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MASTER

Argonne National Laboratory

REACTOR DEVELOPMENT PROGRAM

PROGRESS REPORT

March 1966

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

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REACTOR DEVELOPMENT PROGRAM
PROGRESS REPORT

March 1966

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FOREWORD

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

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

The last six reports issued
in this series are:

September 1965	ANL-7105
October 1965	ANL-7115
November 1965	ANL-7122
December 1965	ANL-7132
January 1966	ANL-7152
February 1966	ANL-7176

REACTOR DEVELOPMENT PROGRAM
Highlights of Project Activities for March 1966

EBR-II

Modifications and maintenance to the reactor and plant systems were completed on schedule and Run No. 15, scheduled for 795 MWd, was begun on March 25. The reactor is operating at 45 MWt.

The fuel-transfer arm was repaired by providing a bell crank of new design and material, and it was re-installed in the reactor following the same shroud-enclosed, inert-atmosphere procedure as for its removal. Only minor adjustments in alignment were required for satisfactory operation when the primary system reached 700°F.

Fuel handling for the current run was routine and involved sixteen subassemblies, including the loading of two experimental subassemblies containing capsules of carbide and oxide fuels and structural materials.

ZPPR

Questions posed by AEC Supplement to the Preliminary Safety Analysis Report, which described a new backup containment structure, were answered. A summary report of filtration studies has been sent to the AEC.

Specifications for the procurement of fuel elements have been prepared.

ZPR-3

A second phase of the SEFOR critical experiments, run as part of the AEC assistance program to the SEFOR project, was initiated. Experiments on a two-segment core, to measure reactivity changes in a simulated, expanded fuel condition, were completed. Work was begun on a mockup with a three-segment design of a fuel element.

Plutonium Recycle Program (EBWR)

Calculations have been made of the uniform temperature coefficient of the plutonium critical when poisoned with 5 gm/gal of boric acid by four-group, two-dimensional diffusion theory. The results indicate a positive temperature coefficient between 70 and 170°F, in qualitative agreement with experiment.

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I. LIQUID-METAL-COOLED REACTORS

A. EBR-II

1. Operations

Scheduled modifications and maintenance to the reactor and plant systems were completed, planned checkouts of equipment and systems were performed, and Run No. 15 was begun.

Before replacing the repaired fuel-transfer arm (see Progress Report for February 1966, ANL-7176, pp. 1-2), a periscope and light source were used in the opening of the primary tank cover to inspect the annuli between the large and small rotating plugs and between the large rotating plug and the primary tank cover (see below).

Modifications to the transfer-arm locking mechanism were finished, including the installation of a bell crank of new design. Following comprehensive testing of the operation of the lock pin and sensing indicator out of the reactor, the transfer arm was positioned in its inert atmosphere bag over the reactor and reinstalled. Also, control-drive shaft No. 11 was removed and replaced in position No. 9 with the original unit which had been repaired by the installation of bellows and gripper assembly.

Operation of the transfer arm and its locking pin were first tested in place when the system reached 350°F. The secondary and steam systems were also brought to this temperature while testing out the new control circuitry for the secondary-sodium piping-trace-heater system.

During startup preparations, drilling was begun on vertical holes through each rotating plug, which are intended to extend down to the area of the offset and lower annulus to facilitate inspection of the annulus. Each hole is 3 in. in diameter. One hole was drilled to a depth of 35 in. (to the level of the cooling air collector plenum). Further drilling was discontinued so as not to delay the startup of the reactor. Drilling will be resumed during future periods of reactor shutdown.

As heatup approached normal standby conditions of 700°F, the transfer-arm alignment was again checked with respect to the fuel-unloading port, storage basket, and main fuel gripper. With only minor adjustments, alignments proved satisfactory.

Seven subassemblies, including two experimental irradiation subassemblies, were loaded into the fuel storage basket in preparation for loading the reactor core for Run No. 15.

During the preparations for unrestricted fuel handling (transfer of subassemblies between the reactor and the storage basket), it was discovered that the jaw actuator for control-rod drive No. 7 had failed. Evidence indicated that the bellows which seals the jaw-actuating shaft had failed, allowing sodium to enter and freeze in the upper portion of the drive shaft.

This rod was removed and assembly No. 8, which had been repaired previously by the replacement of the bellows and the gripper assembly, was installed. Subsequent checkout of this drive and the unit replaced in position No. 9 was satisfactory. Rod drive No. 7 had previously been removed from the primary tank, washed, and replaced.

Unrestricted fuel handling was then begun to prepare the core for Run No. 15. Sixteen subassemblies were changed: eleven core-type, one control-rod, one safety-rod, and three enriched inner-blanket types. One of the two sources in the reactor was removed from position 7-E-6 for examination of the beryllium. The shutdown neutron count level decreased from ≈ 120 to 70 cps as a result of this change. This value is still well in excess of the 5-cps minimum count rate required for startup. The beryllium thimble will be destructively tested and its helium content determined to provide information on the remaining life of such sources.

With fuel handling completed, the reactor was started for Run No. 15, scheduled for 795 MWd. Criticality was achieved on March 25; following physics measurements, 45 MWt was reached the next day. The power-generation system, with overhauled feedwater pump and new main feedwater-control valve, is operating satisfactorily.

2. Maintenance and Minor Modifications

All of the maintenance and modifications planned for this shutdown period were completed as scheduled. Installation of the new secondary-sodium piping-trace-heater system was completed. Checkout and operation during heatup from ambient to 700°F was satisfactory. The installation of the feedwater pressure-control valve and its control system was completed. Operational checkout appears to be satisfactory above approximately 10% feedwater flow. Evaluation of this system will continue during this next period of power operation.

The annual leak-rate test of one electrical penetration and the seals between the secondary sodium piping and the Reactor Building was satisfactorily concluded.

Numerous other minor items were completed, such as: replacement of flange gaskets and leaking steam traps in the Sodium Boiler Building; inspection and repair of major control valves in the steam system; inspection

and replacement of sight glasses on the feedwater heater, flash tank, and steam-drum; and improvement of sodium control-valve operation in the primary purification system.

a. Rotating Plugs. Early in the month, attempts were made to view (from beneath) the annuli between the large and small rotating plugs and between the large rotating plug and the primary tank cover, by use of an optical device and light source inserted through the transfer-arm nozzle. The purpose of this was to determine whether the sticking problem with the rotating plugs might be caused by deposition of sodium and/or sodium oxide in these annuli.

Because of the limitation on length of the viewing device imposed by the dimensions of the transfer-arm nozzle, it was possible to view only a limited arc (a few degrees) of the small plug annulus. However, points on an arc of about 60° of the large plug annulus, in the vicinity of the transfer-arm nozzle, were viewed. The small plug annulus at the point viewed appeared to be free of deposits. Deposits were observed near the bottom of the large plug annulus at several locations viewed. These deposits appeared to be collections of "globules." It is surmised that these are droplets of sodium from "refluxing" of sodium vapors in the annulus. There was no evidence of annulus blockage at any location viewed. However, because of the limited area viewed and because of the difficulty in providing a sufficiently strong light source to permit clear viewing in the upper part of the annulus, it cannot be stated unequivocally that no obstructions exist in the annuli.

To provide additional information concerning the possibility of sodium deposition in the annuli, temperature measurements were made above the step in the large plug annulus. Measured temperatures were above the melting point of sodium, even with the bulk sodium temperature of 280°F .

b. Nuclear Instrumentation. When the compensated ion chambers for Channels 4, 5, 6, and 7 were removed for routine maintenance, a check indicated deterioration of their performance. The reason for this could not be readily identified, but the effect appeared externally as a current and open-circuit voltage. The current when ten or more volts were applied to the positive electrode was 7×10^{-10} Amp. The open-circuit voltage was as much as 12 V, depending on the detector. A current level of this magnitude would create an overlap problem in the startup range; therefore, replacement chambers were required. The ones chosen were Type NA09 with characteristics nearly the same as the original detectors and available with mica-alumina insulators and alumina connectors. Because the reason for the breakdown of the original detectors could not positively be identified, radiation damage to the electrical insulators was assumed. The mica-alumina insulators in the new detectors should offer protection against

radiation damage. The new detectors have been installed for Channels 4, 5, and 6. The Channel 7 detector is presently scheduled for replacement following the next operating period.

c. FERD Loop Instrumentation. The fission counters used as detectors for Channel A of the FERD loop instrumentation have been replaced by BF_3 counters, which have a sensitivity of 28 counts per nv, or roughly 50 times that of the fission counters. The preamplifiers, linear amplifier, and count-rate meter used have been modified to match the detectors. Readout of the signal will be on the second pen of a two-pen recorder, with Pen 1 assigned to Channel B of the same system.

In an effort to reduce the spurious noise level on the incoming line, the input voltage regulator has been replaced with a unit isolated by 90 dB between input and output.

3. Reactor Improvements

a. Control of Primary-tank Immersion Heater. A continuously variable heater-power control was installed for each of the six primary-tank heater assemblies. Variation of heater power is accomplished by varying the firing point of silicon control rectifiers (SCRs), using a timed trigger pulse to fire the SCRs. The SCRs are standard units, commercially available, rated at 235 Amp rms, 800 V. Two SCRs, connected in parallel and reverse, are used for each assembly, which contains four heaters connected in parallel and rated at 28 kW and 480 V. By using two SCRs, it is possible to take advantage of the complete cycle of the input sine wave, and the control system is smoother in operation.

Noise was introduced into the nuclear channels because of the routing of the nuclear signal and the heater-power cables. To correct this interaction, the heater power cables were removed, twisted to form a twisted pair, and reinstalled in flexible conduit. This has corrected the difficulty, and operation of the system has been satisfactory from both a control and operating standpoint.

b. Trace Heating for Secondary System. The scanning section of the control system has been replaced. The replacement equipment is a packaged unit comprised of solid-state switching modules whose purpose is primarily complete elimination of relays and stepping switches. Basically, the functions performed are identical to those of the original equipment; only the method of performing the functions has been changed.

The 76 heating units to be controlled are scanned consecutively for the pipe temperature for comparison to a preset control point. If the temperature difference is greater than 5°F , the heater is either energized

or de-energized, depending on the difference temperature. The scanning rate is preset at 2 sec per point. At this rate, each heated section of pipe is checked once each 2.5-min interval. This rate appears adequate for control based on operating experience gained to date. Following completion of this part of the project, the scanning rate can be increased. For the present, the rate has been kept purposely slow to reduce the number of operations of mechanical relays still in the system.

c. Oscillator System, Mark II. The physical configuration and objectives of this new oscillator rod system have been described in the Progress Reports for October 1965, ANL-7115, p. 3, and November 1965, ANL-7122, p. 3. The positions of the various parts of this complex mechanism will be monitored electrically. For example, limit switches will indicate the discrete positions of the jaws which grip the oscillator rod. Also, the rotational position of the rod will be monitored by several different electrical devices, and the settings of the transmission controlling the rotary speed will be controlled and displayed remotely. The various functions of the oscillator drive will be controlled either from the Reactor Control Room or from the Fuel Handling Console, depending on the reactor status. The conceptual design for the control and interlock circuitry has been completed, the detailed design of all wiring on the mechanism has been completed, and all the electrical components are on order. A temporary test console is being fabricated to be used for checking all of the electrical functions of the device during trial assembly.

4. Reactor Kinetics

a. Control-rod Calibrations. All control rods were recalibrated at zero power. Control rods No. 1 and 6 were period-calibrated. The total worth of each of the other nine rods (excluding No. 9) was obtained by intercomparison with rods 1 and 6.

The values as measured are given below:

<u>No.</u>	<u>Ih</u>	<u>No.</u>	<u>Ih</u>
1	132	2	151
3	122	4	149
5	120	6	147
7	123	8	146
9	SS	10	167
11	132	12	159
Avg	126	Avg	153

The average value agrees with previous calibrations, even though the worths of individual rods have changed due to changes in core loading.

b. Power Coefficient. The power coefficient was measured (see Fig. 1). The total reactivity defect from 0 to 45 MWt was 67 lh. The distinct change in the power coefficient previously observed between 20-25 MW apparently has disappeared. It appears that the effects of sub-assembly bowing (if this were the cause of the lower negative coefficient at low powers) are spread out over the entire power range. Each circled point on the curve represents a minimum of 1/2-hr equilibrium at power. At 45 MW, the point "X," taken after a wait of over 2 hr, shows an increase of 2 lh. This gives some insight into the time required to reach equilibrium at any power level. The stainless steel rod in control rod position No. 9, which is used for the rod-drop tests, was calibrated by a period measurement and by comparison with a calibrated control rod, giving values of 21 and 23 lh, respectively. Rod-drop tests from 10-in. insertion (15 lh) were made at both 0.5 and 45 MW. The data obtained from these tests have not yet been analyzed.

Fig. 1. Power Coefficient at Start of Run No. 15

5. Experimental Irradiations

a. Status. Two experimental subassemblies, X009 and X010, were inspected and then tested in the water flow loop. Following this, they were loaded into the reactor. One experimental fuel subassembly, XA01, and two α -measurement subassemblies were removed from the reactor.

The two subassemblies loaded contain the following experimental capsules:

<u>X009</u>	<u>X010</u>
2 Pu-U mixed carbide capsules (ANL-MET)	4 Pu-U mixed oxide capsules (GE)
2 Pu-U mixed oxide capsules (ANL-MET)	4 structural materials (PNL)
2 cermet (PuO ₂ in SS) capsules (ANL-MET)	11 structural materials (ANL-MET)
3 Pu-U mixed carbide capsules (UNC)	
4 structural materials (PNL)	
2 structural materials (GE)	
2 structural materials (ANL-MET)	

Subassembly X009 was loaded in position 4-A-2; X010 in 7-F-3.

Subassembly XA01, containing 19 pins of Pu-U-Fz alloy, was removed from position 4-D-2 at the experimenter's request.

The two α -measurement subassemblies, C220X and C221X, were removed from positions 1-A-1 and 3-B-2, respectively. Unloading was required because the burnup limit had been reached on the Mark-I driver fuel, which comprised the largest portion of the 91 elements in the subassemblies.

The status of experimental subassembly irradiations at the start of Run No. 15 remains as reported in the Progress Report for January 1966, ANL-7152, p. 6.

b. Neutron Radiography. The use of boron nitride-coated vidicon tubes is being investigated for use on neutron-imaging systems for the non-destructive examination of experimental capsules and fuel elements.

Two neutron-sensitive vidicon pickup tubes were received and tested. One tube was unsatisfactory; silver from a silver-bearing epoxy had migrated and shorted out the target. Testing of the other tube revealed that the desired signal could not be isolated from the system noise. Increasing the target voltage and changing alignment of the camera may improve the signal somewhat.

The low-bay area of the Inspection and Test Facility, Bldg. 772, which will be used in support of the experimental irradiations program, has been completed and accepted for occupancy by the Laboratory. The benches, etc., have been moved in, and the area is now being utilized for setup and alignment of the television system to be used in neutron-radiography studies. A welding machine, to be used eventually in the final assembly of experimental subassemblies, is also in the area.

6. Reactor Physics

Work is in progress for the calculation of power generation, flow, and temperature distributions in a 91-subassembly core of Mark-IA fuel, with 14 experimental subassemblies "homogenized" in Row 4.

By use of the MACH-1 computer code, fission rates have been computed for two cases. The first case assumes a radial blanket of depleted uranium as in the presently constituted reactor; the second, a stainless steel radial blanket in Row 7 and Row 8. In Table I, fission rates for these cases are compared with rates in a reference core of 73 subassemblies.

TABLE I. Fission Rates

	Reference Core	Case 1 (Uranium Blanket)	Case 2 (SS Blanket)
Total number of core subassemblies	73	91	91
Maximum fission rate at core center for 45 MW (fissions/g/sec $\times 10^{-12}$)	9.3	8.0	8.5
Calculated k_{eff}		0.993	0.989
Ratio of maximum fission rate in indicated row to maximum fission rate at core center			
Row 4	0.850	0.860	0.857
Row 5	0.755	0.795	0.790
Row 6	0.595	0.705	0.705
Row 7	0.420	0.582	0.595

The contribution of gamma heating to reactor power is being calculated. Also in progress is a study of pressure drops and flow distribution in the 91-subassembly core. For the latter work, reference is being made to the EBR-II model study performed by the Franklin Institute during EBR-II design.

Power-generation data, based upon the aforementioned fission rates, and flow-distribution data will be used in calculating temperatures in the 91-subassembly core.

7. Fuel Cycle Facility

a. Operations. Preliminary examination of the cladding on a few elements from an irradiated radial blanket subassembly (A-711) indicated no increase in cladding diameter. This subassembly had been in Row 6 of the reactor for 5050 MWd of reactor operation. These elements have been shipped to ANL-Illinois for a more detailed examination of the uranium rods.

Analytical results from melt-refining runs show that at 1400°C essentially quantitative removal (greater than 99.5%) of barium, lanthanum, cesium, and iodine is obtained for liquation periods ranging from 0.25 to 4.0 hr, and that the effect of liquation time, within this range, does not appreciably affect the removal of these elements. Data for the removal of cerium indicate that greater than 2.5 hr of liquation at 1400°C are required for quantitative (greater than 99%) removal of this element. For periods less than 2.5 hr, the removal of cerium falls off from 99% removal at 2.5-hr liquation time to about 80-85% removal for 0.25-hr liquation time.

Examination of additional fuel pins from subassembly C-162 (the first subassembly fabricated in the FCF from reprocessed irradiated fuel and thus had received two cycles of irradiation) failed to show the scale on the outside of the fuel pin that had previously been observed on other pins from this subassembly (see Progress Report for January 1966, ANL-7152, p. 7). This examination was conducted on the pins after they had been decanned and the sodium removed by an alcohol wash. As previously reported, the increase in volume resulting from this scale did not agree with measurements of the sodium level within the can, which had not indicated an abnormal increase in fuel pin volume. Based on this latest examination and the previous sodium-level measurements, it is surmised that the scale resulted from a leak in the sodium washing vessel which allowed air to contact the fuel pins. This would have resulted in the formation of a uranium oxide scale layer on the surface of the pins. This investigation will be continued when the next subassembly (C-195) fabricated from reprocessed irradiated fuel is received from the reactor.

With achievement of normal reactor tank operating temperature and power operation, routine fuel transfers between FCF and the reactor plant were resumed. Production and transfer is summarized in Table II.

TABLE II. Production Summary--March 1966

Subassemblies received:*	5**
Subassemblies dismantled:	5
Subassemblies fabricated:	15
Subassemblies transferred to reactor:*	5
Pins decanned:	461
Melt Refining: Irrad. 4 Recycle 9	
Avg pour yield: 92.9% 93.0%	
Injection casting runs:	16
Pin processing:	
Accepted	1352
Rejected	214
Pins welded:	1325
Leak testing:	
Accepted	1482
Rejected	40
Bond testing (completed runs):	
Accepted	1383
Rejected	84
Surveillance:	A711, A776X, B311, B312, C195

*Transfer of fuel between the reactor and the FCF was temporarily suspended during part of the month as a result of the reactor modification program.

**Beryllium source thimble included.

b. Vertical Assembler-Dismantler. A specific-purpose machine is being developed which will enable the disassembly of EBR-II experimental subassemblies for nondestructive examination of the experimental elements and then the reassembly of these experimental elements for further irradiation. In order to carry out such examinations without disrupting the liquid-metal bond of the fuel pins, the samples must be kept vertical at all times. The capability of interim examination and of regrouping test elements during the course of long irradiations will materially increase the efficiency with which irradiations can be carried out in EBR-II.

The conceptual design of this combination machine is in progress and is based on the experience with the dismantling and assembling machines currently in operation in the Fuel Cycle Facility. A wooden mockup of a combination fuel element removal and loader has been evaluated. This device is currently being designed in sufficient detail to permit the fabrication of a prototype unit.

B. General Fast Reactor Physics

1. ZPR-3

The core loading of the SEFOR reactor mockup in ZPR-3 has been changed to represent a two-segment fuel element loading by placing a 2-in. gap in the fuel columns in the core drawers of Half No. 2 (see Fig. 2). The gap occupies the space from 5 to 7 in. from the reactor interface. Two inches of axial reflector were removed from the core to make room for the gap. The two-segment fuel-element design is intended to reduce the effect of fuel expansion due to heating.

Fig. 2. Typical Core Drawer Showing Gap Location

The reactivity loss measured in the simulated fuel-expansion experiment for the two-segment fuel-element loading was still larger than desired, so a three-segment fuel element design was mocked up. The 2-in. gap which was placed in Half No. 2 remained the same, but a $1\frac{5}{8}$ -in. gap was placed in the fuel columns of the core drawers in Half No. 1, located between 7 and $8\frac{5}{8}$ in. from the reactor interface. Measurements of power distribution have begun, but have not yet been analyzed. A more detailed report on this mockup will be forthcoming.

a. Reactor Composition. The reactor composition for the two-segment fuel-element loading (Assembly 47A) is given in Table III (see Progress Report for February 1966, ANL-7176, pp. 5-19 for the location of the various reactor regions; Region 1C is the gap). The radius of the core region 1A (new fuel region) has been increased from 30.1 cm to 33.8 cm to have more of the 1.1 w/o Al-Pu plates available for increasing the fuel

TABLE III. Composition of Assembly 47A for Loading 30

	Reactor Region		Material Composition (g/cc)												
Name	No.	Volume (liters)	Pu ²³⁹ + Pu ²⁴¹	Pu ²⁴⁰	U ²³⁸	U ²³⁵	Be	O	Mo	C	Na	Al	304 SS	Ni	Fe
Core (New fuel)	1A	255.5	0.6071	0.0525	3.086	0.006	0.081	0.407	0.079	0.067	0.253	0.322	1.32		
Core (Old fuel)	1B	272.1	0.6279	0.0299	3.045	0.006	0.084	0.412		0.067	0.252	0.337	1.45		
Gap	1C	31.0			1.297	0.005	0.082	0.285		0.036		0.290	1.34		0.336
Radial Sodium and Stainless Steel	2	661.5									0.383	0.022	2.20		
Radial Reflector	3(Ni)	579.2										0.276	0.60	6.36	
Radial Reflector	3(SS)	155.6										0.274	6.29		
Axial Reflector	4A	97.7								0.035		0.239	1.62	4.25	
Axial Reflector	4B	66.1								0.037		0.211	1.59	4.54	
Axial Sodium and Stainless Steel	5	127.7								0.075		0.482	3.58		

enrichment in core region 1B. The volume of the axial reflector in one-half of the reactor has been decreased because of the insertion of the gap in the fuel. The composition of the radial sodium and stainless steel region 2 includes the increased sodium mentioned in the Progress Report for December 1965, ANL-7132, p. 13. However, the value for this region given in the following should be used in place of that given in the earlier report.

The composition given in Table III is for a loading with the fuel columns not expanded and SEFOR rod II in the "out" position (withdrawn 40 in.). The fuel in the reactor at this loading is 326.0 kg of $\text{Pu}^{239+241}$. If the SEFOR rod II and the ZPR-3 control rod were both placed at their "in" positions, the multiplication constant k would be 1.0042 ± 0.0001 .

