere between 425 and 645 cal/g for the HFIR fuel), flow, breakup, and ignition of the samples occur. At these higher energies, the extent of reaction is governed by the duration of burning, which, in turn, depends on several factors, such as particle size, depletion of the reacting fuel, and the rate of energy loss by the particles to the surroundings. Here again, the subcooled water tends to decrease the extent of reaction by providing more efficient cooling.

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