The plutonium mass for the above loading is different from that given last month because the original loading with the gap was with the fuel in the expanded condition. The location of the boron carbide thermal shield mockup was also different.

b. Calibration of SEFOR Control Rod. The results of calibration of SEFOR control rods II and III is shown in Fig. 3. The two rods were

Fig. 3. SEFOR Rod Calibration in Assembly 47A of ZPR-3

worth an average of 117 cents of reactivity. The gap appears not to affect the average worth of the SEFOR control rods. (The locations of the SEFOR control rods are shown in Progress Report for December 1965, ANL-7132, Fig. 3, p. 12.)

c. Simulated Fuel Expansion. The purpose of the two-segment fuel-element design was to provide a reduced change in reactivity from fuel expansion due to heating of the core during SEFOR Doppler-effect measurements. One fuel segment expands into the gap, reducing its size, while the other fuel segment expands out of the core. Both fuel movements reduce reactivity, but the reduced gap size is expected to increase reactivity due to reduced neutron leakage.

Figure 4 shows the simulated fuel-column expansion when 1/8-in.-thick aluminum shims are used. The shims are placed only in the drawer columns containing either fuel or depleted uranium; the remaining core materials are not moved. By placing the two shims in the columns of Half No. 1, the axial core-reflector interface is moved in that half of the reactor by 1/4 in. Although the fuel-segment base remains fixed in the SEFOR reactor, the net effect of the expansion is changed, and this is shown in Fig. 4(b). The 22-in. fuel segment expands 3/8 in. into the gap, reducing the gap width by that amount. The 12-in. fuel segment expands out of the core 1/8 in., making the overall core length 36 1/8 in. The net effect of the simulated fuel expansion resulted in a loss of 43.4 ± 2.3 cents of reactivity. This includes the correction for the worth of the aluminum in the shims.

Fig. 4. Shims and Gap Relationship in Fuel-expansion Experiment

d. Fission and BF_3 Traverses. To determine the effect of the 2-in. gap in the fuel on power distribution in the core, axial traverses were made at the center of the reactor, using Pu^{239} and U^{238} fission counters and a BF_3 proportional counter. The results from the three counters are given in Fig. 5-7. The softening of the spectrum in the gap can be seen from these traverses.

Fig. 5. Axial Traverse with Pu^{239} Counter for Assembly 47A of ZPR-3

Fig. 6. Axial Traverse with U^{238} Counter for Assembly 47A of ZPR-3

Fig. 7. Axial Traverse with BF₃ Counter for Assembly 47A of ZPR-3

Radial traverses with Pu²³⁹ and U²³⁸ fission counters at the mid-plane of the reactor were made. Figures 8 and 9 show these results. The slight tilting of the curves which showed up in the radial traverses in the unsegmented mockup was not apparent in these results, apparently having been corrected by a slight redistribution of the core fuel.

Fig. 8. Radial Traverse with Pu²³⁹ Counter for Assembly 47A of ZPR-3

Fig. 9. Radial Traverse with U^{238} Counter for Assembly 47A of ZPR-3

e. Reactivity Traverse. An axial traverse was made to determine the reactivity worth of plutonium as a function of axial position. The reactivity effect of a small stainless steel can containing 24.09 g of plutonium, placed in a holder on the end of the traverse mechanism, was measured in the axial direction. A dummy stainless steel can was then placed in the holder and measured to correct for the reactivity effect of the can, holder, and traverse rod. The results are given in Fig. 10.

Fig. 10. Axial Reactivity Traverse for Pu Sample (24.09 g) in Assembly 47A of ZPR-3

2. ZPR-6

a. Fission Traverses in Assembly No. 4Z. The radial mapping of the fission rates for U^{235} and U^{238} in assembly No. 4Z continued.

As in the previous measurements, U^{235} foils (5 mils thick and 3/8 in. in diameter) and U^{238} foils (5 mils thick and 1/2 in. in diameter) were inserted vertically between adjacent material plates and at a distance of 3 in. from the interface of the halves. Some foils were placed horizontally on top of the U^{238} blocks of the reflector. The majority of the foils, however, were located along the horizontal diameter of the stationary half.

The positioning of the U^{235} foils was chosen in such a way as to cover the entire diameter of the central and blanket zones, and together with previous measurements, to give a complete picture of the radial traverse for the U^{235} fission rates. Figure 11 shows the results.

Fig. 11. Radial Traverse of U^{235} Fission Rates in Assembly No. 4Z of ZPR-6

The U^{238} foils covered a radius from the core center to the first full blanket drawer (from 0.0 to 62.5 cm); Fig. 12 shows the results

of present and previous measurements. The U^{238} fission-rate fine structure has been measured across five drawers (from 26.5 to 56.9-cm radius), so as to cover the buffer and driver zones. A detailed mapping of this fine structure is given in Fig. 13.

Several measurements were also made along a radius at 45° to the horizontal diameter, through the buffer and driver regions; no particular influence due to the orientation of the constituent plates was detected.

As in previous measurements, the results seem to be in good agreement with transport calculations. Diffusion-theory calculations adequately describe the situation in the central zone, but fail to explain the U^{238} fission ratios across the buffer and the driver zones.

Fig. 12. Radial Traverse of U^{238} Fission Rates in Assembly No. 4Z of ZPR-6

Fig. 13. Radial Traverse of U^{238} Fine-structure Fission Rates through the Buffer and Driver Zones of Assembly No. 4Z of ZPR-6

b. Doppler Measurements. Assembly 4Z was returned to the standard loading pattern used in the 1965 Doppler measurements. Recheck of criticality, control-rod worth, and the Doppler effect in a natural U^{238} oxide (expandable) sample indicated a very close reproduction of the conditions of the previous measurements.

A re-run of an axially and radially constrained enriched U^{235} oxide element was carried out to check anomalous results previously obtained. A U^{238} sample, unavailable last summer, has also been run. Reduced data from these runs is not yet available.

Voiding of the entire central zone of Assembly 4Z is presently underway. The final three samples to be measured with this assembly will be done in this sodium-voided configuration.

3. ZPR-9

Conversion of ZPR-9 from an aluminum to a stainless steel matrix is continuing on schedule. The aluminum matrix has been removed from the reactor beds and the stainless steel matrix is being installed. Minor

mechanical modifications, required by the smaller size of the stainless steel matrix and the need to provide needed experimental access holes, are essentially completed. The auxiliary matrix support platforms have been installed.

4. ZPPR

Questions raised by the Commission (CH and DRDT) on review of the Supplement to the ZPPR Preliminary Safety Analysis Report have been answered. A document containing these answers, as well as a summary of the filtration studies done by the Industrial Hygiene and Safety Division, has been transmitted to the Commission.

The following is the current status of reactor components:

a. Bed and Tables. All drawings except the electrical schematics are complete and approved by ANL. One table casting was poured on March 24 and one of the bed castings on March 31. Arrangements have been made with Giddings and Lewis to have an ANL Procurement Services representative inspect the castings when they arrive at the vendor's machine shop. Machining on other portions of the assembly is 5% complete.

b. Matrix Drawers. The vendor, Mechanical Products Mfg. Co., rejected the stainless steel sheets shipped by the steel producer because they were below the minimum thickness dimension. The specifications call for 0.031 ± 0.002 in. of Type 304 stainless steel sheet. A new order was received and is within specifications. The first 400 drawer sheets have been sheared and are now being perforated.

c. Matrix Tubes. The vendor, Van Vetter, Inc., has on hand 1200 tubes which the manufacturer has inspected. Another 1200 are now being inspected by the vendor, and the final 600 tubes are now in the process of being fabricated. Previously, ANL has inspected two lots of 600 tubes each.

d. Fuel-rod Drives. One of the ten rod drives received was returned for repairs. This drive has been satisfactorily repaired. All ten rod drives have been checked out and accepted.

e. Nuclear Instrumentation. Work has stopped on these units pending clarification of ANL's position.

All other ANL procurement items are delayed pending the release of funds by the AEC.

C. General Fast Reactor Fuel Development

1. Metal Fuels

a. U-Pu-Zr and U-Pu-Ti Fuels. Phase relationships, compatibility with potential cladding materials, and properties of the high-melting U-Pu-Zr and U-Pu-Ti fuels are being investigated.

(i) Phases and Reactions. An analysis of reactions in the U-Pu-Zr ternary system indicated that these alloys undergo two class-II reactions during heating from room temperature to reactor operating temperature:

(1) $\gamma + \beta \rightarrow \alpha + \zeta$ at approximately 655°C;

(2) $\alpha + \gamma \rightarrow \delta + \zeta$ at approximately 600°C.

(ii) Thermal Conductivity. The thermal conductivity of a U-13 a/o Pu-22 a/o Zr alloy has been measured. The values range from 0.100 W/cm-°C at 100°C to 0.301 W/cm-°C at 800°C. The thermal conductivity of this alloy correlates well with the values reported for two U-Pu-Zr alloys previously (see Progress Report for December 1965, ANL-7132, p. 19). The data were obtained in a comparative-type apparatus with Armco iron as the standard.

(iii) Compatibility. Further studies have been made of compatibility diffusion couples of U-Pu-Zr and U-Pu-Ti alloys with various cladding materials at temperatures up to 800°C. Our first tests¹ showed that, with Type 304 stainless steel, the U-Pu-Zr alloys were compatible at 800°C but the U-Pu-Ti alloys were compatible only to 650°C. Subsequent investigations have pointed to three factors as possibly influencing these results: the oxygen content of the fuel, the nickel content of the stainless steel, and the composition of the phases present at the diffusion temperatures.

The good compatibility of the U-Pu-Zr alloys with stainless steel seems to depend partly on the oxygen content of the alloys. A phase containing zirconium and oxygen seems to form a barrier film between the fuel and the stainless steel.

The method of formation and stability of this phase is being investigated. Diffusion couples have been run at 800°C for seven days between Zr-10 a/o O and stainless steel, and also between Ti-10 a/o O and stainless steel. The Zr-O/stainless steel couple showed no apparent reaction at 800°C, while a considerable diffusion layer formed between the Ti-O and stainless steel.

¹Reactor Development Program Progress Report for July 1965, ANL-7082, p. 20.

Compatibility couples have also been run between U-14 a/o Pu-26 a/o Zr and Type 304 stainless steel that had been spot welded to the fuel. Gross melting occurred at the fuel-cladding interface at 800°C and 750°C, but not at 700°C after heating for seven days at these temperatures. Metallographic examination of this couple is in progress. A heat-affected zone and small amounts of molten weld metal were observed in the vicinity of the spot welds. The method is being studied to determine its applicability to compatibility diffusion couples.

In most of the diffusion couples, high-nickel alloys have shown progressively poorer compatibility with these metallic fuels as the nickel content increased from 20% to 50%.² A recent test between U-14 a/o Pu-26 a/o Zr and two stainless steels indicated that only a small difference in nickel content might be detrimental. One steel was Type 304 stainless steel containing 8.5 w/o Ni and the other was a modified Type 316 (niobium stabilized) containing 11.5 w/o Ni. The effect of nickel is being studied by means of several Type 304 stainless steel alloys with increasing nickel content.

The tentative ternary phase diagrams have indicated certain phase compositions that should appear in these fuel alloys. Between 670 and 800°C the U-15 a/o Pu-25 a/o Zr is a gamma solid solution, whereas the U-15 a/o Pu-25 a/o Ti alloy contains two phases: a ternary compound isostructural with U_2Ti , and a plutonium-rich gamma solid solution. The plutonium content of this gamma phase may contain up to 25 a/o Pu. The poor compatibility with stainless steel exhibited by U-Pu-Ti may be caused by the greater concentration of plutonium in the gamma phase.

2. Jacket Development

a. Thermal Expansivity of V-5 w/o Ti-15 w/o Cr. The thermal expansivity of annealed V-5 w/o Ti-15 w/o Cr was determined between 25 and 960°C. The dilation specimen was taken from rolled sheet material (parallel to rolling direction) after a one-hour anneal at 900°C in vacuum. Specimen density was 6.16 g cm^{-3} . The values for the mean coefficient of linear expansion are given in Table IV.

TABLE IV. Mean Linear Expansion Coefficients
of Annealed V-5 w/o Ti-15 w/o Cr

Temperature Range (°C)	Mean Linear Expansion Coefficient ($10^{-6} \text{ } ^\circ\text{C}^{-1}$)
0-250	9.72
0-500	10.42
0-750	10.95
0-960	11.45
0-1000	11.55 (extrapolated)

²Reactor Development Program Progress Report for October 1965, ANL-7115, p. 31.

3. Zero-power Reactor Fuel Development

a. U-Pu-based Fuels. Compatibility, dilatometric, and metallographic specimens have been made of two alloys of interest for ZPPR fuel, U-25 w/o Pu-2.5 w/o Mo and U-28 w/o Pu-2.5 w/o Mo. Tests for compatibility of these alloys with Type 304-L stainless steel have been initiated, and dilatometric and metallographic work are in progress.

b. ZPPR Fuel Elements. Sufficient progress was made on the development of ZPPR fuel elements (see Progress Report for January 1966, ANL-7152, pp. 24-32) to write specifications for their procurement. Because of the large quantities of these elements needed, the Laboratory has investigated manufacturing processes that might be used and has carried at least one of these processes into the pilot production stage.

Four U-Mo master alloy melts were made. About 10% of the charged molybdenum failed to appear in the resulting binary alloy. This can be compensated by overcharging molybdenum to the melts. Thirteen ternary melts were made and cast into thirty 9 to 10-in. long castings and six 5 to 6-in. long castings. Composition was varied from 25 to 30 w/o Pu to provide samples for properties measurement, ignition, and gamma-spectrometry calibration over that range.

Yttria-coated carbon crucibles and magnesia crucibles were used for both binary and ternary melts. Magnesia crucibles performed well for both binary and ternary alloying provided that the melt temperature is kept below 1400°C. One magnesia crucible was used five times.

Three methods of parting core plates from the cast bar have been tested. These were (i) hot sawing with high-speed steel cutters and the bar heated to 175°C to 200°C, (ii) sawing at room temperature with tungsten carbide milling saws and (iii) grooving and breaking. The hot-sawing method works well and diminishes the chipping tendency, but it caused considerable difficulty in maintaining an as-cut tolerance closer than ± 0.010 in. This was traced to the difficulty of stabilizing the temperature of the core bar stock, of the milling arbor and of the milling machine. About 2.5 hr was required to bring the core plates, milling machine, and arbor to temperature. This was deemed excessive for production. An added disadvantage is that a high-purity inert atmosphere is required to prevent oxidation of the cores at 200°C.

Very low feed rates were necessary to prevent breaking of the carbide cutter. The most satisfactory results were obtained with 75-mm-dia by 1.5-mm-thick, 72-tooth cutters. The speed was 50 rpm and the feed 0.05 mm per revolution. Edge chipping at the point of milling cutter breakthrough continued to be a problem. This was eliminated by placing a 0.64-mm aluminum pad between the milling fixture and the core plate. The

milling machine was set to cut just below the lower surface of core plate and into the aluminum pad. Chipping was eliminated and core plate length was controlled within ± 0.003 in. The finish was greatly superior to that produced by hot milling.

Notching and breaking requires secondary milling steps to machine the broken surfaces flat and parallel. Chamfering is required and two edges must now be machined even with the saw-cut lengths. A stacked milling cutter, mounted on a vertical mill, was assembled that simultaneously chamfered both corners and milled the edge of the plate. The core plate was placed in a fixture that allowed one end and one longitudinal edge to be milled at a single setup. After the first operation the core plate was rotated 180° and the other edge was end milled. Weight is adjusted during the milling of the final edge.

Successful assembly of the fuel element in the jacket sleeve depends upon (i) thorough cleaning of the core plates to remove loose contamination, (ii) a very low contamination level at the assembly station and on the tools used to handle jacket components, (iii) protection of the lip of the jacket from contact with the core, (iv) deliberate but slow insertion of the core to prevent transfer of contaminants with the displacement of the atmosphere from the jacket, and (v) careful handling of the assemblies until welding. Wire brushing, acetone wiping, and liquid nitrogen scrubbing were tested to clean the core plates. It appears that a combination of all three methods is required to clean the core plate surfaces sufficiently. It has been necessary to flare the open end of the jacket slightly to insert the funnel. This flare is removed when the fuel elements are clamped in welding fixture. The TIG weld technique if properly controlled is adequate for making the closure welds.

4. Supplemental U-5 w/o Fs Alloy Fuel Pins for EBR-II

A production run of 3133 inspected EBR-II fuel pins was completed, and the fuel pins were shipped to Idaho. The use of inert-atmosphere, gloveboxed injection-casting, pin-processing and inspection equipment in Building 350 saved the expense and lost time of setting up an enriched uranium fabrication line and security area, but had the slight disadvantage of glovebox fabrication and plutonium contamination of the fuel pins.

The injection casting furnace had a maximum capacity of 78 Vycor molds and approximately 8 kg of charge weight. Alloy billets weighing 6.0 to 6.5 kg were prepared in the CEN melt refining furnace and were remelted with 1.5 to 2.0 kg of return scrap. Some difficulty was experienced in melting sheared pin ends with the crucible heels and billets, and after the first attempts, these were accumulated and melted in a perforated bottom "strainer" crucible to separate adherent mold wash and oxide. The clean metal was solidified in a mold and the resulting billet was returned to injection casting.

The castings were stripped from their molds, brushed to remove loose mold wash, and visually inspected for surface pits or defects. The 13.5-in.-long fuel rods were sheared from the 17.5-in.-long castings by means of carbide dies in a rod parter. Rod length was indicated by a dial comparator. The rods were fed through a three-contact roller gauge which indicated maximum and minimum diameter. Average diameter was estimated by observing the dial indicator as the rods were passed through the gauge. The rods were weighed on a substitution-type analytical balance (Mettler). Sheared rods and crucible heels were also weighed to balance out the melt.

The inspected fuel rods were placed in a vinyl bandolier and sealed from the glovebox in a large vinyl envelope. They were radiographed by a 4-Ci Co⁶⁰ source through the envelope and bandolier.

Fuel pins showing internal defects larger than 1/16 in. in any direction were rejected, although some of these were sent to Idaho for use in calibration of the remote eddy-current flow detector along with the radiographs. After radiography and removal of reject fuel pins, the vinyl envelope, bandolier, and accepted fuel pins were rolled and sealed in 18-in.-long food-pack cans. The sealed cans were packed in birdcages for shipment to Idaho.

5. Corrosion

a. Fuel-cladding Materials for Sodium-cooled Reactors. Exposure of V-20 w/o Ti in sodium at 650°C containing 3.8 ppm oxygen for times up to 32 hr has been completed. Samples were rotated at 50 rpm to insure access of oxygen to the surface. The oxidation was parabolic with a rate constant of $6.2 \times 10^{-4} \text{ (mg/cm}^2\text{)}^2 \text{ hr}^{-1}$. A previous experiment at the same temperature in sodium containing 7.5 ppm oxygen and with the samples rotated at 5 rpm yielded a rate constant of $30 \times 10^{-4} \text{ (mg/cm}^2\text{)}^2 \text{ hr}^{-1}$. Oxygen concentration is measured by the vacuum distillation method.

It is physically unrealistic to attribute the lower rate to the higher velocity. One would expect either no velocity dependence or an increase in the oxidation rate with increase in velocity if transport of oxygen through the liquid metal were controlling the overall reaction rate.

It appears evident that in the range of variables studied, corrosion rate is much more sensitive to small changes in oxygen concentration than to relatively large changes in velocity. This conclusion regarding the sensitivity to oxygen concentration is in agreement with previous indications concerning the dependence of corrosion rate on oxygen concentration at low values of oxygen concentration. The apparatus will be modified to allow more predictable control of oxygen concentration.

b. Refractory Metal Alloys for Service in Oxygen-contaminated Sodium. A refreshed autoclave test of V-20 w/o Ti in 650°C static sodium has been completed. The oxygen content was determined by the mercury amalgamation method. The internal sample penetration and external sample loss are shown in Table V.

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

Time (days)	Oxygen in Sodium (ppm)	Sample Change per Surface	
		External Loss (μ)	Internal Penetration (μ)
7.0	15	10	38-42
14.0	15	20	49-55
21.1	14	41	45-58
28.1	16	52	44-57
35.1	16	62	45-60
42.1	12	83	50-65

The oxygen level was chosen based on a single analysis of EBR-II sodium at 13 ppm. The V-20 w/o Ti forms a relatively constant hardened zone after the first week. A plot of the total penetration versus time gives a slope of $\sim 2.4 \mu/\text{day}$ (assuming linear kinetics) and an intercept on the penetration axis of $\sim 30 \mu$. If the total penetrations at one week are compared at various oxygen levels (48-52 μ at 15 ppm, 67-62 at 20-25 ppm, and 86-92 at 45-50 ppm), it will be obvious that the alloy is extremely sensitive to oxygen concentration. This conclusion is supported by the work of H. A. Levin (see previous section) in carefully deoxygenated sodium where very little attack is noted.

An alloy of V-20 w/o Cr-10 w/o Al has been shown to have significantly better oxidation resistance than V-20 w/o Ti to 650°C sodium containing 25-30 ppm oxygen, but it is difficult to fabricate. Attempts to modify its fabrication properties with small additions of zirconium or yttrium were not successful, so significant amounts of titanium were added

TABLE VI. Corrosion of V-20 w/o Cr-10 w/o Al-Ti Alloys

Titanium in Alloy (w/o)	Weight Loss (mg/cm^2)
0	1.54
5	2.15
10	1.64
15	2.77
20	3.13
V-20 w/o Ti	7.14

to the basic composition for the same purpose. A one-week corrosion test of these modified alloys was performed in 650°C static refreshed sodium with about 16 ppm oxygen. As shown in the preliminary results of Table VI, all of the modified alloys lost more weight than the V-20 w/o Cr-10 w/o Al but less than V-20 w/o Ti.

The dynamic system has been repaired and V-20 w/o Ti is being tested in 650°C sodium flowing at 6.1 m/sec. The oxygen level of the system is being maintained by a bypass flow through a cold trap held at 120°C.

6. Irradiation Testing

a. Plutonium-alloy Fuels. Instrumented capsules ANL-55-5 and ANL-55-6 were irradiated in the MTR. ANL-55-5 contained one full-length, EBR-II-type fuel rod (C102), and ANL-55-6 contained three fuel rods (C94, 95, and 96) of the same type. On the basis of thermocouple data the central fuel temperature in C102 was calculated as ranging from 555 to 565°C and the maximum cladding temperature as 485 to 500°C. The central fuel temperature for the other three rods was similarly calculated at 550°C and the cladding temperature as 505°C.

Each of the four fuel rods contained a fuel pin of 0.144-in.-dia and 14.21-in.-long U-20 w/o Pu-10 w/o Fs. The fuel pins were clad with stress-relieved Nb-1 w/o Zr tubing, of 0.174-in. OD, 17.5 in. long, and with a 9-mil wall. A sodium bond facilitated heat transfer from the fuel to the cladding. The only obvious difference among these rods was a restrainer-type top plug on C96.

Samples from C102 have been analyzed for technetium. Burnups determined from the analyses ranged from 1.8 a/o of the heavy metal atoms at the bottom to 1.4 a/o at the top of the fuel pin. Analyses on the other three pins are being made. Except for the effect of local perturbations they are expected to be similar to the analyses on C102.

As reported previously, C102 had two areas of cladding failure through which fuel had emerged on the upper third of the length of the pin. In addition, the tube had split from the lower of the two eruptions down to about one inch from the bottom of the rod. C95 also had a split jacket on the lower half of its length. C94 and 96 were intact.

Volume changes on fuel samples from each of the four rods were determined by the method of displacement in toluene. The results are shown in Table VII. In C102 the fuel eruptions occurred between Samples No. 7 and No. 16. Presumably data obtained in this area would have shown much higher volume increases than the highest value obtained for this fuel pin. Indications are that the fuel had expanded 30% or more almost all along its length. This is just about the maximum permitted by the annulus between the fuel and the cladding, considering only radial expansion.

TABLE VII. Volume Increases along Irradiated Fuel Pins
of U-20 w/o Pu-10 w/o Fs

Capsule Number	Fuel Rod	Sample Number	Length of Sample (in.)	Distance from Bottom of Fuel Pin to Centered Sample (in.)	% Volume Increase
ANL-55-5	C102	1	$1\frac{1}{8}$	$9\frac{1}{16}$	17.3
		4*	$3\frac{1}{8}$	$3\frac{11}{16}$	35.6
		7**	$2\frac{1}{16}$	$7\frac{5}{16}$	36.3
		16	$3\frac{1}{16}$	$13\frac{1}{8}$	<u>29.7</u>
					29.7
ANL-55-6	C94 (Intact)	40	$1\frac{1}{2}$	$1\frac{5}{8}$	5.1
		41	$1\frac{1}{2}$	$3\frac{1}{4}$	3.6
		42	$1\frac{1}{2}$	$5\frac{5}{8}$	6.6
		43	$1\frac{1}{2}$	$8\frac{1}{8}$	5.5
		44	$1\frac{1}{2}$	$10\frac{1}{2}$	<u>4.9</u>
					5.1
	C95	34	$1\frac{1}{2}$	$1\frac{1}{4}$	23.5
		35	$1\frac{1}{2}$	$3\frac{5}{8}$	24.6
		36	$1\frac{1}{2}$	$5\frac{1}{4}$	10.6
		37	$1\frac{1}{2}$	$7\frac{3}{4}$	8.2
		38	$1\frac{1}{2}$	$9\frac{3}{4}$	3.8
		39	$1\frac{1}{2}$	$12\frac{1}{4}$	<u>4.4</u>
					12.5
	C96 (Intact)	28	$1\frac{1}{2}$	$1\frac{1}{4}$	6.3
		29	$1\frac{1}{2}$	$3\frac{1}{4}$	6.2
		30	$1\frac{1}{2}$	$5\frac{1}{4}$	7.0
		31	$1\frac{1}{2}$	$7\frac{1}{4}$	7.2
		32	$1\frac{1}{2}$	$9\frac{3}{4}$	5.2
		33	$1\frac{1}{2}$	$11\frac{5}{8}$	<u>4.7</u>
					6.1

*Sample broke into four pieces; % volume change is a weighted average.

**Sample broke into two pieces; % volume change is a weighted average.

In C95 the tube split toward the bottom, and it is at the bottom that the greatest measured volume increases occurred. The top two-thirds of this pin showed only moderate volume increases.

C94 and C96 had relatively low and quite uniform volume increases for the fuel.

The fuel rods described above are based on early fuel element designs and materials selections. As has been noted in previous progress reports, work on U-Pu-Fs alloys is being phased out in favor of the improved alloy U-Pu-Zr. Also, the use of somewhat thicker cladding than 0.009 in. has been found to lead to much higher burnups without jacket rupture.

A total of 41 specimens are under irradiation as shown in Table VIII. The specimens being irradiated in CP-5 are from 4 to 6 in. long and are in five instrumented capsules. The specimens in EBR-II are full length (18 in.) and are located in three special subassemblies.

TABLE VIII. Status of Metal Irradiations in Progress

Test Reactor	Capsule or S/A No.	Design Parameters						Operating Conditions			
		Fuel Composition (w/o)	Effective Density (%)	U + Pu atoms per cc x 10 ^{-22(a)}	Cladding Composition (w/o)	Cladding OD (in.)	Cladding Thickness (in.)	kW/cc(a)	Maximum Cladding Temperature (°C)	Burnup to Date	
										a/o (U + Pu)	fiss/cc x 10 ^{-20(a)}
CP-5	N-15	U-19 Pu-14 Zr	66	2.17	V-20 Ti	0.208	0.015	2.6	620	6.3	13.7
EBR-II	XG06	U-15 Pu-10 Zr	66	2.40	V-20 Ti	0.209	0.016	1.7	540	1.5	3.6
EBR-II	XG05	U-15 Pu-10 Zr	66	2.40	V-20 Ti	0.209	0.016	1.7	535	1.4	3.4
EBR-II	XA07	U-15 Pu-9 Zr	75	2.80	304 SS	0.208	0.021	2.0	650	1.1	3.1
EBR-II	XA07	U-15 Pu-9 Zr	75	2.80	304 SS	0.208	0.021	2.0	650	1.1	3.1
EBR-II	XA07	U-15 Pu-9 Zr	75	2.80	316 SS	0.196	0.015	2.0	645	1.1	3.1
EBR-II	XA07	U-15 Pu-9 Zr	75	2.80	Hast.-X	0.196	0.015	2.0	645	1.1	3.1
CP-5	L-18	U-15 Pu-12 Zr	75	2.56	Hast.-X	0.196	0.015	1.9	490	0.73	1.9
CP-5	F-35	U-15 Pu-12 Zr	75	2.56	304 SS	0.196	0.015	1.9	490	0.73	1.9
CP-5	F-37	U-15 Pu-12 Zr	75	2.56	316 SS	0.196	0.015	1.9	490	0.73	1.9
CP-5	L-19	U-15 Pu-12 Zr	75	2.56	Hast.-X	0.196	0.015	1.9	560	0.73	1.9
CP-5	F-36	U-15 Pu-12 Zr	75	2.56	304 SS	0.196	0.015	1.9	560	0.73	1.9
CP-5	F-38	U-15 Pu-12 Zr	75	2.56	316 SS	0.196	0.015	1.9	560	0.73	1.9
EBR-II	XA07	U-14 Pu-12 Zr	76	2.60	304 SS	0.208	0.021	1.9	630	1.0	2.6
EBR-II	XA07	U-14 Pu-12 Zr	76	2.60	304 SS	0.208	0.021	1.9	630	1.0	2.6
EBR-II	XA07	U-14 Pu-12 Zr	76	2.60	316 SS	0.196	0.015	1.8	625	1.0	2.6
EBR-II	XA07	U-14 Pu-12 Zr	76	2.60	316 SS	0.196	0.015	1.8	625	1.0	2.6
EBR-II	XA07	U-14 Pu-12 Zr	76	2.60	316 SS	0.196	0.015	1.9	635	1.0	2.6
EBR-II	XA07	U-14 Pu-12 Zr	76	2.60	316 SS	0.196	0.015	1.9	635	1.0	2.6
EBR-II	XA07	U-14 Pu-12 Zr	76	2.60	Hast.-X	0.196	0.015	1.9	630	1.0	2.6
EBR-II	XA07	U-14 Pu-12 Zr	76	2.60	Hast.-X	0.196	0.015	1.9	635	1.0	2.6
EBR-II	XA07	U-14 Pu-12 Zr	76	2.60	Hast.-X	0.196	0.015	1.9	640	1.0	2.6
EBR-II	XA07	U-14 Pu-12 Zr	66	2.50	Hast.-X-280	0.208	0.015	1.8	635	1.0	2.5
EBR-II	XA07	U-14 Pu-12 Zr	66	2.50	Hast.-X-280	0.208	0.015	1.8	635	1.0	2.5
EBR-II	XA07	U-14 Pu-12 Zr	66	2.50	Hast.-X-280	0.208	0.015	1.8	635	1.0	2.5
CP-5	N-14	U-15 Pu-10 Ti	69	2.30	V-20 Ti	0.203	0.015	2.4	580	5.3	12.9
EBR-II	XG05	U-15 Pu-10 Ti	63	2.21	V-20 Ti	0.209	0.016	1.6	540	1.4	3.1
EBR-II	XG06	U-15 Pu-10 Ti	63	2.21	V-20 Ti	0.209	0.016	1.6	540	1.5	3.3
CP-5	N-10	U-10 Pu-10 Fz	77	2.94	V-20 Ti	0.196	0.016	1.9	530	5.8	17.1
CP-5	N-11	U-10 Pu-10 Fz	79	3.01	V-20 Ti	0.193	0.016	2.0	530	5.8	17.5
CP-5	N-12	U-10 Pu-10 Fz	84	3.2	V-20 Ti	0.189	0.016	2.1	530	5.8	18.6
CP-5	TC-1	U-15 Pu-10 Fz	57*	2.17	Nb-1 Zr	0.174	0.009	1.5	500	2.9	6.3
CP-5	C-152P	U-15 Pu-10 Fz	58*	2.20	Nb-1 Zr	0.174	0.009	1.5	500	2.9	6.4
CP-5	C-153P	U-15 Pu-10 Fz	59*	2.24	Nb-1 Zr	0.174	0.009	1.5	500	2.9	6.5
CP-5	C-155P	U-15 Pu-10 Fz	58*	2.20	Nb-1 Zr	0.174	0.009	1.5	510	2.9	6.4
CP-5	C-156P	U-15 Pu-10 Fz	61*	2.31	Nb-1 Zr	0.174	0.009	1.5	510	2.9	6.7
CP-5	C-159P	U-15 Pu-10 Fz	59*	2.24	Nb-1 Zr	0.174	0.009	1.5	510	2.9	6.5
CP-5	N-7	U-15 Pu-10 Fz	77	2.94	V-20 Ti	0.196	0.016	1.9	530	5.8	17.1
CP-5	N-8	U-15 Pu-10 Fz	79	3.01	V-20 Ti	0.193	0.016	2.0	530	5.8	17.5
CP-5	N-9	U-15 Pu-10 Fz	84	3.21	V-20 Ti	0.189	0.016	2.1	530	5.8	18.6
CP-5	N-13	U-15 Pu-10 Fz	73	2.79	V-20 Ti	0.199	0.015	2.3	550	4.9	13.7

*Vibratory compacted powders.

(a)Based on effective density.

b. Ceramic Fuel Irradiations. A subassembly containing three mixed carbide specimens and two mixed oxide specimens was loaded into EBR-II during this report period. The carbide specimens contain pellets, vibratorily compacted physically mixed UC-20 w/o PuC, and vibratorily compacted solid-solution ($U_{0.8}Pu_{0.2}$)C. The oxide specimens contain vibratorily compacted Nu-Pak UO_2 -20 w/o PuO_2 . The reactor is expected to exceed 90 MWd this period after a sustained shutdown. Table IX contains a summary of all ceramic irradiations in the reactor and their anticipated status at the end of the report period.

TABLE IX. Status of Ceramic Fuel Irradiations in EBR-II

Capsule or S/A No.	Design Parameters						Operating Conditions			
	Fuel Composition (w/o)	Effective Density (%)	U + Pu atoms per cc $\times 10^{-22}$ (a)	Cladding Composition (w/o)	Cladding OD (in.)	Cladding Thickness (in.)	kW/cc ^(a)	Maximum Cladding Temperature (°C)	Burnup to Date	
									a/o, (U + Pu)	fiss/cc $\times 10^{-20}$ (a)
XG-05	UC-20 w/o PuC	84	2.78	304 SS	0.297	0.020	2.13	645	1.44	4.00
XG-05	UC-20 w/o PuC	80	2.64	Hast.-X	0.297	0.015	2.20	670	1.47	3.88
XG-05	($U_{0.8}Pu_{0.2}$)C	84	2.78	Nb-1 Zr	0.281	0.012	2.06	645	1.49	4.14
X-008	($U_{0.8}Pu_{0.2}$)C	82	2.71	Nb-1 Zr	0.281	0.012	1.64	545	0.36	0.98
X-008	UC-20 w/o PuC	80	2.64	Nb-1 Zr	0.281	0.012	2.43	635	0.52	1.38
X-008	UC-20 w/o PuC	80	2.64	Nb-1 Zr	0.281	0.012	2.13	605	0.48	1.26
X-008	($U_{0.8}Pu_{0.2}$)C	86	2.81	Nb-1 Zr	0.281	0.012	2.43	635	0.52	1.47
X-008	UC-20 w/o PuC	80	2.64	Hast.-X	0.297	0.015	2.07	640	0.47	1.24
X-008	UC-20 w/o PuC	80	2.64	Hast.-X	0.297	0.015	2.28	670	0.52	1.36
X-008	($U_{0.8}Pu_{0.2}$)C	82	2.64	Hast.-X + W	0.297	0.020	1.58	555	0.36	0.94
X-008	($U_{0.8}Pu_{0.2}$)C	83	2.75	Hast.-X + W	0.297	0.020	2.36	685	0.52	1.43
X-009	($U_{0.8}Pu_{0.2}$)C	82	2.70	316 SS	0.306	0.024	1.60	570	0.04	0.09
X-009	UC-20 w/o PuC	80	2.64	316 SS	0.306	0.024	2.10	640	0.05	0.12
X-009	($U_{0.8}Pu_{0.2}$)C	86	2.81	Vanadium	0.301	0.022	2.53	640	0.05	0.15
X-009	UO_2 -20 PuO_2	82	2.05	304 SS	0.297	0.020	1.66	555	0.05	0.10
X-009	UO_2 -20 PuO_2	82	2.05	304 SS	0.297	0.020	1.69	565	0.05	0.10

(a) Based on effective density.

7. Powder-loaded, Radiator-type Fuel Elements

The purpose of this program is to find the factors influencing the economical and simplest method of fabricating entire fuel-element assemblies. Particulate fuels were chosen as the basis for the study. To conserve on costly compounds of uranium and plutonium and to simplify handling requirements, preliminary experiments were carried out with blast-cleaning shot and grit. A three-part study of the packing characteristics of spherical and angular particles has resulted from this interest. During the reporting period the third part of the study, the factors influencing rate of compact formation, has been completed.

The experiments with spherical particles showed that an infiltration technique was a solution to separation of particles of different size caused by the levitation of the larger size by the smaller component. This technique, considered to be tantamount to the uniform distribution of particles in a column, was employed in the rate-of-entry study. It was found that

the rate of entry of particles into a matrix of larger particles was influenced by frequency of vibration, acceleration, the void fraction in the matrix, column height, and ratio of sizes of entering to matrix particles. The specifics of the study are as follows:

a. Driving Force. The rate of entry of particles of constant diameter ratios into a matrix of fixed void fraction was found to respond to variations in the frequency and acceleration of impressed vibration. Infiltration time for both spherical and angular particles decreased with increasing acceleration to about 15 g. Little difference in the time was observed between 15 and 20 g, suggesting that a plateau is reached in the rate of infiltration between these two values. The influence of frequency on infiltration time showed a minimum, for all accelerations to 20 g, between 40 and 80 cps. (This fact had been suggested by screening experiments and 60-cycle vibration has been used in the fabrication of fuel columns of carbides and oxides for irradiation testing.) The minimum is depressed as acceleration increases, but is almost absent in the case of spherical particles. In all cases, at driving frequencies between 100 and 400 cps, infiltration time increased with increasing frequency.

b. Column Characteristics. The infiltration time of a smaller component varies inversely with the void fraction in the matrix. The relationship varies exponentially with the void fraction. In simple terms this implies that as the void fraction becomes small, the void size becomes small and the entering particle must be allowed additional time to seek out and fill the increased number of smaller holes.

The rate of infiltration is also influenced by the ratio of the sizes of infiltrating to matrix particles. Actually the relationship should be governed by the ratio of the size of the infiltrating particle to that of the void but, since the voids are connecting and continuous and depend for their size upon the matrix particle size, the former ratio is preferred. The influence in both spherical and angular particles is somewhat hyperbolic. When the ratio of particle sizes is a minimum, below which infiltration cannot proceed, the rate is low. As the ratio increases, infiltration becomes faster until a maximum is reached. For spherical particles the minimum ratio of small to larger component to allow infiltration is about five; for angular particles this value is about seven. The maximum infiltration rate for spheres is reached when the size ratio is greater than 11. Angular particles respond to increases in size ratio above 18.

Finally, the influence of column height upon infiltration rate was studied. This was done to gain some insight into the mechanism of "hole filling." Our findings indicate that a nearly straight-line relation between column height and infiltration time exists for the first two-thirds of the time required. This would indicate that at first the column fills as water, flowing at a constant rate, fills a vessel. This is, of course, an

oversimplification neglecting, among other factors, the resistance to flow offered by the matrix particles. For the last third of the filling time the rate changes while the local particles settle into voids not occupied during the initial and incomplete filling period. During this latter period we have observed that about 80% of the void space had been filled and the additional time is required to infiltrate the more inaccessible voids.

8. Particulate Fuel Test Specimens--Oxide

Twenty-four UO_2 - PuO_2 fuel rods in the form of a vibratorily compacted matrix of angular particles are being fabricated at the request of the Engineering Irradiation Group.

The fuel rods are being loaded with UO_2 -20 w/o PuO_2 which had been processed at Battelle-Northwest by the Nu-Pak consolidation technique and then powdered. The rods are jacketed in Type 304 stainless steel, Hastelloy-X, or V-20 w/o Ti (TV-20), and have either a rod-type or spring-type fuel retainer. Fuel densities of 80 to 82, 83 to 85, and >85% of theoretical are desirable. At this time twenty-one rods have been fabricated and delivered. Table X is a resumé of the pertinent characteristics of the completed fuel specimens.

TABLE X. Vibratorily Compacted UO_2 - PuO_2 Fuel Rods for Irradiation

Specimen Number	Fuel Weight (g)	Fuel Volume (cc)	Packing Fraction (%)	Theoretical Density (%)	Jacket Material	Retainer Form
SOV-1	85.045	9.71	81.9	79.9	304 SS	Rod
SOV-2	85.053	9.72	81.8	79.8	304 SS	Rod
SOV-3	88.546	9.72	85.2	83.1	304 SS	Rod
SOV-4	87.317	9.70	84.2	82.1	304 SS	Rod
SOV-5	88.647	9.70	85.5	83.4	304 SS	Rod
SOV-6	88.611	9.71	85.4	83.3	304 SS	Rod
SOV-7	90.704	9.72	86.8	85.1	304 SS	Rod
SOV-8	90.554	9.77	86.2	84.5	304 SS	Spring
SOV-9	91.002	9.72	87.1	85.4	304 SS	Rod
SOV-10	84.740	9.73	81.4	79.4	304 SS	Spring
SOV-11	85.355	9.74	81.9	79.9	304 SS	Spring
HOV-1	92.201	10.51	81.7	80.0	Hast.-X	Spring
HOV-2	92.173	10.52	81.6	79.9	Hast.-X	Rod
HOV-3	92.200	10.52	81.6	79.9	Hast.-X	Spring
HOV-4	92.213	10.51	81.7	80.0	Hast.-X	Rod
HOV-15	91.749	10.48	81.8	79.8	Hast.-X	Rod
HOV-6	92.177	10.50	82.2	80.1	Hast.-X	Rod
HOV-8	97.814	10.50	86.7	85.0	Hast.-X	Spring
HOV-9	97.585	10.51	86.4	84.7	Hast.-X	Rod
HOV-10	97.702	10.50	86.5	84.8	Hast.-X	Rod
HOV-11	92.045	10.52	81.8	79.8	Hast.-X	Rod

Three rods jacketed in TV-20 with spring retainers will complete this series of fabricated elements. The preparation of the rods is complete and the actual loading is imminent. The tabulated rods were loaded by an infiltration technique with emphasis placed on fuel distribution and density control. The loading of the fine fraction in the last three rods will emphasize speed to correlate rate-of-entry studies with actual fuel-loading conditions.

9. Doppler-element Loading Techniques

Thirty-two capsules containing plutonium oxide were fabricated during 1965 by the Metallurgy Division for Doppler effect experiments by Reactor Physics Division. Additional Doppler elements are required.

The Doppler capsules previously supplied contained glass-bonded pellets. Glass-bonding was required to meet the specification that the fuel body be made up of low-density cored pellets. These bodies were designed to be easily constrained to provide constant volume upon heating.

All necessary conditions may be met with columns of particles. To test the hypothesis, five capsules were loaded and supplied for testing. The contents of the five capsules are described in Table XI. It appears from the tests performed to date that a column of granules will meet the requirements for Doppler specimens. If so, the number of steps and man-hours required to fabricate such elements will be reduced to about two-thirds of that required for preparation of the glass-bonded pellets.

TABLE XI. Contents of Doppler Capsules

Capsule Number	Feed Material			Charge (g)	Performance
	Composition	Source	Size		
N-12	Natural UO_2	Crushed High-density Pellets	-14 $\pm$ 25	983.7	Unsatisfactory--capsule showed excessive void during constraint.
N-13	Natural UO_2	Crushed High-density Pellets	-25 $\pm$ 50	948.6	Satisfactory.
N-14	Natural UO_2	Crushed High-density Pellets	-50 $\pm$ 100	877.5	Excellent.
N-15	Natural UO_2	Mallinckrodt Powder	-325	589.5	Test not complete to date.
HE-5	Enriched UO_2	Fired Granules	-50 $\pm$ 100	813.68	Test not complete to date.

D. General Fast Reactor Fuel Reprocessing Development

1. Skull Reclamation Process

The skull reclamation process is being developed in order to recover and purify the uranium-bearing residue or skull that is left in the crucible following the melt refining of enriched uranium alloy fuel pins discharged from the EBR-II reactor. The uranium in the skull amounts to 5 to 10% of the original uranium charge. The chemistry and technology that are being accumulated in the course of this process development also have considerable potential utility for the processing of plutonium-uranium oxide and carbide fuels. Currently, the skull-reclamation process is being tested in remotely operated plant-scale equipment (~4 kg of uranium per batch) comprising a reduction furnace and a retorting furnace.

Three additional runs (SRR-15 through SRR-17) with nonradioactive uranium-bearing charges have been completed in the reduction furnace. The processing of the skull oxides in the furnace consists of the following three steps: (1) removal of the noble metal fission products, (2) reduction of skull oxides to uranium metal and removal of essentially all the rare earth, alkali, and alkaline earth fission products, and (3) dissolution of the uranium in zinc-magnesium, which product solution is the feed to the subsequent retorting operation. In these runs, which are part of a new series, the noble metal-extraction step and the uranium oxide-reduction step were conducted under more vigorous reaction conditions (higher temperatures, longer contact times, and faster mixing) than in earlier runs (see Progress Report for December 1965, ANL-7132, pp. 23-24). The noble metal-extraction step was carried out for 2 hr at 750°C. The reduction step was conducted for 2 hr, of which 3/4 hr was required to heat the charge from 700°C to the operating temperature of 800°C. Mixing speed in both steps was 800 rpm.

In the three recent runs the effect of fluoride ion in the salt on fission product decontamination in the noble metal-extraction step and on the reduction of uranium oxides in the reduction step was again studied. In these runs, 10 m/o fluoride ion (as NaF) was present in the salt used for the noble metal-extraction step. In runs SRR-11 through SRR-14, in which fluoride ion was added just prior to the reduction step, high fission product element removals and excellent (~99%) uranium oxide reductions were obtained (see Progress Report for January 1966, ANL-7152, p. 42).

Uranium oxide reductions exceeding 99% were achieved in the three runs, which confirmed the beneficial effect of fluoride ion in the reduction step. Data on the fission product removals for these runs are not yet available. Overall fission product removals will be determined by analyses of the uranium buttons obtained after retorting the zinc-magnesium-uranium product solutions.

The final step in the skull-reclamation process involves retorting, or solvent evaporation, of the Zn-Mg alloy to recover the product uranium from the Zn-12 w/o Mg-12 w/o U ingot cast from the product solution that is transferred out of the reduction furnace. Previous retorting runs have shown that the product uranium button will be contaminated with oxide unless the residual flux that accompanies the Zn-Mg-U solution during its transfer from the reduction furnace is removed before retorting (see Progress Reports for January and February 1966; ANL-7152, p. 43 and ANL-7176, p. 35). In previously reported small-scale tests, Fiberfrax strainers were successfully used to remove salt flux from molten Zn-Mg alloy by pouring the material through the strainer at temperatures ranging from 600 to 775°C (see ANL-7176, p. 35). The flux preferentially wetted the strainer and was retained, while the clean metal passed through. Full-scale strainers were successfully tested in the recently completed reduction furnace runs. The Zn-Mg-U product solution, after being poured through the strainer, which was mounted on the graphite product mold, appeared essentially free of flux. Visual examination indicated no holdup of product metal in the strainer.

2. Processes for Future Fast Reactor Fuels

Compact pyrochemical processes are being developed for processing fast breeder reactor fuels of the ceramic (e.g., oxide or carbide) type. Salt-transport processes are currently being investigated. In these processes, the fissile and fertile constituents of a reactor fuel are selectively transferred from one liquid metal solvent (donor) to another (acceptor) by cycling a molten salt phase, which acts as a carrier, between the two metal solvents. The salt-transport processes have the inherent advantage over other pyrochemical processes that only small solution volumes are required, since not all of the uranium or plutonium needs to be in solution at any one time. The salt-transport processes also offer the possibility of excellent separations of fission products from uranium and plutonium and excellent recoveries of uranium and plutonium.

A process based on the selective transport of uranium and plutonium from a Cu-Mg solvent alloy to appropriate Zn-Mg alloys through a chloride salt phase is currently under investigation. A conceptual flowsheet³ was presented in a previous report (see Progress Report for September 1965, ANL-7105, p. 34).

³ The flowsheet involves first an oxidative decladding operation in which oxide fuels are released from cladding. Carbide and metal fuels are converted to oxides in such a step. These oxides are then reduced by a 90 w/o Cu-Mg alloy. The highly electropositive fission elements (alkali, rare earth, and alkaline earth elements) distribute strongly to the salt phase and are removed in it. By taking advantage of the widely different distribution coefficients and solubilities for uranium and plutonium in metal phases of different compositions, the uranium and plutonium can then be separated by selective transfer to separate metal phases. Transfer of uranium and plutonium can be done successively or simultaneously; the latter is shown schematically in the referenced flowsheet.

In the oxide-reduction step of the process, the oxides are suspended in a molten salt which is then agitated in the presence of a liquid magnesium alloy. By this procedure the fissile element oxides are reduced and dissolved in the liquid alloy. Since the molten salt is a waste stream, it is desirable to achieve complete oxide reduction and to minimize the concentration of dissolved fissile element compounds. These two requirements depend on the magnesium chloride concentration of the molten salt in opposite ways; thus a high MgCl_2 concentration favors rapid oxide reduction whereas a low MgCl_2 concentration is required to obtain low concentrations of dissolved fissile compounds and thereby minimize losses. Therefore, it is necessary to optimize the process step with respect to these conflicting requirements.

One aspect of this problem was investigated in the laboratory by measuring the dependence of the concentration of uranium and cerium in a CaCl_2 - NaCl - MgCl_2 salt mixture on the MgCl_2 concentration when in equilibrium with a liquid copper-magnesium alloy at 800°C . These measurements indicated the following:

(a) The concentration of UCl_3 and CeCl_3 in the molten salt may vary by several orders of magnitude when the concentration of MgCl_2 varies from about 1 to 60 m/o and the composition of the metal phase is constant.

(b) The distribution coefficient (ratio of w/o uranium or cerium in salt to w/o uranium or cerium in metal) is proportional to the three-halves power of the MgCl_2 concentration.

(c) The ratio of uranium to cerium in the salt phase is approximately independent of the MgCl_2 concentration of the salt.

Another method of reducing the overall loss of fissile elements in the oxide-reduction step is to use the same molten salt mixture for several reductions. This would also have the added advantage of reducing the waste volume. To reuse the salt after a reduction it is necessary to separate a major portion of the suspended magnesium oxide which is produced when the magnesium alloy reduces the fissile element oxides. A sample of a typical reduction salt containing suspended MgO was examined. The MgO was found to consist of particles of from 1 to $5\ \mu$ in diameter ($2\ \mu$ average) which were found to settle rapidly. A sharp boundary was observed between the supernatant solution and the precipitate, indicating that decantation might be a feasible technique for the recovery of a salt having a low MgO content. A sample of the salt containing MgO was maintained at 500°C for 6 hr; no significant change in the average particle size was observed.

To lower the operating temperature of the salt-transport process from about 800 to 600°C a Cd-Zn-Mg alloy has been considered (see Progress Report for February 1966, ANL-7176, p. 36) in place of the Cu-Mg donor

alloy indicated in the conceptual flowsheet. To evaluate the Cd-Zn-Mg alloy for this use, measurements were made of the distribution coefficients of uranium, plutonium, and cerium between a 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl molten salt and the liquid ternary alloy in which the cadmium-to-zinc weight ratio was 9 and the magnesium concentration was varied between ~1 and 7 w/o. In these same experiments, the co-solubilities of uranium, plutonium, and cerium in the liquid metal phase were also determined. The results indicate that a 9-to-1 Cd-Zn alloy containing one percent magnesium could be used in the salt-transport process as a donor alloy at 600°C. However, the magnesium concentration of the alloy would have to be closely controlled. The Cd-Zn-Mg alloy was found to be more suitable for use as a donor alloy than a Cd-Mg alloy; the addition of 10 w/o zinc increases the co-solubilities of uranium, plutonium, and cerium without markedly decreasing the distribution coefficients.

Recent engineering-scale experiments were performed in support of the development of equipment for the salt-transport process. In these experiments, the settling behavior of metals in salts was investigated under conditions of process interest.

An engineering-scale demonstration of the salt transport concept was previously reported (see Progress Report for December 1965, ANL-7132, pp. 27-28). In this run, reduction and salt-transport steps were performed at 830°C to recover uranium from an unirradiated charge composed of a mixture of oxides of uranium and selected fission products. Except for a copper-entrainment problem, the results of this run were very encouraging. An excessive amount of copper had been entrained in the transferred uranium; the copper concentration in the uranium was 2.8 w/o of the uranium. The entrainment behavior of copper in a liquid Cu-Mg-U alloy and liquid MgCl_2 system was therefore investigated in a three-part experiment having the following conditions: (a) all metal alloy components were in solution, (b) precipitated copper fines were present, and (c) precipitated copper and uranium fines were present. In each part of the experiment, the metal alloy and salt phases were mixed for 15 min at 650 rpm and the phases were allowed to separate by settling. Salt-phase samples were taken simultaneously from three different depths below the salt surface after various settling times of up to 15 min.

The results of this experiment were as follows:

(a) After a 2-min settling period, no copper was found suspended in the salt for the first case. For the second and third cases, considerable quantities of copper were still suspended in the salt. The presence of precipitated copper fines seemed to increase the time required to separate the metal and salt phases.

(b) In all cases, after a 5-min settling time, there was no evidence of any copper having remained suspended in the salt phase. However, a small equilibrium amount of copper remained dissolved in the salt.

(c) After the first 5 min of settling, little additional decrease in the copper concentration in the salt phase was observed. For the conditions of this experiment, the minimum copper level obtainable appears to be about 0.002 w/o.

The results of this experiment would indicate that the copper entrainment experienced in the demonstration run was too high to have been caused solely by suspended copper, since a 10-min settling time was used. The entrainment was probably a result of metallic copper having been resuspended during the salt transfer. Therefore, the entrained copper metal might well have been in the form of particles or droplets which had settled to the metal-salt interface, but because of the high interfacial tension had remained above the interface.

For the next demonstration run, the reduction step will be modified to avoid the formation of precipitated copper fines, and the transfer line will be modified to eliminate the formation of a vortex at the inlet.

In other engineering work related to salt-transport processes, information on the effect of mixing speed on entrainment of the metal phase in the salt phase was obtained in two experiments. These data will be useful for designing continuous, differential-type extraction equipment, such as a circulating-salt loop or a system in which a small crucible is suspended within a large crucible. This crucible-within-a-crucible technique, which was described in Progress Report for December 1965, ANL-7132, pp. 25-26, was used in the two experiments. Each experiment utilized a different liquid metal-salt system; one system was 60 w/o Mg-Zn alloy and MgCl_2 -30 m/o NaCl-20 m/o KCl, and the other system Cu-Mg-U and MgCl_2 . The metal and salt phases were mixed at various speeds for 10 min. After the mixing period, the amount of the metal phase found in the small crucible positioned in the salt phase indicated the extent of metal-phase suspension. Excessive entrainment of the metal phase was found at about 650 rpm, the mixing speed used to carry out the extractions in the previously reported demonstration run. However, at mixing speeds below ~500 rpm, low entrainment of the metal phase was observed. Additional experiments are planned to determine if the extraction kinetics are satisfactory at the lower agitation speeds and if de-entrainment devices can be employed effectively at the higher mixing speeds.

A number of equipment components, such as valves, pumps, flowmeters, liquid-level detectors, and pressure-measuring devices, are required for processes employing liquid metal and liquid salt systems.

A test loop has been assembled for the testing of such equipment (see Progress Report for July 1965, ANL-7082, p. 28). A centrifugal pump, an orifice flowmeter, two types of pressure sensors, a freeze valve, and a bellows-sealed throttling valve have been installed in the loop for evaluation of their performance. The loop was operated for 60 hr during which liquid 50 m/o MgCl_2 -30 m/o NaCl -20 m/o KCl at 500°C was circulated at an average flow rate of 0.9 gpm. Operation of the loop was satisfactory. The freeze valve was opened and closed six times without difficulty to permit filling and emptying of the loop. Following this initial operating period, the loop was shut down to carry out minor modifications in the auxiliary equipment, after which the loop will be placed in operation again.

E. Sodium Technology

1. Component and Materials Loop (CAMEL)

Construction of this loop, originally known as the Components Surveillance Loop, has been continued to the point where the pipe welding and main closure connections are finished. The loop has been helium leak tested for integrity. Copper bus bars for the electromagnetic pump were fabricated and delivered on site for installation. The circuitry for leak detection and thermocouple location is being prepared and arrangements have been made for the required control instrumentation.

a. Falex Wear Tester. An additional three units are under assembly at the D-206 shop. The essential components for these three units have been fabricated and delivered from the ANL Central Shops.

The results of previous tests with 304 stainless steel and carbon steel drill rod demonstrated the inadequacy of these materials to perform at 1200°F with any substantial bearing load. The tests have been extended to include the Carbolloys in contact with Stellite. The first tests are still directed at a screening selection for the best materials, and these tests, therefore, involve the exposure of the materials to test under an inert atmosphere at 1200°F . It was demonstrated in this period that the Carbolly samples can be fastened to the test shaft by means of induction brazing with Nicrobraz alloy. Samples are now being prepared to test.

2. Static Tank Facility (STATE)

A conditional acceptance of the pit in D-206 was made from the contractor since all of the construction has been completed with the exception of the cover grid delivery. The cover grids will be installed as soon as they are delivered to the contractor. In the meantime, preparations for the installation of the tank and thimbles continue. The 200-gpm pump in D-308 is to be transferred to D-206 and prepared for installation in the circuit external to the tank. Layouts have been made for the mounting of the components and instruments are being selected.

3. Dynamic Component Test Loop (DYCOTL)

The 2-in. line, inlet and outlet, of the cold trap system to the main loop is completely welded, radiographed, and leak checked. Heaters are installed. Insulation is about 50% completed.

About 85% of the line heaters on the main sodium circuits are installed and connected to a source of power. There is a shortage of Teflon-covered wire to complete the electrical work to connect the balance of heater circuits to the control panel.

The mechanical-pump electrical wiring was checked for polarity and direction of rotation. A tachometer located on the control panel registered 300 rpm with Dynamatic control reduced to lowest speed. There was no indicated vibration or mechanical interference. This pump was shipped to ANL after dynamic water-flow tests at the vendor's plant. There has been no disassembly of motor or coupling from the pump rotor.

Three relay racks have been added to the floor-level instrument panel for strain-gauge readout equipment. About 50% of the strain-gauge wiring is completed, strain gauge to relay rack wire. All strain gauges to pipe (sensing elements) have been completed previously.

The spool-piece insulation has been completed. The flanged instrument thimble nozzles have not been insulated. Separate sheet-metal detachable covers, insulated, for each nozzle are now being fabricated.

The interior of the large pressure vessel is being cleaned. The final electrical connections to the patch board are being made. It is probable that a dry run at low temperatures, to test the heaters and thermocouples, can be initiated soon.

4. Materials Compatibility Test (Small Soaking Pots)

In this test, a ball is forced into an inverted frustum of a cone to test whether or not the ball will stick to the side of the cone at the line-contact area under different loads and at different sodium temperatures. The ball has a diameter of about 1 in., and the cone wall slope is 45° to the base. There are five pairs of balls and cones composed of 304 stainless steel material and each pair is loaded differently. The ball is actually a spherical head of a rod with loads of $11\frac{1}{2}$, 21, 38, 80, and 119 lb applied at the opposite end of the rods. The five cones with the rod ball ends are immersed in a pot of sodium through bellows acting as lids of the pots. The loads are applied outside the pot.

To check the effect or amount of ball-cone sticking, the rod is lifted. The cone is not attached to the base of the pot but is allowed to lift with the ball if there is any adhesion. At a known point, the cone encounters interference and is forced to part from the ball. The measured force is an indication of the amount of sticking.

The five samples, each with a different load, were tested at 400°F, 600°F, 1000°F, and 1200°F. Indication of sticking has been observed at 1200°F condition for the $11\frac{1}{2}$ -lb and 38-lb loads. The other samples are to be checked visually under the microscope. To date, all that can be said is that sticking of pairs has been observed at 1200°F. This work is in the preliminary stage and much more work will be done and reported.

F. General Fast Reactor Component Development

1. Remote Handling Systems and Tools

a. Head-controlled TV for Manipulator Systems. Fabrication of the major components for this new head-controlled television system has been completed. The system will slave a remote camera and a local monitor to follow 5 motions of the manipulator operator's head. This system will be used to evaluate the effectiveness of adding the X, Y, and Z motions to the previously tested pan and tilt motions. The earlier system was briefly reported in the Progress Report for August 1965, ANL-7090, p. 45. It is expected that the assembly of these components will be delayed by the shortage of personnel until about the middle of May.

II. GENERAL REACTOR TECHNOLOGY

A. Experimental Reactor and Nuclear Physics

1. Absolute Measurements of Neutron Sources

With the use of the Manganese Bath Facility,⁴ an independent determination of the standard neutron source NBS-II gave a value of $(1.186 \times 10^6 \text{ n/sec}) \pm 0.7\%$, within the range $(1.180 \times 10^6 \text{ n/sec}) \pm 1.1\%$ quoted by the National Bureau of Standards⁵ and just touching the measured value upper limit of Axton.⁶ The error in the source calibration is composed of a 0.5% rms spread in counting data (two separate counts at two different concentrations about a half year apart), 0.3% error in the manganese/hydrogen absorption correction, and a few 0.1% errors associated with other correction factors, all added in quadrature. There may be some uncertainty in the 0.3% error range associated with the manganese/hydrogen thermal cross-section ratio, because it is based on the optimistic viewpoint that the best values of the cross section can be chosen without fear of unknown effects.

Additional neutron sources were calibrated either by direct comparison with NBS-II (or a similar mode of calculation) or from a series of concentration experiments. The ratio of source intensity is determined by counting in the same solutions and, as with the NBS-II standard, is corrected for differences in neutron-energy spectrum. These ratios are given in Table XII. The consistency of the indicated ratios between various sources (measurements at two concentrations separated by a half year) guarantees some precision and accuracy.

TABLE XII. Neutron Source strength Ratios*

$\frac{\text{Cf}^{252} (1964)}{\text{Cf}^{252} (1965)} = 1.074$	$\frac{\text{NBS-II} (1964)}{\text{Cf}^{252} (1964)} = 0.6608 \pm 1.4\%$	$\left. \begin{array}{l} \\ \end{array} \right\} 0.6617 \pm 0.8\%$
	$\frac{\text{NBS-II} (1965)}{\text{Cf}^{252} (1965)} = 0.6625 \pm 1.0\%$	
$\frac{\text{Ra-}\alpha\text{-Be} (1964)}{\text{Ra-}\alpha\text{-Be} (1965)} = 1.071$	$\frac{\text{Ra-}\alpha\text{-Be} (1964)}{\text{Cf}^{252} (1964)} = 2.929$	
$\frac{\text{NBS-II} (1964)}{\text{NBS II} (1965)} = 1.072$	$\frac{\text{Ra-}\alpha\text{-Be} (1965)}{\text{Cf}^{252} (1965)} = 2.939$	
$\frac{\text{NBS-II} (1964)}{\text{Ra-}\alpha\text{-Be} (1964)} = 0.2256 \pm 1.3\%$	$\left. \begin{array}{l} \\ \end{array} \right\} 0.2255 \pm 0.5\%$	
$\frac{\text{NBS-II} (1965)}{\text{Ra-}\alpha\text{-Be} (1965)} = 0.2254 \pm 0.7\%$		

* Cf^{252} decay corrected to 3/10/64.

⁴DeVolpi, A., Porges, K. G., and Larsen, R. N., Mn⁵⁶ Coincidence Counting Facility, ANL-6790 (Aug 1963).

⁵Spiegel, V., Jr., National Bureau of Standards, Private Communication (July 1965).

⁶Axton, E. J., Cross, P., and Robertson, J. C., J. Nucl. Energy, Parts A/B 19, 409-422 (1965).

All elements of a manganese bath system have been individually tested for accuracy and precision on an individual basis and as a complete system. The system test, which involves a determination of the intensity of NBS-II, supports the accuracy and precision of the individual tests in assigning approximately 0.7% standard error for neutron-source calibrations.

2. High Conversion Critical Experiments (Hi-C)

The analysis of the microparameter measurements made in the 1.349-cm square pitch stainless steel-clad zone Hi-C core has been completed. This core contained a central zone of 400 stainless steel-clad fuel pins surrounded by about 450 aluminum-clad fuel pins. The microparameter measurements were made at a radius of about 5 cm in the 15-cm radius stainless steel-clad central zone.

A value of 0.518 ± 0.006 for the initial conversion ratio (ICR) was obtained by coincidence counting with metal foils. A value of 0.225 was used for α^{25} to compute the value of the ICR.

As in previous cores, the thermal activation method was used to determine the U^{238} capture cadmium ratio C_{28} . Three runs were made with dysprosium as the intermediate material. An average value

$$C_{28} = 1.328 \pm 0.004$$

was obtained. The epi-cadmium to sub-cadmium capture rate is then

$$\rho_c^{28} = 3.05 \pm 0.04.$$

The values of the U^{235} fission cadmium ratios were measured with shaped uranium-aluminum alloy foils with 17.5 w/o uranium enriched to 93 w/o U^{235} , as was done in previous cores. The values were also corrected for thermal self-shielding and extrapolated to zero mass of cadmium. The U^{235} fission cadmium ratios obtained were:

$$\text{in the fuel: } 6.13 \pm 0.20;$$

$$\text{in the moderator: } 6.55 \pm 0.20.$$

The epi-cadmium to sub-cadmium fission ratio in U^{235} in the fuel is then

$$\rho_f^{25} = 0.195 \pm 0.008.$$

The U^{235} fission ratio of a bare moderator foil to a bare fuel foil was found to be

$$\frac{\text{Bare moderator}}{\text{Bare fuel}} = 1.152 \pm 0.006.$$

From this ratio and the fission cadmium ratios in the fuel and moderator, the thermal disadvantage factor is

$$\zeta_{25} = 1.166 \pm 0.011.$$

3. Nuclear Constants

a. Radiative Capture. Neutron radiative capture cross sections of substances important to the fast reactor program are being studied as a function of neutron energy between 4 keV and 3 MeV, thus covering a large part of the neutron-energy spectrum of fast reactors. Capture cross sections are needed to make proper use of materials proposed for structural and control use in reactors. The data are also useful in testing theories of nuclear reaction. The information gained by comparison of experimental data with theoretical values could allow the prediction of other neutron cross sections in the absence of experimental data.

In these studies, the activation method, in which the nucleus being studied is irradiated with a monoenergetic neutron beam, and the radioactive species produced is counted with a beta counter or a gamma-ray analyzer, was used. The counting data are converted to capture cross sections by suitable calibrations.

Capture cross sections have been determined for Y^{89} . In Fig. 14 the data are compared with those in the literature.^{7,8} A significant extension of the data is shown.

Fig. 14. Capture Cross Section of Yttrium-89

⁷ Macklin, R. L., Gibbons, J. H., and Inada, T., Phys. Rev. 129, 2695 (1963).

⁸ Hughes, D. J., Garth, R. C., and Levin, J. S., Phys. Rev. 91, 1423 (1953).

Work has begun on measuring the cross sections of K^{41} , Rb^{87} , Sr^{86} , V^{51} , and Mn^{55} .

B. Theoretical Reactor Physics

1. The ARC System

a. Burnup and Fuel Management. Preliminary results of a study to develop a flexible Burnup Module have been reported (see Progress Report for February 1966, ANL-7176, pp. 51-53). The following is a revision of what has already been reported and some further developments. Most of the symbols used here are defined in the previous report.

In considering the fuel management, it is convenient to work with the total number of atoms. Define a matrix $\underline{V}$:

$$\underline{V} = \begin{bmatrix} v_1 \underline{I} & & 0 \\ & v_2 \underline{I} & \\ & & \ddots \\ 0 & & & v_K \underline{I} \end{bmatrix}, \quad (1)$$

where v_i is the volume of region i , and $\underline{I}$ is the $(M \times M)$ unit matrix. The atomic composition for the $(i+1)$ -st irradiation step is given by a vector

$$\begin{aligned} \underline{n}^{i+1} = & \underline{V}^{-1} [(\underline{I} - \underline{D}) + \underline{A}(\underline{I} - \underline{C} - \underline{G}) \underline{D}] \underline{B}^i \underline{V} \underline{n}^i \\ & + \underline{V}^{-1} \underline{E} \underline{R} \underline{G} \underline{D} \underline{B}^i \underline{V} \underline{n}^i + \underline{V}^{-1} \underline{F} \underline{q}^i, \end{aligned} \quad (2)$$

where the makeup vector $\underline{q}^1$ is found by

$$\underline{q}^i = [\underline{c} - \underline{S}\{(\underline{I} - \underline{D}) \underline{B}^i \underline{V} + \underline{A}(\underline{I} - \underline{C} - \underline{G}) \underline{D} \underline{B}^i \underline{V} + \underline{E} \underline{R} \underline{G} \underline{D} \underline{B}^i \underline{V} \underline{n}^i\}], \quad (3)$$

and

$$\underline{c} = \underline{S} \underline{V} \underline{n}^i \text{ for } i = 0, 1, 2, \dots, \infty. \quad (4)$$

In practice, however, the material returning to the reactor after reprocessing to make up the composition for $(i+1)$ -st irradiation step will not be the material discharged after the i -th irradiation step, as Eq. (3) implies, but the material discharged after some earlier irradiation step. Thus, let the time interval between the discharge from the reactor and the recycle back to the reactor be $\ell\tau$, where τ is the length of irradiation,

assumed to be constant for each step, and ℓ is some nonnegative integer. Then, Eqs. (3) and (4) are modified as

$$\begin{aligned} \underline{n}^{i+1} = & \underline{V}^{-1}[(\underline{I} - \underline{D}) + \underline{A}(\underline{I} - \underline{C} - \underline{G}) \underline{D}] \underline{B}^i \underline{V} \underline{n}^i \\ & + \underline{V}^{-1} \underline{E} \underline{R} \underline{G} \underline{D} \underline{B}^{i-\ell} \underline{V} \underline{n}^{i-\ell} + \underline{V}^{-1} \underline{F} \underline{q}^i, \end{aligned} \quad (5)$$

and

$$\underline{q}^i = [\underline{c} - \underline{S}\{(\underline{I} - \underline{D}) + \underline{A}(\underline{I} - \underline{C} - \underline{G}) \underline{D}\} \underline{B}^i \underline{V} \underline{n}^i - \underline{S} \underline{E} \underline{R} \underline{G} \underline{D} \underline{B}^{i-\ell} \underline{V} \underline{n}^{i-\ell}]. \quad (6)$$

(i) Recycling Process. Let the matrix $\underline{R}$ be associated with the recovery from the reprocessing plant. Two factors contribute in making up the matrix $\underline{R}$:

- (a) the decay and buildup resulting from radioactivity of nuclides;
- (b) the efficiency of reprocessing stages.

Suppose that, once the material enters the reprocessing plant, it loses the information regarding the past history, i.e., the information as to which region it came from, etc. This makes it possible to consider an M-vector $\underline{U} \underline{G} \underline{D} \underline{B} \underline{V} \underline{n}$ instead of the original KM-vector $\underline{G} \underline{D} \underline{B} \underline{V} \underline{n}$, where

$$\underline{U} = \left[\begin{array}{c|c|c|c|c} \underline{I} & \underline{I} & \dots & \underline{I} \\ \hline \underbrace{\hspace{10em}} & & & & \\ K & & & & \end{array} \right]. \quad (7)$$

Further, suppose that the plant consists of P reprocessing stages, where the p -th stage recovers the fraction Pr_m of the nuclide m entering the stage. It will be assumed that each reprocessing stage is instantaneous.

Let the interval between the time at which material leaves the p -th reprocessing stage and the time at which it enters the $(p+1)$ -st reprocessing stage be denoted by Δt_p , and let Δt_0 denote the interval from the time the material is removed from the reactor to the time it enters the first reprocessing stage, and Δt_p denote the interval from the time the material leaves the last (P -th) reprocessing stage to the time it is returned to the reactor. Then,

$$\sum_{p=0}^P \Delta t_p = \ell \tau. \quad (8)$$

Define the following matrices:

(a) the diagonal $(M \times M)$ -matrices $\underline{P}_R$ such that

$$\underline{P}_R = \begin{bmatrix} P_{R1} & & 0 \\ & \ddots & \\ 0 & & P_{RM} \end{bmatrix}, \quad p = 1, 2, \dots, P; \quad (9)$$

(b) the $(M \times M)$ -matrix $\underline{T}$ such that

$$\underline{T}(t) = \underline{B}_{ii}(\phi_i = 0, t), \quad i = 1, 2, \dots \text{ or } K, \quad (10)$$

where $\underline{B}_{ii}(\phi_i = 0, \tau)$ is the $(M \times M)$ -matrix, which is the i -th diagonal element of the block matrix $\underline{B}$ with ϕ replaced by zero.

The atomic composition of the material leaving the reactor for reprocessing is of the form

$$\underline{s} = \underline{B}(\phi, \tau) \underline{V} \underline{n}(0), \quad (11)$$

where $\underline{s}$ is a KM -vector. Since the past history of the materials reaching reprocessing plant is lost, the vector $\underline{s}$ may be operated upon by the matrix $\underline{U}$ to yield an M -vector $\underline{w}$, i.e.,

$$\underline{w} = \underline{U} \underline{s}, \quad (12)$$

where the element w_m of $\underline{w}$ is the total number of nuclide m reaching the reprocessing plant. Now, referring to Fig. 15, the atomic composition reaching the reprocessing stage 1 is, with ${}^1\underline{T} = \underline{T}(\Delta t_1)$,

$${}^0\underline{u} = {}^0\underline{T} \underline{w}; \quad (13)$$

the atomic composition leaving the same stage is

$${}^1\underline{w} = {}^1\underline{R} {}^0\underline{T} \underline{w}. \quad (14)$$

Fig. 15. Reprocessing Plant

Repeating the same operations, the atomic composition leaving the P -th stage is given by

$$\underline{P}_w = \underline{P}_R \underline{P}^{-1} \underline{T} \underline{P}^{-1} \underline{R} \dots \underline{R} \underline{0} \underline{T} \underline{w}, \quad (15)$$

and the atomic composition returned to the reactor is

$$\underline{P}_u = \underline{P}_T \underline{P}_R \underline{P}^{-1} \underline{T} \underline{P}^{-1} \underline{R} \dots \underline{R} \underline{0} \underline{T} \underline{w}. \quad (16)$$

Thus,

$$\underline{R} = \underline{P}_T \underline{P}_R \underline{P}^{-1} \underline{T} \underline{P}^{-1} \underline{R} \dots \underline{R} \underline{0} \underline{T} \underline{u}. \quad (17)$$

Consider Eq. (5). In deriving the equation, it was assumed that the material discharged at ℓ irradiation steps earlier is recycled back to the reactor. In the sequel, this process will be referred to as the "delayed reprocessing." In addition to delayed reprocessing the following two reprocessing processes are of interest:

(a) Sequential Reprocessing--Under the sequential reprocessing Eq. (5) takes the form

$$\begin{aligned} \underline{n}^{i+1}(0) = & \underline{V}^{-1}[(\underline{I} - \underline{D}) + \underline{A}(\underline{I} - \underline{C} - \underline{G}) \underline{D}] \underline{B}(\phi^i, \tau) \underline{V} \underline{n}^i(0) \\ & + \underline{V}^{-1} \underline{E} \underline{R} \underline{G} \underline{D} \underline{B}(\phi^i, \tau) \underline{V} \underline{n}^i(0) + \underline{V}^{-1} \underline{F} \underline{q}^i; \end{aligned} \quad (18)$$

for example, the material recycled may be treated formally as of the same composition as that recovered from the composition discharged at the immediately preceding irradiation step. This process corresponds to the case of having infinitely many identical reactors, operating under the same policy of fuel management, where at least one reactor is started up at $(t - k\ell\tau)$ for each positive integer value of k . The reactor under study starts up at t , discharging the first irradiation product, $\underline{B}(\phi^0, \tau) \underline{n}^0$, to the reprocessing plant at $(t + \tau)$. The reactor that started up at $(t - \ell\tau)$ must have done the same thing, i.e., it discharged its first irradiation product, $\underline{B}(\phi^0, \tau) \underline{n}^0$, to the plant at $[t - (\ell - 1)\tau]$. At $(t + \tau)$ the reactor under study restarts with $\underline{n}^1$, which includes the recycle originating from the other reactor that started up at $(t - \ell\tau)$. The recycled material, by the time it reached the reactor, has stayed out of reactor for $\ell\tau$. The generalization to the later steps is straightforward.

The consideration of this process is intended to represent a situation that may arise in the future, where a large number of identical reactors are commercially manufactured and are operating under some optimized fuel management policy.

(b) Equilibrium Reprocessing--In this process, Eq. (5) takes the form

$$\underline{n}^{i+1}(0) = \underline{V}^{-1}[(\underline{I} - \underline{D}) + \underline{A}(\underline{I} - \underline{C} - \underline{G}) \underline{D}] \underline{B}(\underline{\phi}^i, \tau) \underline{V} \underline{n}^i(0) + \underline{V}^{-1} \underline{E} \underline{R} \underline{G} \underline{D} \underline{B}(\underline{\phi}^\infty, \tau) \underline{V} \underline{n}^\infty(0) + \underline{V}^{-1} \underline{F} \underline{q}^i, \quad (19)$$

where $\underline{n}^\infty$ denotes the equilibrium composition. This process is also intended to represent a similar future situation. Suppose that at least one of the operating reactors has reached equilibrium under sequential reprocessing. The material recovered from the discharge originating from such an equilibrium reactor is assumed to be always available as recycle. With this assumption, Eq. (19) is easily arrived at.

(ii) Equilibrium. In all three cases considered here, it will be reasonable to assume that an "equilibrium" situation will eventually be reached as $i \rightarrow \infty$. Progressing step by step as i is increased in Eq. (5), (18), or (19), the behavior of the reactor may be observed as the equilibrium is approached, that is, the "approach to equilibrium." Computationally, however, it is often of interest to find the equilibrium reactor composition and the feed composition, without having to go through the step-by-step procedure of approach to equilibrium. For the three cases considered, a direct analytical determination of equilibrium reactor and feed compositions is possible, if the flux vector $\underline{\phi}^i$ varies slowly from step to step.

Thus, the assumption will be made that the flux vector $\underline{\phi}^i$ remains constant for all steps, i.e.,

$$\underline{\phi} = \underline{\phi}^i \text{ for all } i. \quad (20)$$

(a) Sequential Reprocessing--Suppose that the process has reached an equilibrium; then Eq. (18) may be written as

$$\underline{n}^\infty = \underline{Z} \underline{n}^\infty + \underline{V}^{-1} \underline{F} \underline{q}^\infty, \quad (21)$$

where

$$\underline{Z} = \underline{X} + \underline{Y}; \quad (22a)$$

$$\underline{X} = \underline{V}^{-1}[(\underline{I} - \underline{D}) + \underline{A}(\underline{I} - \underline{C} - \underline{G}) \underline{D}] \underline{B} \underline{V}; \quad (22b)$$

$$\underline{Y} = \underline{V}^{-1} \underline{E} \underline{R} \underline{G} \underline{D} \underline{B} \underline{V}. \quad (22c)$$

Rewriting Eq. (21),

$$(\underline{I} - \underline{Z}) \underline{n}^\infty = \underline{V}^{-1} \underline{F} \underline{q}^\infty = 0. \quad (23)$$

But Eq. (5) gives, at equilibrium,

$$\underline{S} \underline{V} \underline{n}^{\infty} = \underline{c}. \quad (24)$$

To combine Eqs. (23) and (24), use matrix augmentation to find

$$\left[\begin{array}{c|c} (\underline{I} - \underline{Z}) & -\underline{V}^{-1} \underline{F} \\ \hline \underline{S} \underline{V} & \underline{0} \end{array} \right] \begin{bmatrix} \underline{n}^{\infty} \\ \underline{q}^{\infty} \end{bmatrix} = \begin{bmatrix} \underline{0} \\ \underline{c} \end{bmatrix}. \quad (25)$$

Hence, if the matrix in Eq. (25) is nonsingular,

$$\begin{bmatrix} \underline{n}^{\infty} \\ \underline{q}^{\infty} \end{bmatrix} = \left[\begin{array}{c|c} \underline{I} - \underline{Z} & -\underline{V}^{-1} \underline{F} \\ \hline \underline{S} \underline{V} & \underline{0} \end{array} \right]^{-1} \begin{bmatrix} \underline{0} \\ \underline{c} \end{bmatrix}. \quad (26)$$

The left-hand side of Eq. (26) is the desired compositions.

(b) Delayed Reprocessing--Simply by noting that $i = (i - \ell)$ for large values of i , it can be immediately concluded that the equilibrium compositions under delayed reprocessing are identical to those under sequential reprocessing.

(c) Equilibrium Reprocessing--Consider Eq. (19), where $\underline{n}^{\infty}$ is same as that in Eq. (26); hence, it can be found independently. To keep the nomenclature clear, let $\underline{p} = \underline{n}^{\infty}$ as determined above. Then, Eq. (19) may be rewritten as, at equilibrium,

$$(\underline{I} - \underline{X}) \underline{n}^{\infty} - \underline{V}^{-1} \underline{F} \underline{q}^{\infty} = \underline{Y} \underline{p}. \quad (27)$$

Combining Eqs. (27) and (5) by matrix augmentation as before,

$$\left[\begin{array}{c|c} \underline{I} - \underline{X} & -\underline{V}^{-1} \underline{F} \\ \hline \underline{S} \underline{V} & \underline{0} \end{array} \right] \begin{bmatrix} \underline{n}^{\infty} \\ \underline{q}^{\infty} \end{bmatrix} = \begin{bmatrix} \underline{Y} \underline{p} \\ \underline{c} \end{bmatrix}. \quad (28)$$

Therefore, if the matrix in Eq. (28) is nonsingular,

$$\begin{bmatrix} \underline{n}^{\infty} \\ \underline{q}^{\infty} \end{bmatrix} = \left[\begin{array}{c|c} \underline{I} - \underline{X} & -\underline{V}^{-1} \underline{F} \\ \hline \underline{S} \underline{V} & \underline{0} \end{array} \right]^{-1} \begin{bmatrix} \underline{Y} \underline{p} \\ \underline{c} \end{bmatrix}. \quad (29)$$

No consideration is given so far to such important questions as:

- (a) maintaining the criticality of the reactor;
- (b) achieving the desired burnup by, for example, varying the step length τ from step to step;
- (c) finding the correct flux to use to compute $\underline{B}$ as the vector $\underline{n}$ varies within an irradiation step. These, among others, will be the topics of further study.

2. Hybrid Computer Techniques

A method for analyzing a reactor system has been developed based on the use of a hybrid computer and the principle of contraction mapping (see Progress Report for February 1966, ANL-7176, pp. 49-50).

From the reactor system analysis point of view, calculation of the fuel and coolant temperatures are essential to the accurate determination of reactivity feedback. When thermal properties, such as conductivity of the fuel material vary significantly with temperature (for example, UO_2), a large amount of computer equipment and programming effort would be necessary if either an analog or digital computer was employed separately.

The analog component is programmed to solve the heat flow equation for radial temperature distribution in a fuel rod for an axial increment ΔZ_i . The coolant temperature at the exit of increment ΔZ_i is sampled in time, stored in the memory of the digital component, and then is used as the entering temperature to axial segment ΔZ_{i+1} .

Therefore, when axial conduction of heat in the fuel rod is relatively trivial compared to the radial component (which is reasonable in ceramic fuels), the determination of the temperature profile along the Z axis may be determined by the detailed simulation of radial heat flow in one segment ΔZ_i .

From the detailed determination of the transient temperature profile (radial and axial) for a given number of fuel cells at representative positions in a reactor core, the feedback reactivity as a function of temperature is computed in the digital component for each ΔZ and returned to the neutron kinetics equations in the analog component. The feedback reactivity results from Doppler effect, thermal expansions in the core, and density variation in the sodium.

The overall system analysis is accomplished as follows: An arbitrary expression for feedback is made initially and the transient temperature profile for a given input of reactivity is determined. The digital

component calculates a feedback reactivity concomitant with this temperature profile. The iterative process is continued until the required convergence of the system variables is achieved. Convergence to within 1/4% is usually achieved with four or five iterations.

As an example consider a fuel cell in the form of a cylinder of UO_2 with concentric shells of stainless steel and sodium flowing in the axial direction. For simplicity, a heat generation rate of 1000 W/cm^3 is taken as uniform in space. The sodium temperature at Z_0 is 500°K . Reactivity is inserted at the rate of $\$1.25/\text{sec}$ for one second and then held constant. The feedback of reactivity is given by

$$\Delta k = -0.005 \ln \left(1 + \frac{\Delta T_f}{T_{0f}} \right) - 0.4 \Delta T_f \times 10^{-6} + 0.2 \Delta T_c \times 10^{-6},$$

where ΔT is the variation of average temperature, T_0 is initial temperature, and subscripts f and c pertain to fuel and coolant.

Convergence of the neutron flux is shown by curves A_1 to A_5 of Fig. 16. The converged maximum temperature of UO_2 is given by curve B.

Fig. 16

The Convergence of the Neutron Flux is Shown in the Curves A_1 to A_5 . The converged value for the maximum UO_2 temperature is shown in Curve B. Curve C shows what the UO_2 temperature would be if the radiative component were removed from the temperature-dependent equation for the thermal conductivity of UO_2 .

Curve C demonstrates the importance of internal radiative heat transfer in ceramic fuels. It was obtained by removing the radiative component (photon conduction) from the temperature-dependent equation for the thermal conductivity of UO_2 .

3. Isotope Irradiation

An intense isotopic neutron source has been used to perform low-level irradiations at known, constant fluxes for the development of automated gamma-scanning techniques. To increase the flux level and irradiation volume it has been proposed that this source be placed in a multiplying assembly. To take advantage of the source intensity, a Godiva-type assembly has been suggested as the multiplier with a variety of reflectors.

Two types of multipliers were studied: a spherical assembly of highly enriched uranium at 90% of theoretical density, surrounded by a reflector, and a cylindrical annular assembly immersed in water. In each case the effective multiplication of the assembly in water was adjusted to 20 ($k_{\text{eff}} = 0.95$). The source strength was $3.75 \times 10^{10} \text{ n/sec}$, distributed

over the outer 2 cm of the spherical assembly or the inner 2 cm of the cylindrical annulus. The source neutrons were assigned the spectrum of neutrons from fast-neutron-induced fission in U^{235} .

The conclusions are:

- a. The spherical assembly multiplies the irradiation volume, not the peak flux.
- b. The cylindrical assembly with a 6.0-cm internal thermal column gives a multiplied thermal flux as well as an extensive irradiation volume.
- c. The fast flux for irradiation can be enhanced by use of a metal reflector.
- d. The choice of reflector considerably affects the reactivity of the system; thus it is unlikely that there would be a frequent change of reflector.

The results for the spherical assemblies are summarized in Table XIII, for the cylindrical annuli in Table XIV. No study has yet been made of the effect of replacing the water outside the cylinder by a metal reflector, but the effect should be similar to the spherical cases. These results are preliminary in the sense that the cross sections have not been critically reviewed, there has been only a cursory check with critical experiments, etc., but it is felt that there is no substantial error.

TABLE XIII. Spherical Assemblies

Uranium radius: 6.28 cm

Reflector	Peak Thermal Flux (n/cm^2 -sec)	Peak Fast Flux (>2.5 MeV) in Reflector	k_{eff}
1. Water	7.68×10^7 at $r = 8.78$ cm	2.85×10^7 at $r = 6.78$ cm	0.95
2. Beryllium	9.09×10^6 at $r = 12.78$ cm	2.09×10^7 at $r = 6.78$ cm	0.995
3. Nickel	9.22×10^3 at $r = 13.78$ cm	2.21×10^7 at $r = 6.78$ cm	1.02
4. Lead	8.27×10^3 at $r = 20.78$ cm	2.47×10^7 at $r = 6.78$ cm	0.93

TABLE XIV. Cylindrical Assemblies* ($k_{\text{eff}} = 0.95$)

All immersed in water: height = 6 cm

Case	Inner Radius (cm)	Outer Radius (cm)	Peak Thermal Flux at $r = 0$ ($\text{n/cm}^2\text{-sec}$)	Peak Thermal Flux Outside ($\text{n/cm}^2\text{-sec}$)	Peak Fast Flux in Reflector
1	4	8.65	4.48×10^8	1.38×10^8 at $r = 12.22$ cm	4.22×10^8 at $r = 3.75$ cm
2	6	10.5	5.04×10^8	1.23×10^8 at $r = 14.96$ cm	2.4×10^8 at $r = 5.75$ cm

*A 2-cm inner radius was examined and discarded.

4. Code Modifications, R-153

R-153 is the version of AISITE⁹ for the CDC-3600. This version has been modified to include building-attenuation factors in calculating the direct gamma dose from the building. The procedure is to calculate the attenuation factor for a given gamma-ray energy (EGAMMA) by linear interpolation of the attenuation for 10-MeV gammas (BSFH) and 0.1-MeV gammas (BSFL). The resulting number, BSF, multiplies the computed building dose rate, BLDR, so that the building dose rate at a distance from the building is proportional to $\text{BLDR} \times \text{BSF}$.

5. Subroutine Development, BN

Subroutine BN has been written to solve the one-group BN equations for an arbitrary order N. Source convergence is used when in-group scattering is present.

C. Instrumentation1. High-temperature Instrument Development (Out-of-pile)

a. "Hot Zone" Errors. "Hot zone" errors (see Progress Report for November 1963, ANL-6808, p. 29) have been measured using selected high-temperature thermocouples. The thermocouple construction is as follows: Sheath--0.062-in. OD with a 0.01-in.-thick wall, 20 in. long, made of tantalum; Insulation--99.8% crushable thoria; Wires--0.01-in. dia, W-3% Re/W-25% Re, spaced on 0.02-in. centers. The measured "hot zone" errors are listed in Table XV. By comparison, the results of the calculations with four different insulation-resistivity representations (A, B, C, and D) are also presented. The constants used in the resistivity equation are listed in Table XVI.

⁹Blaine, R. A., and Bramblett, E. L., AISITE II, A Digital Computer Program for Investigation of Reactor Siting, NAA-SR-9982 (Oct 15, 1964).

TABLE XV. "Hot Zone" Errors

t Max Temp (°C)	Measured Error (°C)	Calculated Error (°C)			
		A	B	C	D
1600	4	1.0	16.0	2.0	8.0
1700	16	1.5	32.0	7.0	20.0
1800	33	2.0	53.0	20.0	38.0
1900	59	4.0	76.0	42.0	61.0
2000	91	6.0	99.0	69.0	85.0
2100	122	7.0	122.0	98.0	110.0
2200	153	10.0	145.0	126.0	144.0
2300	184	14.0	169.0	154.0	159.0
2400	216	19.0	197.0	190.0	189.0

TABLE XVI. Resistivity Constants Used

Case	Constants			
	k_4	k_5	k_6	k_7
A	7×10^9	-11.5×10^{-3}	1×10^6	-3.00×10^{-3}
B	7×10^9	-11.5×10^{-3}	1×10^6	-5.76×10^{-3}
C	1×10^{11}	-11.5×10^{-3}	0	0
D	7×10^9	-11.5×10^{-3}	1×10^8	-8.05×10^{-3}

b. Resistivity of Refractory Oxide Insulators. The resistivity of alumina is very much a function of its time-temperature history. This seems to be the result of two separate effects. One effect is the "freezing-in" of defects by quickly cooling the alumina, and the other is an apparent stoichiometric change in the alumina when it is heated to temperatures in excess of 1800°C in the presence of the tantalum electrodes used in the resistivity experiment. The resistivity data (see Fig. 17) were taken with the aid of a transistor curve tracer and the resistivity values represent the initial slope of current-voltage traces. All data were taken in a dry helium atmosphere.

Fig. 17. Resistivity of Alumina

The alumina used was of 99.7% or better purity and was pre-fired for 3 hr at 1950°C so that all shrinkage would take place before the resistivity probe was assembled. The pre-firing was accomplished while the sample of alumina was suspended on a W/25 Re wire in a dry helium atmosphere. After this firing, the alumina tube was made a part of a coaxial resistivity measuring probe that used a Ta outer conductor and center wire. The parts of this probe were sized to give tight press fits.

2. Wide-range Neutron Monitoring

The wide range of neutron flux that must be monitored in nuclear reactors has conventionally required systems that use two or more types of detectors placed at various locations relative to the reactor core.

Suitable discrimination against unwanted gamma, alpha, and noise signals can be difficult to achieve in this type of system. A neutron-detection system has been tested which uses a single, fixed-position neutron detector coupled to a "nonswitched" electronic system to monitor continuously up to ten decades of reactor power. Superior gamma discrimination compared to conventional systems is realized over the entire range of neutron flux.

In the test, a conventional counting technique was used for the source and low intermediate ranges, and a statistical chamber-current fluctuation technique, sometimes referred to as a "Campbell System," was used to monitor the intermediate and power ranges. Sufficient linear overlap of the two output signals was experimentally verified, thus demonstrating the continuous monitoring feature of this system. A mean chamber current could be measured and used for the linear high-power trip signals.

A standard fission counter operating at 300 V provided the signal to a narrow-band pass preamplifier. The preamplifier output was fed to a linear pulse amplifier, with a count-rate meter output, and a true rms voltmeter connected in parallel. The mean chamber-current signal was measured with a picoammeter. The various experimental responses of the system to neutron flux are presented in Fig. 18. The lowest measurable flux level will be determined by either the alpha output from the fissionable coating, as was shown by this experiment, or the gamma output. It has been calculated that a gamma flux of 10^5 R/hr would be required to produce a 10% error in a measured neutron flux of 10^5 nv.

Fig. 18. System Responses to Neutron Flux

D. High-temperature Materials Studies

1. Uranium-Urania Phase Diagram

The results of recent arc-melting experiments make it possible to narrow the uncertainty of the monotectic composition in the uranium-urania system at 2470°C from 1.30 ± 0.10 to 1.33 ± 0.03 O/U atom-ratio units (see Progress Report for November 1965, ANL-7122, pp. 47-50).

2. Uranium-Urania-Tungsten Phase Diagram

The uranium-urania-tungsten system shows no evidence for the existence of intermediate phases. The general form reported¹⁰ for the binary W-U system, namely, that of a simple eutectic system, was confirmed. The binary W- UO_2 system appears to be the same type, with the eutectic close to the composition UO_2 . The liquid miscibility gap that was found to exist in the uranium-urania system above 2470°C was found to extend into the ternary system as well. The distribution of tungsten between the two immiscible liquids is strongly in favor of the metallic phase. Tungsten solubility in solid urania appears to be quite low; for example, at 2300°C the solubility in urania of O/U atom ratio ≈ 1.75 is less than 0.5 w/o. The solubility of tungsten in liquid UO_2 is also reported¹¹ to be low, about 0.5 w/o at 2900°C.

Further study of this ternary system is not planned in view of the very limited solubility of tungsten in urania.

3. Ceramics

a. Thermodynamic Study of Nonstoichiometry in Urania. The transpiration measurements of total pressure of uranium-bearing species and (simultaneously) of oxygen partial pressure over urania have continued (see Progress Report for October 1965, ANL-7115, pp. 53-55). The accumulated data obtained between 2080 and 2705°K are presented in Figs. 19 and 20.

The isotherms of total pressure of uranium-bearing species given in Fig. 19 are effectively total pressure curves since the contribution from the species O(g) and $\text{O}_2(\text{g})$ is negligible. The triangular end-points represent conditions at the hypostoichiometric urania phase boundary and were calculated by means of the equations

$$(1873 \text{ to } 2743^\circ\text{K}) \log X_{\text{U}} = 1.335 - (5608/T); \quad (1)$$

$$(1543 \text{ to } 2317^\circ\text{K}) \log P(\text{atm}) = 7.25 - (27,024/T). \quad (2)$$

¹⁰Schramm, C. H., Gordon, P., and Kaufman, A. R., Trans. AIME 188, 195 (1950).

¹¹Weidenbaum, B., and Hausner, H., A.N.S. Trans. 8(1), 32 (1965).

Fig. 19. Total Pressure of Uranium-bearing Species as a Function of Urania Composition

Fig. 20. Partial Pressure of Monatomic Oxygen as a Function of Urania Composition

These equations, previously reported,¹² give compositions and total (effusion) vapor pressures, respectively, at the hypostoichiometric urania phase boundary as functions of absolute temperature. The filled circles in Fig. 19 are extrapolations of the effusion data for stoichiometric urania.¹³ Linear extrapolations of their low-temperature vapor pressures were used rather than their experimental values, because the evaporating solid urania probably became hypostoichiometric at the higher temperatures. Figure 19 indicates that the results obtained by the transpiration and (two) effusion studies are consistent. The isotherms have been extended into the hyperstoichiometric urania region, and it is of interest to note that minima occur close to the stoichiometric composition. Mass-spectrometric investigation¹² of this system has shown that UO is the predominant gaseous species in the far hypostoichiometric region, and that its partial pressure decreases rapidly as the oxygen content of the solid increases. The partial pressures of UO_2 and UO_3 increase as the oxygen content of the solid increases; UO_2 becomes the predominant gaseous species in the vicinity of the stoichiometric composition, and UO_3 in the far hyperstoichiometric region. The minima are not likely to be as sharp as drawn in Fig. 19; they probably become less pronounced, i.e., tend to flatten, as the temperature increases. The location of these minima with respect to composition is not accurately determined by the transpiration experiments. However, such determinations are the objective of the measurements discussed later.

The measured oxygen partial pressures in Fig. 20 (open points) were computed from the moisture content of the exit carrier-gas streams. The computations are based on the equation

$$\log P_{\text{O}}(\text{atm}) = \frac{-26,240}{T} + \log \left(\frac{P_{\text{H}_2\text{O}}}{P_{\text{H}_2}} \right) + 6.38, \quad (3)$$

which is derivable from well-known equilibrium equations^{14,15} involving $\text{H}_2\text{O}(\text{g})$, $\text{H}_2(\text{g})$, $\text{O}_2(\text{g})$, and $\text{O}(\text{g})$. The filled points in Fig. 20, representative of conditions at the hypostoichiometric urania phase boundary, were

¹²Ackermann, R. J., Rauh, E. G., and Chandrasekharaiah, M. S., A Thermodynamic Study of the Urania-Uranium System, ANL-7048 (July 1965); see also Progress Report for November 1965, ANL-7122, pp. 47-50.

¹³Ackermann, R. J., Gilles, P. W., and Thorn, R. J., *J. Chem. Phys.* **25**, 1089 (1956).

¹⁴Kubaschewski, O., and Evans, E. L., Metallurgical Thermochemistry, Pergamon Press, New York, N. Y. (1958), p. 339.

¹⁵Stull, D. R., and Sinke, G. C., Thermodynamic Properties of the Elements, Advances in Chemistry Series, No. 18, American Chemical Society, Washington, D. C. (1956), p. 144.

calculated by means of Eq. (1) and the following equation¹² derived in the mass-spectroscopic effusion study of the uranium-urania two-phase system:

$$\log P_0(\text{atm}) = 7.59 - (40,980/T). \quad (4)$$

The isotherms of monatomic oxygen partial pressure smoothly interconnect the single-phase transpiration work with the two-phase effusion work. Of considerable interest is the fact that the isotherms rise less steeply at the stoichiometric composition as temperature increases. This behavior has important consequences for defect model theories¹⁶ and could imply that stoichiometric urania is intrinsically disordered, with equal numbers of vacancies and interstitials.

One may use the data of Fig. 20 to obtain a value of ~ -45 eu for the relative partial molar entropy of solution of diatomic oxygen in stoichiometric urania. The experimental values obtained by other workers in the range 900 to 1400°C have been summarized by Hagemark;¹⁷ they seem to fall in the range from -7 to 20 eu. The value of -45 eu is in good accord with estimates¹⁸ based on defect theory.

b. Congruently Vaporizing Composition of Urania. An investigation of the congruently vaporizing composition(s) of urania has been initiated to clarify certain discrepancies in the literature concerning the vapor pressure of UO_2 . The usual methods for the experimental determination of vapor pressure require that the composition of the condensed phase attains some fixed value and remains there during the course of the measurement. However, residual gases in a vacuum system (or, in the case of transpiration measurements, contaminants in the inert carrier gas) can be oxidizing or reducing, depending on temperature, and can affect the solid composition. On cooling the residual solid prior to analysis, the composition may change and thus differ from that of the evaporating solid. To avoid some of these difficulties, an extremely clean system is being used for the effusion measurements now in progress. The residual gases ($<10^{-9}$ Torr at temperature) are being monitored by a quadrupole-type analyzer to assure that they are safely below values which could affect the composition of the residual solid. The experiments are performed by heating urania of known composition to high temperatures in a tungsten Knudsen effusion cell within an ultrahigh-vacuum system. A considerable fraction of the initial sample is evaporated over several hours to ensure attainment of the congruently vaporizing solid composition. After the residual solid is cooled, its composition is determined by ignition to U_3O_8 .

¹⁶Anderson, J. S., Nonstoichiometric Compounds, Advances in Chemistry Series, No. 39, American Chemical Society, Washington, D. C. (1963), Ch. 1, p. 6.

¹⁷Hagemark, K., KR-67, Institutt for Atomenergi, Kjeller Research Establishment, Kjeller, Norway (Feb 1964).

¹⁸Thorn, R. J., and Winslow, G., Argonne National Laboratory, private communication.

The results obtained so far, shown in Table XVII, indicate that between 2170 and 2290°C the congruently vaporizing compositions of urania (the minima of Fig. 19) are hypostoichiometric, and that the oxygen deficiency increases with increasing temperature. The similarity of the final O/U ratios in Exp. 4 and 6, in which equilibrium was approached from opposite directions, furnishes evidence that equilibrium is being attained in these experiments. Further measurements are now in progress.

TABLE XVII. The Congruently Vaporizing Compositions of Urania

Exp No.	Temp (°C)	Weight of Urania (g)		Composition, O/U Atom Ratio (± 0.004)	
		Initial	Final	Initial	Final
1	2170	2.10	1.20	2.112	1.997
3	2219	3.32	1.40	2.093	1.990, 1.991
2	2225	2.00	1.07	2.093	1.981
5	2267	3.10	1.90	2.093	1.991, 1.993
4	2290	3.00	1.50	2.093	1.940
6	2290	3.60	2.20	1.89	1.945, 1.950

c. Th-U-Pu Phosphides. Heating of UP pellets in vacuum above 1400°C generally produced a decreased lattice constant, which accompanied selective loss of phosphorus. Reduction in cell size with the loss of the relatively large phosphorus atom suggested formation of nonmetal lattice vacancies. The lattice parameter reached a minimum on heating between 2000 and 2100°C, but showed an increase at higher temperature.

Recent experiments revealed a difference in the vaporization behavior of UP powder and pellets (see Table XVIII). As might be expected,

TABLE XVIII. Vaporization Behavior of UP in Vacuum

			Pellet with 1% Binder	Pellet without Binder	Powder with 1% Binder	Powder without Binder
<u>A.</u>	4 hr at 2000°C	w/o Loss	12.2	11.2	21.2	19.8
		Lattice Constant, Å	5.586	5.586	5.588	5.588
<u>B.</u>	4 Additional hr at 2000°C	w/o Loss	9.2	9.7	17.8	17.7
		Lattice Constant, Å	5.585	5.585	5.588	5.588

NOTE: Original material had a lattice constant of 5.589 Å.

weight losses of powders were considerably greater than those of the pellets, the difference being due to the larger surface areas presented by powders. However, surprisingly, the lattice parameters of the powders changed very little from those of the original material, while those of the pellets exhibited greater changes. Apparently, loss of phosphorus by the powders was almost balanced by the loss of uranium, indicating that UP vaporized almost congruently at 2000°C. However, phosphorus was clearly lost selectively from the pellets. The pellets had a density of 93% of theoretical, with very little open porosity; consequently, most of the vaporization came from pellet surfaces. Movement of atoms from the center of the pellet to the surface would be mainly through some diffusion process, and the relatively low lattice constant of the pellets suggests that diffusion of phosphorus in UP was more rapid than that of uranium. The presence of binder did not appear to have had any effect on vaporization behavior; chemical analysis did not reveal any carbon residue in the samples.

d. Thermal Stability of Plutonium Ceramics

(i) Gd₂O₃ Evaporation. Final testing is being conducted on the glovebox thermogravimetric-analysis apparatus. In order to discover potential difficulties before the closing of the glovebox, the apparatus and operating procedures were tested by experiments with a nonactive material. Evaporation experiments were started with gadolinium sesquioxide, whose use seemed appropriate because of the possibility of using it as a stabilizing agent for (U,Pu)O₂ fuels. In addition, the temperature range (2150 to 2600°K) at which its vapor pressure is significant is similar to that of interest for the planned (U,Pu)O₂ evaporation experiments. Finally, experimental data exist for comparison with the present results.¹⁹

Evaporation of sintered Gd₂O₃ pellets was studied. The system pressure was maintained at about 10⁻⁶ Torr during the evaporation runs. The automatic recording system for the balance made it possible to make several isothermal runs on a single specimen without removing it from the furnace.

Figure 21 shows that weight-loss versus time curves for a Gd₂O₃ pellet evaporating from an open tungsten crucible are linear for the test temperatures. The Knudsen cell measurements of White et al., showed an initial period of decreasing rate followed by a linear period. They suggested that the initial period could have been due to reaction of the sample with the tungsten cell, outgassing, and loss of oxygen from the specimen. The present results indicate that, at least under conditions of free evaporation, the other processes are insignificant and that the chief cause of the

¹⁹White, D., Walsh, P. N., Ames, L. L., and Goldstein, H. W., "Thermodynamics of Vaporization of the Rare-earth Oxides at Elevated Temperatures; Dissociation Energies of the Gaseous Monoxides," Thermodynamics of Nuclear Materials, pp. 417-443, IAEA, Vienna (1962).

weight loss is the evaporation process. This conclusion is supported by the result that one specimen, evaporated at temperatures from 2150 to 2380°K, lost 98.5 mg while the tungsten plate on which it rested lost only 0.6 mg. Evidently, the weight-loss contribution from reaction with the tungsten crucible was very small.

Fig. 21. Weight Loss vs. Time for a Gd_2O_3 Pellet

ing due to reflections from the crucible walls. Blackbody conditions were very poorly approached by the bare pellet, so the temperatures given are those measured by sighting on the crucible for the same set point.

Despite the uncertainties mentioned above, the data appear consistent for each specimen, and the three slopes agree. The rates are all less than, and the slopes greater than, those reported by White *et al.* This disagreement is in the right direction to suggest the possibility of an evaporation coefficient. However, it may only represent the influence of uncontrolled parameters in the present experiments. It is planned to conclude the Gd_2O_3 work by Knudsen-cell measurements. These measurements will permit a comparison between free-evaporation and Knudsen-cell data obtained in the same apparatus under the same conditions.

Figure 22 summarizes the weight-loss data and includes an extrapolation of the data of White *et al.* for comparison. Specimen G-1 was 5.74 mm in diameter and 3.81 mm thick; specimen G-4 was 11.00 mm in diameter and 2.39 mm thick. Specimen areas were calculated from micrometer measurements with allowances being made for area changes due to weight loss. For the specimens that were evaporated from the tungsten crucible, it was assumed that only the top and sides of a specimen contributed to the evaporating area. No rate corrections were made for molecular flow effects or for rate lower-

Fig. 22. Summary of Weight-loss Data for Gd_2O_3 Pellets

cooled laser head between them. The length of the cavity is adjustable over a wide range, and both high-quality ruby and neodymium rods can be interchanged in the head. An autocollimator used to align the reflectors with the laser crystal to a high degree of precision is part of the installation.

A vibration-free, adjustable mounting platform has been built at the Laboratory to enable the laser pulse to be directed into the horizontal diffusivity furnace. The beam passes through a quartz window installed into a plastic pouch in the glovebox wall.

Output pulses ranging from 9 to 15 J were used to obtain thermal diffusivity values for Armco iron and uranium monocarbide standards mounted in the furnace. These values agree with our previously published data. It is expected that the new laser, capable of energy pulses up to 34 J, will enable us to extend the temperature range and improve the accuracy of our measurements.

e. Development of Thermal Diffusivity Rig for Ceramic Materials. A laser installation specifically designed for the thermal diffusivity project has been received. The energy pulses produced by this unit are reproducible to 3% as measured in a laser calorimeter, which is part of the installation. This reproducibility is obtained by a closed-cycle, refrigerated heat exchanger capable of maintaining the temperature of the laser crystal to within $1^\circ C$. This feature is needed to obtain heat-capacity values of the specimen so that the thermal conductivity can be calculated from the experimental data. A power supply capable of charging a capacitance bank adjustable from 140 to 420 μF to voltage levels between the lasing threshold and a maximum ensures flexibility in the high-output range.

To obtain well-collimated pulses at low-energy levels a set of calibrated filters is used. The laser cavity is formed by two reflectors mounted on an optical bench with the water-

f. U-S-O System. Procedures have now been developed for obtaining all the starting materials for this study in the desired purity. Uranium (or UH_3), US , and U_3O_8 are prepared by conventional gas-solid reactions. Uranium disulfide was prepared by reacting an excess of H_2S with reactive uranium powder, US_2 being the highest sulfide stable at the reaction temperatures. Mixtures of hexagonal γ US_2 and orthorhombic β US_2 were obtained, with the γ form predominantly formed at 400°C and the β form at 500°C . A stoichiometric $\text{UO}_{2.00}$ powder was produced by the reduction of as-received Kerr-McGee uranium dioxide ($\text{O/U} = 2.13$) in H_2 at 1500°C . The high temperature was required to reduce the surface area of this very active powder to the point where it would not readily reoxidize. Powders reduced at 1000°C consistently reoxidized to $\text{UO}_{2.035}$ on cooling. An equimolar mixture of UO_2 and US_3 showed complete conversion to UOS when pelletized at 1400 kg/cm^2 and fired at 1200°C in vacuum for 3 hr. This completes the list of six starting materials.

The low-temperature oxidation behavior of US (see Progress Report for October 1965, ANL-7115, pp. 49-50) was further clarified. It had been thought that progressive intensity changes of certain lines in the US powder diffraction pattern during oxidation might be indicative of a rather extensive oxygen solubility. Polished sections of powders withdrawn at various stages of oxidation, however, indicated that only a small amount of oxygen was soluble in the US phase, this amount probably being reflected in an initially lowered lattice parameter. Oxidation to UO_2 began at the surface of the particles and proceeded inward by diffusion through the adherent layer until conversion to a strained UO_2 phase, containing as much as 0.5 moles of S per mole of UO_2 , was complete. (The X-ray data, then, simply resulted from the superposition of a diffuse UO_2 pattern having only low angle lines on a sharp, complete US pattern, the cation lattices of the two phases being almost identical.) The sulfur remained in the oxide structure during conversion to U_3O_8 and was eventually lost in an endothermic reaction at 750°C .

4. Liquid-metal Corrosion

a. Polarization Studies. Experimentation has continued with zirconium electrodes in 540°C sodium nearly saturated with oxygen. The presumed likelihood of polarization-cell short circuits due to sodium condensing on the electrode shank has been discussed previously. The effectiveness of various insulating surface films, including oxide films, in insuring the absence of such shorts has been poor. Oxide corrosion films have been found during overnight exposures at 540°C to dissolve sufficiently in underlying zirconium to impair the insulating quality of the film. Recently, the feasibility was explored of retarding oxide dissolution by prior enrichment with oxygen of underlying zirconium metal.

The surface of a 0.25-in.-dia rod electrode of zirconium was enriched with oxygen in a tube furnace at 900°C by evacuating and adding oxygen at a known rate. A weight gain of 3.7 mg/cm² was obtained. Subsequent metallographic inspection revealed the average thickness of a resulting uneven black surface film to be about 7.5 μ ; thus little of the oxygen gained was dissolved. However, a sectional microhardness traverse showed metal hardness to decrease rather linearly from a surface value comparable to that of oxide corrosion product to the (unchanged) bulk metal value in a distance of 150 μ . While the absorption technique may be improved, the sample was considered worthy of further treatment in oxygenated sodium to develop a uniform film and subsequent test of the resistance to dissolution of the film.

Accordingly, the sample was exposed for one day at 540°C in the cell containing oxygenated sodium. After this exposure, areas exposed to the vapor phase were no longer satisfactory insulators at room temperature, while areas exposed to the liquid phase showed excellent room-temperature resistance to surface contacts and displayed a dull gray appearance.

A test of resistances of the film to dissolution by the metal was made by sealing the sample in an evacuated Vycor capsule for an overnight treatment at 527°C. Afterwards, the room-temperature insulation resistance to surface contacts had decreased, although the degree of resistance remaining may prove to be satisfactory.

Subsequent overnight exposure of part of the same sample to oxygenated sodium at 540°C, utilizing a portion of the "satisfactory" area as an insulated shank section, and imposition of anodic polarization current, yielded evidence of some cell polarization that decayed with time. Afterwards, the liquid-phase area of the sample displayed low resistance at room temperature to surface contacts. This behavior is consistent with previously observed effects of anodic polarization. The shank surface-resistance behavior is marginal in spite of metal enrichment with oxygen. Means of improving the enrichment technique are being considered.

b. Dissolution Kinetics. Measurements of the diffusivity of Ta¹⁸² in liquid tin over the temperature range from 500 to 1100°C have been completed. A sufficient number of 2.5-mm-bore Vycor capillaries have been successfully used at the higher temperatures.

Additional dissolution runs have been made at 900 and 1100°C to explore further the velocity dependence of the dissolution rates. The region of laminar flow in the vicinity of the disc sample ($Re = 1 \times 10^3$ to 1×10^5) has been investigated at 900, 1000, and 1100°C. The data indicate that the transition from diffusion or mixed reaction control to surface control moves to higher Reynolds numbers as the temperature increases.

Several more runs at a temperature around 1400°C are planned to determine if complete diffusion control can be achieved.

c. Lithium Corrosion Studies at Elevated Temperatures. In the continuing study of the interaction of refractory metals with multicomponent liquid metals, tantalum in lithium or lithium-silicon are systems of interest. Tantalum is inherently quite ductile, and displays good fabricability and weldability. However, tantalum may suffer localized attack by lithium at temperatures above 1000°C. Results of exploratory tests of these systems at 1200°C are given below. Effects on the molybdenum container capsules are also described.

A small coupon of high-purity tantalum and test lithium containing 1.6 a/o Si were sealed in a high-purity molybdenum capsule. The assembly was tested isothermally at 1200°C for seven days. A weight loss of 4.7 mg/cm² of tantalum was observed at the end of this period. A microhardness transverse of a tantalum cross section revealed a moderate increase of surface hardness (325 KHN near the surface and 296 KHN in the center area: 50-g load--average of six indentations). However, there was no evidence of localized corrosion as previously experienced. The tested surface was smooth, indicating a uniform solution type of attack. The molybdenum container was not corroded, but it was found that internal cracks had developed below the liquid level. There was no abrupt increase in microhardness near cracked areas (229 KHN--100-g load--average of six indentations). A discontinuous surface layer was present on the molybdenum surface areas that had been exposed to liquid lithium.

In the absence of silicon and under otherwise identical conditions, a weight gain of 0.30 mg/cm² for the tantalum insert was observed. Microhardness measurements indicated that there was no disturbed layer discernible on the exposed surfaces of either tantalum or molybdenum, nor was any intergranular attack of the tantalum observed.

The behavior of the tantalum in the lithium test described above was markedly different from that observed in similar tests in the past. The only apparent difference in these tests is that molybdenum (rather than tantalum) was used as the containment material in the latter test. The reasons for the difference in behavior will be investigated. The role of silicon in apparently causing cracking of molybdenum will also be investigated.

5. Irradiation Testing

A series of irradiations are in progress in the MTR. A summary is shown in Table XIX.

The latest capsule in this series, ANL-56-13, inserted into the MTR on March 14, 1966, contained three specimens of vibratorily compacted mixed UC-20 w/o PuC and one of PuC in Nb-1 w/o Zr jackets. Nominal irradiation conditions call for 2 a/o burnup at a cladding temperature of 1000°C.

TABLE XIX. Status of Ceramic Fuel Irradiations in Progress

Capsule or Specimen No.	Design Parameters						Operating Conditions			
	Fuel Composition (w/o)	Effective Density (% Theory)	U + Pu atoms per cc x 10 ^{-22a}	Cladding Composition (w/o)	Cladding OD (in.)	Cladding Thickness (in.)	kW/cc ^a	Max Cladding Temp (°C)	Burnup to Date	
									U + Pu (a/o)	fiss/cc x 10 ^{-20a}
56-11(MV-2)	UC-20 w/o PuC	79	2.58	Nb-1 Zr	0.281	0.012	1.2	520	4.8	12.4
56-8(MV-3)	UC-20 w/o PuC	81	2.65	Nb-1 Zr	0.281	0.012	1.2	715	5.3	14.0
56-8(MV-5)	UC-20 w/o PuC	80	2.62	Nb-1 Zr	0.281	0.012	1.2	705	5.0	13.1
56-11(MV-6)	UC-20 w/o PuC	80	2.62	Nb-1 Zr	0.281	0.012	1.2	530	5.3	13.9
56-13(Z-4)	UC-20 w/o PuC	79	2.58	Nb-1 Zr	0.174	0.015	1.3	995	0.2	0.52
56-13(Z-5)	UC-20 w/o PuC	79	2.58	Nb-1 Zr	0.174	0.015	1.3	875	0.2	0.52
56-13(Z-7)	UC-20 w/o PuC	79	2.58	Nb-1 Zr	0.174	0.015	1.3	850	0.2	0.52
56-13(C-45)	PuC	84	2.74	Nb-1 Zr	0.174	0.009	1.4	1045	0.2	0.55
56-8(S-7)	US	80	1.94	Nb-1 Zr	0.281	0.012	1.0	535	3.6	7.0
56-8(S-8)	US	89	2.16	Nb-1 Zr	0.281	0.012	1.0	725	5.6	12.1
56-8(S-9)	US	76	1.84	Nb-1 Zr	0.281	0.012	1.0	750	5.6	10.3
56-8(S-10)	US	91	2.21	Nb-1 Zr	0.281	0.012	1.0	690	5.6	12.3
56-11(S-15)	US	82	1.99	Nb-1 Zr	0.281	0.012	1.0	430	2.7	5.4
56-11(S-16)	US	90	2.18	Nb-1 Zr	0.281	0.012	1.0	560	4.6	10.0
56-11(S-17)	US	88	2.13	Nb-1 Zr	0.281	0.012	1.0	550	3.3	7.0
56-11(S-18)	US	77	1.87	Nb-1 Zr	0.281	0.012	1.0	660	5.1	9.5

^aBased on effective density.

Initial metallographic examination was begun on four vibratorily compacted UC-20 w/o PuC specimens that were irradiated to an estimated burnup of 6.8 a/o of U + Pu (1.8×10^{21} fiss/cc) in the MTR (see Progress Reports for July 1965, ANL-7082, p. 38; for October 1965, ANL-7115, p. 58 for previous findings on these specimens). The uranium was not enriched. Two specimens were jacketed with 0.009-in. Nb-1 w/o Zr and two with 0.009-in. Type 304 stainless steel. One specimen in each jacket material contained hypostoichiometric PuC; the other contained hyperstoichiometric PuC. One transverse metallographic section was taken from the center of each specimen; more will be taken at a later date.

All specimens showed that extensive sintering and PuC swelling had occurred. Copious quantities of fission gas bubbles were found in the PuC areas. The PuC phase formed an almost continuous network around the large 20/40 mesh UC particles. Particles of fine -325 mesh UC were found trapped in PuC areas and along boundaries of PuC particles. Sintering had taken place between PuC particles and between the PuC and the fine UC particles. For the most part the coarse UC fraction did not react directly with the PuC. The growth of the plutonium areas is attributed to both swelling and reaction sintering with the UC. The product of the reaction sintering would be a (U, Pu)C solid solution of indeterminate composition resulting in added plutonium mobility. Figures 23 and 24 show examples of trapped UC in PuC areas and the swelling of PuC into a void area between two large UC particles, respectively. Both areas are from a specimen containing hyperstoichiometric PuC.

The fission gas bubbles in the hypostoichiometric PuC appeared to be generally larger and more interconnected than those in the hyper-PuC, although no free plutonium has been identified. Due to the flux perturbation in a thermal reactor, the burnup is higher at the outer edge of the specimens. Consequently, the gas bubbles were considerably larger at the

250X

Fig. 23. Sintered PuC with Trapped UC Particles and Fission Gas Bubbles

250X

Fig. 24. PuC Swelled into Void between UC Particles

very edge than in the interior in all specimens. A white phase of undetermined origin was found around many bubbles and also unassociated in the cooler regions of all specimens. In general, very little structure in the PuC was discernible even in the etched condition.

The UC particles originally contained a dicarbide second phase but showed a loss of this phase at particle free surfaces and, to some extent, adjacent to grain boundaries. Also, a white grain-boundary phase was present and the former "needle" structure took on greater breadth. It is tentatively thought the new white phase is U_2C_3 formed during irradiation from the UC_2 . These phases are shown in Fig. 24. The loss of the second phase at surfaces may be the result of a carbon-oxygen reaction in which CO is given off, thereby depleting the area of carbon. Oxygen would be present either in solution or absorbed on the surface of the particles. The eventual disposition of the CO gas has not been determined.

The PuC was found to react slightly with the 304 stainless jackets. Occasionally reaction was found with Nb-1 w/o Zr. Maximum jacket temperatures were thought to be around 700°C.

Measurements of fission gas release had indicated that the hypofuel released 20 and 40% of the theoretical yield, whereas the hyperfuel released < 1%. Although from previous results it was expected that the hyperfuel would release less gas, the very low actual release was surprising, particularly since it was less than that from similar specimens of much lower burnup. This discrepancy may now be reconciled because the high-burnup specimens sintered whereas the low-burnup ones did not. Sintering would have the effect of closing the original open porosity and lengthening the paths of escape. Hypostoichiometric fuel, by nature of its free-metal phase, would release more gas even if sintering occurred.

The information gathered from the metallography substantiates the two hypotheses of the experiment:

- a. Initial void space in a vibratorily compacted fuel can accommodate fuel expansion without cladding failure;
- b. Physically mixed UC and PuC will react in-pile, at least to a limited extent, to form a (U, Pu)C solid solution of indeterminate composition.

E. Other Reactor Fuels and Materials Development

1. Fast-neutron Irradiation of Jacket Materials

A total of 12 capsules containing 60 tube-burst specimens and 112 tensile-type specimens are being irradiated in EBR-II subassemblies XA07 and XA08 at temperatures between 540 and 590°C, and have accumulated maximum total neutron exposures of 6.9×10^{21} and 3.7×10^{21} n/cm².

Fourteen additional capsules containing a total of 182 tensile-type specimens have been inserted in EBR-II. Two capsules containing V-20 w/o Ti alloy and one capsule containing Type 304 stainless steel and Hastelloy-X were placed in subassembly XO09 for irradiation to a maximum total neutron exposure of 1.7×10^{22} n/cm² at a maximum specimen temperature of 670°C. Six capsules containing V-20 w/o Ti alloy, four capsules containing Type 304 stainless steel, and one capsule containing Hastelloy-X were placed in subassembly XO10 for irradiation to a maximum total neutron exposure of 3.5×10^{22} n/cm² at a maximum specimen temperature of 500°C. The status and identification of the 26 capsules are given in Table XX.

TABLE XX. Status of Irradiations of Jacket Materials in EBR-II

S/A No.	Capsule Number	Design Parameters				Operating Conditions	
		Cladding Material	Type of Specimen	No. of Specimens	Specimen Environment	Maximum Specimen Temperature (°C)	Exposure to Date (n/cm ²)
XA07	AS-9	V-20 Ti	Tensile	16	Argon-helium	590	6.9×10^{21}
XA07	AS-10	Hast.-X	Tensile	16	Argon-helium	590	6.4×10^{21}
XA07	AS-11	304 SS	Tensile	16	Argon-helium	590	6.6×10^{21}
XA08	AS-1	V-20 Ti	Tube-burst	12	Argon-helium	540	3.5×10^{21}
XA08	AS-2	V-20 Ti	Tube-burst	12	Argon-helium	540	3.5×10^{21}
XA08	AS-3	Hast.-X	Tube-burst	12	Argon-helium	540	3.6×10^{21}
XA08	AS-4	Hast.-X	Tube-burst	12	Argon-helium	540	3.6×10^{21}
XA08	AS-5	304 SS	Tube-burst	12	Argon-helium	540	3.6×10^{21}
XA08	AS-6	V-20 Ti	Tensile	16	Argon-helium	580	3.7×10^{21}
XA08	AS-7	Hast.-X	Tensile	16	Argon-helium	580	3.5×10^{21}
XA08	AS-8	304 SS	Tensile	16	Argon-helium	580	3.5×10^{21}
XA08	AS-12	V-20 Ti	Tensile	16	Argon-helium	580	3.3×10^{21}
XO09	AS-14	V-20 Ti	Tensile	13	Argon-helium	670	3.1×10^{20}
XO09	AS-15	V-20 Ti	Tensile	13	Argon-helium	670	3.0×10^{20}
XO09	AS-27	304 SS, Hast.-X	Tensile	13	Argon-helium	670	3.2×10^{20}
XO10	AS-16	V-20 Ti	Tensile	13	Argon-helium	500	1.2×10^{20}
XO10	AS-17	V-20 Ti	Tensile	13	Argon-helium	500	1.3×10^{20}
XO10	AS-18	V-20 Ti	Tensile	13	Argon-helium	500	1.1×10^{20}
XO10	AS-19	V-20 Ti	Tensile	13	Argon-helium	500	1.2×10^{20}
XO10	AS-20	V-20 Ti	Tensile	13	Argon-helium	500	1.3×10^{20}
XO10	AS-21	V-20 Ti	Tensile	13	Argon-helium	500	1.6×10^{20}
XO10	AS-22	Hast.-X	Tensile	13	Argon-helium	500	1.4×10^{20}
XO10	AS-23	304 SS	Tensile	13	Argon-helium	500	1.6×10^{20}
XO10	AS-24	304 SS	Tensile	13	Argon-helium	500	1.3×10^{20}
XO10	AS-25	304 SS	Tensile	13	Argon-helium	500	1.2×10^{20}
XO10	AS-26	304 SS	Tensile	15	Argon-helium	500	1.3×10^{20}

2. Nondestructive Testing

a. Elastic Moduli of High-temperature Materials by Ultrasonics. Measurements have been made to 1100°C with a sample of Type 304 stainless steel, and calculations of sound velocities and elastic constants are now being made. Echoes from longitudinal waves were very discernible throughout this temperature range. However, the signals due to shear waves, the so-called trailing pulses, were not very clear in the high-temperature region. There was much scattering of sound from the interior of the specimen, presumably from grain boundaries. The room-temperature attenuation of both longitudinal and shear waves was markedly increased after the sample had been cycled to 1100°C twice.

b. Ultrasonic Instrument and Transducer Development. Two ultrasonic transducer probes with lead zirconate-titanate piezoelectric elements and porous sintered nickel backings are being fabricated to evaluate further the potential of porous sintered nickel as a backing medium for ceramic transducers. When completed, these probes will be compared to others with conventional backings of impregnated epoxies.

Additional schlieren photographs of radiation patterns from ultrasonic transducer probes were taken; continuous-wave ultrasound was utilized.

c. Development of a Neutron-image-intensification System. While Juggernaut reactor was not operating, the time was used to reorganize and perform maintenance on the neutron-imaging equipment. Some troubleshooting work was done on the television equipment, and an intermittent gain variation appears to have been eliminated. Television modifications for a gamma control circuit are also under study. Some progress in this area has been made but further work is necessary.

One imaging test made during this period involved the movement of an irradiated, experimental fuel capsule through the neutron-image area. Observation of the image on the television monitor indicated that it was possible to conduct a preliminary inspection of each fuel pin to observe swelling and/or cladding rupture and to inspect the capsule itself to observe broken components, thermocouple connections, etc. Image quality was satisfactory for such observations. This was the first time that the television system was used to inspect a complete irradiation capsule.

A new image-intensifier tube has just been received and initial tests are now in progress. The new tube is of smaller size (15-cm-dia input target) and requires some changes in electrical operation. Preliminary tests with a previously supplied X-ray intensifier tube of this size did indicate that we could satisfactorily operate the smaller tube.

d. Thermal Conductivity of Irradiated Fuel as a Function of Burnup and Temperature. Room-temperature thermal-diffusivity and conductivity measurements have been completed on the irradiated uranium-5 w/o Fs fuel specimens (see Progress Report for November 1965, ANL-7122, p. 54). These included fuel pins of 0.3, 1.8, and 3.0 a/o burnup. Final reduction of the data is now being made.

F. Engineering Development

1. Boiling Liquid-metal Technology

a. Niobium-1% Zirconium Loop. This facility is designed to investigate the heat transfer and two-phase flow characteristics of boiling sodium to a temperature of 2100°F, equivalent to a pressure of approximately 8 atm. Among the variables to be investigated are boiling heat flux

and temperature difference to the point of critical flux occurrence, boiling and adiabatic two-phase pressure losses, vapor volume fraction, and boiling stability parameters.

Instrumentation, power, and sodium service connections through the vacuum chamber ports to the Nb-1% Zr loop are nearing completion. The pressure transducers, fill line, electrical power leads, flowmeter lines, and the condenser shutter drive have been completed. Installation of the thermocouple assemblies, flowmeter output leads, radiation insulation, and preheater assembly is underway. Following this, the entire chamber and loop will be thoroughly cleaned before the bell jar is placed over the loop assembly.

The sodium-handling and -sampling system has been placed in operation. Filtered sodium was fed to the dump tank until the tank was approximately 75% filled; the sodium-cleanup system was then charged and circulation through the zirconium hot trap (at 1300°F) was begun. The temperature of the remainder of the system is being maintained at 350°F.

Work is continuing on the future design of a transient heater. Feasibility of initiating a special service contract with an outside vendor for determining the characteristics of a diode or triode electron heater is being investigated.

b. Heater Experiments

(i) Electron-bombardment Heater Experiment. Modifications to the 20-kV, 200-kW power supply have shown that high-voltage arcing (caused by dust collection) is prevented by installing an air filter in the inlet air cooling duct, and that replacement of the saturable reactor by powerstats provides the necessary input voltage control required for smooth operation of the electron heater voltage. A new heater arrangement has been installed and further tests are underway.

2. General Heat Transfer

a. Heat Transfer in Double-pipe Heat Exchangers

(i) Countercurrent Turbulent Liquid-metal Flow. Computer programs for the prediction of temperature distribution and overall heat transfer rates for the test section heat exchangers of the experimental study, and for the performance of "computational experiments," have been prepared. The former is necessary for selection of sets of specific operating condition desired for the mercury heat transfer experiments. The latter relates to investigations of alternate computational procedures (see Progress Report for February 1966, ANL-7176, pp. 63-64). Both programs are presently being "de-bugged."

The first test section and a new orifice assembly for calibrating flow rate are being fabricated. When completed, operation of the mercury flow loop will begin.

3. Electric Master-Slave Manipulators

The configuration and characteristics of an improved 50-lb-capacity electric master-slave manipulator have been described (see Progress Report for September 1965, ANL-7105, p. 50, and November 1965, ANL-7122, p. 58). This manipulator from now on will be referred to as the Mark E4A to distinguish it from Mark E4B, designed for heavier duty. The slave arm of the Mark E4A manipulator is essentially complete in mechanical and electrical detail, except that the cables have yet to be connected. The master arm is also essentially complete with one exception: the delivery of the synchro gearing from the vendor is several weeks overdue. It is expected that the complete manipulator will be ready for operation about one week after the gears are delivered.

Design work is under way and the manufacture of some parts has been started for the slave arm of a 100-lb-capacity manipulator, Mark E4B. This manipulator, together with the Mark E4A, will provide a pair of test arms to be used in the development of power tools and techniques for carrying out remote maintenance. The new manipulator will also be used to determine the effect on the overall operating characteristics of a high-force boost from master to slave. This force boost for the Mark E4B will be variable up to a factor of 10:1.

The Mark E4B Manipulator will differ from the E4A principally in the load capacity of the slave arm, which will be in the range from 100 to 150 lb for all motions. The slave arm of the E4B will be similar in configuration to the E4A, and the master arm will be nearly identical to the E4A. Each servo-controlled motion will be powered by a two-phase, 60-cycle, cup-type servo motor. This motor has very low inertia for a motor of this capacity. The motor currently being fabricated has undergone some redesign on the basis of the tests performed on the previous prototype.

G. Chemical Separations

1. Fluidization and Volatility Separations Processes

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

(i) Reactions of BrF_5 with UF_4 and U_3O_8 . Experimental evaluation of bromine pentafluoride as a selective fluorinating agent for uranium in uranium-plutonium mixtures continued. A series of experiments were performed to determine the products of the reactions (other than UF_6) between BrF_5 and UF_4 and between BrF_5 and U_3O_8 . For the reaction of

UF_4 with BrF_5 , it was observed that the color of the condensed reaction products was deep red-brown, indicative of bromine. Titration of the condensed products with fluorine²⁰ at 30°C showed that BrF_5 is reduced to bromine in the reaction. Bromine could result from either the direct reduction of BrF_5 or from a two-step process involving the formation of a BrF_3 intermediate compound. In the event that BrF_3 is formed, it would be necessary to determine which reaction was rate controlling. Results from preliminary experiments with 26 v/o BrF_3 in nitrogen at 200°C indicate that the rate of reaction of UF_4 with BrF_3 is approximately 5 to 10 times the rate of reaction of BrF_5 with UF_4 . For the reaction of BrF_5 with U_3O_8 , again BrF_5 was reduced completely to bromine. This indicates that the fluorinating agent is efficiently utilized, since all of the fluorine in the BrF_5 is consumed. Moreover, with bromine as the sole reaction product, cold-trapping and recycling problems are expected to be simpler than would have been the case if BrF_3 were a reaction product.

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

(i) Engineering-scale Alpha Facility. The third run with plutonium-containing materials was completed. No results are available at this time.

Results from the second run (see Progress Report for January 1966, ANL-7152, pp. 68-69) indicated that about 99% of the plutonium in the feed was collected on the fluid-bed material during the thermal decomposition of PuF_6 to PuF_4 from a mixed UF_6 - PuF_6 feed. It was observed that thermal decomposition occurs preferentially on the surface of the bed particles rather than in the gas phase.

(ii) Process Development Studies for Uranium Dioxide Fuels. Studies have continued in the 3-in.-dia fluid-bed reactor to investigate alternative schemes for processing uranium dioxide fuels. Oxidative pulverization of the UO_2 to U_3O_8 powder followed by fluorination is being tested. One objective of these studies is to determine whether samples of the fluid-bed material taken after oxidation are representative of the fuel charge. If this should be the case, the problem of obtaining an input sample for plutonium accountability would be resolved. To obtain information on the possible distribution of plutonium in the fluid bed after oxidation, UO_2 pellets were used which contained U^{235} in concentrations approximating the plutonium concentrations expected in UO_2 - PuO_2 fuels. The assumption is made in these experiments that the distribution behaviors of U^{235} and plutonium are the same; final verification of this assumption will require experiments utilizing plutonium.

Uranium dioxide pellets of two enrichments were employed in recent experiments. In two experiments, 6-in.-deep beds of UO_2 pellets, consisting of equal quantities of depleted (0.2198% U^{235}) and normal

²⁰Sheft, I., Hyman, H. H., and Katz, J. J., Anal. Chem. 25, 1877 (1953).

(0.7218% U^{235}) UO_2 pellets, were charged to the reactor. Approximately 6.9 kg of sintered alumina, equivalent to a static-bed height of 30 in., was used as the fluid-bed material. In one experiment the alumina was -100 mesh size, in another, -60 mesh. The fluidizing gas, 13 to 19 v/o oxygen in nitrogen, was fed at a superficial velocity of 1.5 ft/sec. Oxidative pulverization of the pellets was completed in 5.5 hr in the first experiment and 11 hr in the second.

Bed samples taken at the end of the 5.5-hr run 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, in the uranium thus indicating that the uranium content of the sample is homogeneous with respect to U^{235} . The concentration of uranium in the bed samples varied from 28.3 w/o uranium in the lower portion of the bed to 32.3 w/o uranium in the upper portion of the bed. In a test in which the oxidation was carried out for 11 hr, analyses of bed samples taken periodically throughout the run from the three sampling points indicated that the uranium in the samples remains homogeneous with respect to U^{235} after 5 hr. Within the range of fluid-bed particle sizes used in these tests, it appears that homogeneity of the uranium isotopes within the U_3O_8 powder is unaffected by the bed particle size. The distribution of U_3O_8 powder in the fluid bed appeared to be unaffected by the size of the fluid-bed particles.

Homogeneity of the uranium isotopes within the U_3O_8 powder was further demonstrated in a separate test involving the oxidation of 0.22 kg of depleted UO_2 pellets and 1.98 kg of normal UO_2 pellets having the same nominal concentrations of U^{235} as those of the pellets in the previous tests. In this test, 2.91 kg of -60 mesh alumina was used as the fluid-bed material. After 5.5 hr of oxidation with 18 v/o oxygen in nitrogen at 450°C, bed samples showed 0.671% U^{235} , indicating that the uranium content of the samples was homogeneous with respect to U^{235} .

The experiments described above demonstrated the homogeneity of the uranium fraction in the fluid bed after oxidation. It is reasonable to expect that plutonium, when present in low concentrations in uranium, will also be distributed homogeneously within the uranium fraction after oxidation in the fluid bed.

In a separate study of the oxidation and fluorination procedures, 2.2 kg of fragmented UO_2 pellets were oxidized to U_3O_8 powder and the powder reacted with fluorine. Approximately 2.9 kg of -60 mesh alumina was used as the fluid-bed material. Oxidation of the fragmented pellets was carried out for 5 hr at 450°C with 20 v/o oxygen in nitrogen as the fluidizing gas. Fluorination of the U_3O_8 fines was conducted as follows: 1 hr at 315°C with 6 to 10 v/o fluorine in nitrogen, 1.5 hr at 350-400°C with 15 v/o fluorine, and 1.5 hr with 20-30 v/o fluorine at 400 to 475°C. Both the oxidation and fluorination steps proceeded smoothly without any operational problems. Uranium hexafluoride production rates as high as 83 lb/(hr)(sq ft) were achieved during the fluorination period. No caking of the alumina bed was observed when the bed was drained from the reactor at the end of the test.

c. Safety in Volatility Processing

(i) Environmental Contamination Control for Volatility Processing. The environmental contamination control for fluoride volatility processing involves treatment of gas streams containing plutonium hexafluoride and fission product fluorides (such as tellurium fluorides) as well as treatment of gas streams containing reagents such as fluorine and bromine fluorides. Owing to the highly toxic nature of plutonium, studies of the hydrolysis and filtration of PuF_6 in gas streams have received the major attention to date. These studies have yielded considerable information on the design of the filter systems and are being continued to gain additional information needed for safe containment of PuF_6 within ventilation facilities. Experimental programs have also been initiated on the chemistry of tellurium fluorides and on the disposal of gaseous reagents.

In the continuing study on the containment of PuF_6 by means of vapor-phase hydrolysis and filtration, a series of seven experiments are being made to improve the reproducibility of the results. In previous experiments²¹ in which PuF_6 was released into air containing an excess of moisture, the fraction of plutonium penetrating two successive AEC filters in series ranged from 1 ppm to 2 ppb, and that penetrating the first filter ranged from 0.2 to 140 ppm. The variations in these penetrations may be due to several factors: (1) variations in the filter sections used, (2) the procedure of mixing the moist air stream and PuF_6 stream, with resulting differences in agglomeration of the hydrolysis product and loading effect on the filters, and (3) the chemical reaction on the surface of the filters.

In the current work, two air streams, one containing PuF_6 and the other moisture, were rapidly mixed to reduce vapor-phase hydrolysis and agglomeration, and were passed through a series of four filters (of 10.0-cm diameter) which had been pretested by use of a DOP aerosol (see Progress Report for September 1965, ANL-7015, p. 54). The apparatus for preparing and mixing the two air streams was the same as that used in previous studies on surface reaction kinetics for the hydrolysis of the hexafluorides (see Progress Report for May 1965, ANL-7046, p. 59). Preliminary results from three experiments indicate that the fraction of plutonium penetrating the first AEC filter ranged from 0.4 to 2 ppm. These tentative results indicate a considerable improvement over the results of the earlier experiments. The reasons for this improvement are not yet known but should become evident when the complete results of the current work are available.

Studies of the chemistry of tellurium fluorides have been initiated to select a suitable adsorbent for these fluorides. Some of the inorganic adsorbents to be tested are activated charcoal, activated alumina, soda lime, and tellurium metal. The studies will be made with a tubular

²¹Chemical Engineering Division Summary Report, July, August, September 1962, ANL-6596, pp. 123-125.

reactor and gram quantities of the adsorbents. The construction of the equipment is nearly completed. As an aid in the investigation, a literature survey of the properties of tellurium and its oxygen and fluorine compounds was made.²²

An experimental program has also been initiated to study the disposal of gaseous reagents used in fluoride volatility processing of nuclear reactor fuel materials. Equipment has been constructed to evaluate the use of solid agents, such as activated alumina, lime, limestone, and soda ash, in the disposal of fluorine, hydrogen fluoride, and bromine fluorides. The equipment consists basically of a 2-in.-dia fluid-bed reactor and a system of instrumentation for analysis of the off-gas from the fluid-bed reactor. Initial tests will be performed to study the reaction between fluorine and activated alumina.

H. Plutonium Recycle Program (EBWR)

1. Reactor Plant Preparation and Maintenance

Completion of repairs to the Reboiler Steam Piping was delayed by weld flaws joining the 6-in. lateral connection to the 8-in. main steam line. This lateral connection is now radiographically sound, and an early completion of welding is anticipated in April.

2. Theoretical Calculations

Calculations have been made of the uniform temperature coefficient of the plutonium critical when poisoned with 5 g/gal of boric acid. Computations by four-group, two-dimensional diffusion theory of the multiplication constant k at 70, 120, 220, 270, and 325°F were made by means of the PDQ03 code. Spectrum-dependent cross sections for each temperature were obtained by use of the GAM code for the three fast groups and by use of the Argonne-revised THERMØS code (with successive flux-weighted cell homogenizations) for the thermal group. The results are given in Table XXI. The results are in qualitative agreement with experiment between 70 and 170°F in that a positive temperature coefficient was observed experimentally for this temperature range. These calculations underestimate the reactivity of the system slightly because the PDQ03 code does not permit group skipping. When these cross sections were used in the CANDID-2D code, which permits group skipping, we obtained $k = 0.999$ for 70°F, which is in excellent agreement with experiment in which the system was critical with 4.9 g/gal of boric acid.

²²Vissers, D. R., and Steindler, M. J., Laboratory Investigations in Support of Fluid-bed Fluoride Volatility Processes. Part X. A Literature Survey on the Properties of Tellurium, Its Oxygen and Fluorine Compounds, ANL-7142, in press.

TABLE XXI. Results of PDQ Calculations of the Temperature Coefficient for a Boric Acid Concentration of 5 g/gal

Temperature (°F)	k, Plutonium Critical (all rods out)	k, Full System (8, noncenter rods in)
70	0.9942	0.9541
120	0.9941	0.9513
170	0.9959	0.9501
220	0.9957	0.9471
270	0.9953	0.9435
325	0.9941	0.9397

Calculations by PDQ03 have also been made of the uniform temperature coefficient of the full system when poisoned with 5 g/gal of boric acid. Here the center control rod was fully withdrawn and the other eight control rods fully inserted. The cross sections for the shim and natural uranium zones were obtained from neutron spectra appropriate for these zones. The results of these calculations are also given in Table XXI. Note that in contrast to the plutonium critical, the calculations predict a negative temperature coefficient for the entire temperature range.

III. ADVANCED SYSTEMS RESEARCH AND DEVELOPMENT

A. Argonne Advanced Research Reactor (AARR)

1. Reactor Physics Experiments

Summarized below are results of experiments with the present, 1215/1620 loading of 1215 fuel foils of standard thickness (~ 62 kg U^{235}) and 1620 B-SS strips (~ 850 gB). The data include: (1) isothermal-temperature coefficient of reactivity; (2) water-void coefficients of reactivity; (3) reactivity effects of aluminum and boron in the ITC; and (4) mean prompt neutron lifetime, by Rossi- α measurements. Preliminary results are given for measurements of effectiveness of neutron windows in raising the thermal neutron flux beyond a steel slab which is used to simulate the pressure vessel of AARR. In addition, data are given for flux levels from multiplication of Sb-Be source neutrons.

The isothermal temperature coefficient of reactivity was measured for two conditions of reactor control: (1) water at its normal level, and reactivity controlled by adjusting the positions of control blades; (2) all control blades withdrawn fully, and reactivity controlled by adjusting water level. With control by control blades, the average isothermal temperature coefficient of reactivity over the range from 24 to 70°C was approximately $-5 \times 10^{-3}\%/^{\circ}\text{C}$. This value includes the effect of the temperature change in increasing the reactivity-control effectiveness of the control blades which were inserted. (At 24°C, control blades Nos. 7 and 9 were 20.0 cm up from reference zero, control blade No. 11 was at 31.1 cm, and all other blades were withdrawn fully.)

Recalibration of the control blades at 70°C provided data with which to determine the contribution of the increased worth of control blades at the higher temperature. It is concluded that the net isothermal temperature coefficient is due mostly to the increasing effectiveness of the control blades, with an indicated net reactivity contribution of approximately $-4 \times 10^{-4}\%/^{\circ}\text{C}$ from all other effects.

With reactivity control by water level alone, no significant difference in the criticality setting of water level was found between 24 and 71°C. The minimum level for criticality was logged at a temperature of 55°C. Note, however, that the net isothermal temperature coefficient of reactivity is the algebraic sum of positive reactivity contributions from the internal thermal column (ITC) and from the heated beryllium, and negative reactivity contributions from the reactor core. The negative reactivity effect due to a reduction in coolant density in the core at the higher temperature is only one such effect, but it is the negative reactivity effect of largest magnitude; it is sizeable even without the additional negative reactivity feedback due to the increased control worth of control blades.

Void-coefficient measurements were made in the ITC and in the coolant water in the reactor core. ITC voids were simulated with distributed 0.5-in.-diameter solid Teflon rods; voids in the core water were simulated with a uniform distribution of full-length thin Teflon strips. The data are summarized in Table XXII, which includes also a list of void coefficients for earlier loadings.

TABLE XXII. Differential Reactivity Effect of Voids in Water

Reactor Core Loading (Number of fuel foils/ number of boron-stainless steel poison strips)	Void Coefficient of Reactivity (%/ % Void in Region Water)	
	ITC	Reactor Core
315/0	-	-0.159
615/420	+0.044	-0.228
810/810	+0.044	-0.190
1215/1620	+0.040	-0.116

Reactivity effects of introducing full-length aluminum rods and of dilute solutions of boric acid in tubes in the ITC have been measured. The purpose of these experiments is to assist the evaluation of anticipated reactivity changes due to insertion of irradiation samples in the ITC of the actual reactor. The net reactivity effect of displacing small volumes of ITC water with aluminum is +0.029% per percent ITC volume displaced. The maximum differential reactivity effect, +0.036%/ % ITC, occurred for local substitutions at the flat face of the hexagonal outer boundary of the ITC; the minimum effect, +0.024%/ % ITC, occurred at a vertex of the boundary.

The average reactivity effect of introducing a small full-length tube of boric acid solution was measured to be -0.68%/g boron. The tube contained a total of 0.021 g H_3BO_3 at a tube dilution of 3.65 g/liter. This reactivity effect is approximately 1/3 less than the effect determined for the 810/810 loading (see Progress Report for November 1965, ANL-7122, p. 74).

The Rossi- α measurements of the mean prompt-neutron lifetime in the various core loadings have been re-evaluated to include corrections for the degree of subcriticality necessitated by the low flux levels required during the measurements. The effect of such a correction increased somewhat proportionally to the total irradiation level of the fuel. In Table XXIII are listed the revised lifetimes to the accuracy presently known. It is not expected that further adjustments will change these values substantially.

Measurements have been made of the effectiveness of an aluminum window to raise levels of thermal-neutron flux beyond the AARR pressure vessel, both during reactor operation and during reactor startup. In earlier

measurements (see Progress Report for September 1965, ANL-7105, pp. 59-60), the steel vessel was not simulated. The new measurements are based upon a more realistic simulation of conditions to be expected in the actual AARR. The pressure vessel was approximated by a steel slab located at the inner radius of the pressure vessel (4 ft).

TABLE XXIII. Summary of Corrected Values of Mean Prompt-neutron Lifetime (Rossi- α Measurements)

Reactor Core Loading (Number of fuel foils/ number of boron-stainless steel poison strips)	Prompt Neutron Lifetime (microseconds)	
	Corrected Values	Values Reported Earlier*
315/0	39.4 ± 1.0	38.8
615/420	33.1 ± 2.0	26.5
810/810	32.3 ± 2.0	29.4
1215/1620	27.2 ± 1.0	-

*See Progress Report for May 1965, ANL-7046, p. 70. In the 615/420 experiment, there was no Plexiglas in the beryllium reflector.

The results reported here, while preliminary, indicate that it should be possible to monitor and regulate the reactor power level at 100 MWt by means of neutron detectors located outside the reactor pressure vessel.

For these measurements, an aluminum window was made up of close-fitting, thin, 18 x 18-in. layers of aluminum to fill the space between the radial beryllium reflector and the pressure vessel. Measurements with indium foils positioned at the midheight of the active fuel zone indicate that, with the equivalent of a 1/2-in.-thick layer of water plus $24\frac{1}{2}$ in. of aluminum, the operating flux level at the outer surface of a $4\frac{1}{2}$ -in.-thick steel slab is approximately 2×10^{-6} as high as the flux level at the axis of the ITC. Four inches farther out, in water, the flux level was 1/3 as high. For these conditions, neutron detectors should sense a flux level of approximately $4 \times 10^7 P$ n-cm/(cm³-sec), where P is the reactor power level in megawatts. If more water is introduced between the beryllium and the pressure vessel, e.g., in coolant passages in the aluminum or in coolant-distribution layers, the measured flux will drop somewhat. Also, it should be noted that corrections will be required for the differences between the actual beryllium reflector and the idealized beryllium zone in the Criticality Facility. These corrections will be inferred from theoretical analysis, supplemented, if necessary, by further experiments.

To estimate the external source required for reactor startup with new cores, an antimony source, with an activity level of $\sim 1\frac{1}{2}$ Ci, was installed in the beryllium reflector, 6 in. from the fuel zone and at the

midheight of this zone. The source was positioned so that the fuel zone lay between the source and the aluminum window. In this case, there was the equivalent of a 1-in. thickness of water in the window. The steel slab was augmented by placing a 1-in. thickness of steel at its inside surface. All control blades were inserted, except for the 2 peripheral control blades nearest the antimony source. With a BF_3 counter, with a sensitivity of 5-10 cps per thermal nv, a count rate of 13 cpm was obtained at the outside surface of the steel slab. This count rate is more than ten times background levels, and it corresponds to a thermal-neutron flux of approximately unity per 100 Ci of Sb^{124} activity.

2. Theoretical Reactor Physics

From computations of fuel burnup of the FGAR physics-reference reactor, it has been determined that at the end of the anticipated core life the magnitudes of the void coefficients of reactivity in the ITC and in the core water would increase slightly. For the 100%-water ITC, the increase is marginal; the computed end-of-life coefficient is $+0.037\%/ \%$ void, versus $+0.036\%/ \%$ void at the beginning of the core cycle. For the core water, the end-of-life value is $-0.027\%/ \%$ void in core water; the value for the fresh core is $-0.023\%/ \%$ void.

On the basis of the burnup analyses, it is concluded that per unit of reactor power:

- (a) the peak thermal-neutron flux in the 100%-water ITC is independent of fuel burnup;
- (b) the average thermal-neutron flux in the ITC varies little during the core life; computations indicate that it increases somewhat, during the core life;
- (c) the peak thermal-neutron flux in the radial beryllium reflector increases somewhat, during core life.

These results are in accord with results obtained earlier, namely, that to a good approximation the peak thermal-neutron fluxes per unit reactor power are not sensitive to the gross radial distribution of power.

Perturbation theory has been applied to the computation of the mean prompt-neutron lifetime in the various Criticality Facility loadings. The computer code used requires that the $\{1/v_j\}_j$ vector, of reciprocal average group velocities of the multi energy-group model, be space independent. Two sets of computations have been run: (1) using the $\{1/v_j\}_j$ vector characteristic of the asymptotic spectrum of the fuel zone; (2) using the $\{1/v_j\}_j$ vector characteristic of the asymptotic spectrum in the internal and external reflectors. The results are presented in Table XXIV. Note that, for the 615/420 experiments, introducing Plexiglas in the beryllium markedly

reduced the prompt-neutron lifetime. This corresponds to the introduction of coolant water in the actual reflector. The computed mean value for the neutron lifetime will be much closer to the value for the reflector $\{1/v_j\}_j$; this statement is substantiated by calculations performed for the PSAR reference system, wherein the α/v method was employed but using a space-dependent velocity vector. The results of theory and experiment appear to be in reasonable agreement.

TABLE XXIV. Computed Mean Prompt-neutron Lifetimes for AARR Criticality Facility Loadings^a

Reactor Core Loading (Number of fuel foils/ number of boron-stainless steel poison strips)	Beryllium Reflector Composition	Relative Prompt- neutron Lifetime	
		Using Fuel Zone $\{1/v_j\}_j$	Using ITC $\{1/v_j\}_j$
315/0	10% Plexiglas	33.1	37.8
615/420	10% Plexiglas	25.6	30.4
615/420	No Plexiglas	29.2	34.7
810/810	No Plexiglas	26.5	31.9
1215/1620 ^b	No Plexiglas	22.4	27.6
Reference PSAR:		17.9	21.9

^aSee Table XXIII for comparison with experiment.

^bThe computations actually were performed for a loading of 1215 fuel foils and 2000 boron-stainless steel strips, but the difference in lifetime should be small.

Erratum: In the Progress Report for December 1965, ANL-7132, Fig. 31 is in error; the labels of the two curves are reversed. For convenience, the corrected figure is reproduced here as Fig. 25.

3. Reactor Vessel

An alternative design of domed-head vessel closure was developed to accommodate the four additional large ($7\frac{1}{2}$ -in.-diameter) instrument nozzles required by the users. The new closure diameter is equal to the ID of the vessel and is slightly larger than the original. The flange-ring closure members (vessel and head) are similar geometrically and thereby eliminate the problem of differential elastic and thermal strains between the two clamping rings, i.e., the elimination of rubbing on the sealing gaskets.

Fig. 25. Radial Distributions of Neutron Flux in the Preliminary Reference Design AARR.
(Normalized to unit Group-16 flux at center of internal thermal column.)

Several design rearrangements of internal structural members permitted the foreshortening of the vessel to a minimum length of 20 ft 6 in. The shortest vessel, however, does not provide an adequate depth of shielding water for manipulations of active core fuel elements. A vessel approximately 24 ft long is the best compromise to satisfy this functional requirement.

4. Primary System and Components

a. Corrosion Studies. The first heat flux-corrosion test with a stainless steel specimen has operated continuously for 308 hr. The specimen is a Type AISI 304 stainless steel tube with a 0.125-in. ID and 0.015-in. wall thickness. Heat is generated in the wall by passage of 60-cycle a.c. current, and heat is removed by the flow of water through the tube. Thermocouples are spot-welded on the outside of the tube, which is then insulated so that heat is removed only from the interior surface. In the present test, the heat flux across the water cooled surface is 4×10^6 Btu/hr-ft². The coolant is deionized water flowing at 45 fps. The test loop is pressurized to 600 psig, and aluminum and beryllium surfaces in ratios corresponding to reactor conditions are exposed to the coolant.

The pertinent thermal data are given in the following table:

Distance from Inlet End (in.)	Thermocouple Temp (°F)	Temp at Water Surface (°F)	Bulk Water Temp (°F)	Fluid Film Coefficient (Btu/hr-ft ² -°F)
1.00	568	350	132	18,100
1.25	584	366	136	16,700
2.25	592	374	148	17,500
3.25	613	395	161	16,700
3.25	574	356	161	20,500
4.25	619	401	176	17,500
5.25	630	412	187	17,500
5.50	629	411	190	17,800
6.25	637	419	200	18,000

During the test, there has been no significant increase in temperature at the thermocouple locations, indicating no deposition or formation of corrosion products on the interior wall of the tube. This test will continue for a total time of at least 1000 hr.

The isothermal loop run, in which various crevice and galvanically coupled specimens are exposed to deionized water at 200°F, was interrupted after 1000 hr to examine the specimens. Crevice areas between stainless steel and aluminum, stainless steel and beryllium, and aluminum and beryllium were completely free of visible attack. The specimens have been reassembled and returned to the loop for an additional 1000-hr testing period.

5. Hydraulic Loop

Inspection of the Type 304 SS piping welds continued. All 8-in. piping welds have been completed and radiographed. Work on the smaller branch connections to the main loop continues.

IV. DESALINATION

A. Steam Supply from a 10,000-MWt Breeder

As part of the AEC's desalting program, a conceptual design study of a nuclear steam-supply system utilizing a very large sodium-cooled fast reactor was examined with the aid of Westinghouse Electric Corporation as a subcontractor. The design appears to be feasible. Sodium-cooled fast breeder reactor plants of 10,000-MWt size should be considered as being evolutionary from nominal 2500-MWt commercial fast reactor designs.

In Table XXIV is a summary of the design characteristics adopted.

TABLE XXIV. Summary of Data for Very Large Fast Breeder

Gross Reactor Power	10,000 MW
Net Plant Output	3,880 MWe
Sodium Inlet Temperature	720°F
Sodium Outlet Temperature	1050°F
Steam Pressure	2400 psia
Steam Temperature	900°F
Type of Fuel in the Core	(U + Pu)C
Type of Core	Annular
Volume of Core	16,700 liters
Reactor Fissile Pu Weight	10,500 kg
Reactor Breeding Ratio	1.4
Fissile Material Doubling Time	7 yr
Linear Heat Rate (avg)	16.8 kW/ft
Core Burnup (avg)	110,000 MWD/MT
Interval between Refuelings	1/2 yr
Equilibrium Fuel-cycle Cost	1/4 mill/kWh

The neutronic and thermal designs of the reactor were closely linked. The thermal design was based on reasonable expectations for fuel development to satisfy the required thermal performance. The neutronic design was based on holding the sodium voiding effect within workable bounds while achieving a satisfactory breeding ratio. The breeding gain and specific power for the fissile material are sufficient to provide a doubling time of about 7 yr on a geometric, equilibrium basis.

The annularly shaped reactor has a mean core diameter of about 22 ft. Core fuel is uranium-plutonium carbide clad in 304 stainless steel and helium bonded. Axial blankets also are of uranium carbide; the radial blankets consist of a uranium-zirconium alloy also clad in 304 stainless steel.

Control of the reactor is achieved by 84 poison rods whose drives are mounted on the reactor vessel cover.

The reactor is contained in a cylindrical steel vessel. Sodium inlet and outlet lines enter the vessel above the reactor core level. The 14-ft-diameter region interior to the inner radial reflector provides a space for storage of irradiated and fresh fuel. Recharging is accomplished by a mechanism having a radial arm operating through a rotating plug in the vessel cover. Fuel is transferred in either direction between the reactor and the storage area with the reactor shut down. After reactor operation is resumed, irradiated fuel subassemblies are removed from storage through the rotating plug and placed in a transfer cask for shipment to a processing plant. The storage area is replenished with fresh fuel for the next recharging cycle.

The reactor coolant circulates in six primary loops, each containing a 140,000-gpm pump and an intermediate sodium-sodium heat exchanger. Intermediate sodium loops transfer the heat to six once-through steam generators located in three buildings outside of the containment sphere. The steam generators (Incoloy 800 is used for tube material) supply steam at the throttle of 2400 psia and 900°F. Live steam reheat to 660°F is used rather than a sodium reheat cycle. Steam is delivered to three sets of turbine generators in the turbine hall.

Safety problems faced in the design are basically the same as those encountered in the design of a 1000-MWe reactor. A negative power coefficient of reactivity provides stability during normal operation. The design makes incredible the total loss of sodium from the reactor. An investigation of the safety was pursued on a broad basis because the present state of fast reactor safety knowledge placed specific safety considerations beyond the scope and cost of the study. Nominal containment of the reactor and the six primary loops is provided by a 240-ft-dia spherical shell which also contains an emergency-shutdown cooling loop.

Fuel-cycle costs were estimated in order to aid in the development of the design. With a 90% annual plant factor, 5% charge on working capital, and 10 dollars per gram fissile plutonium, the fuel-cycle cost is 1/4 mill/kWh. No complete estimate of capital costs was made; however, certain equipment items were costed on an installed basis. Sodium-to-sodium heat exchangers, secondary sodium pumps and drives, secondary sodium piping, and steam generators were estimated at 10 to 12 dollars per net kilowatt. A brief analysis of the refueling operation indicated that the plant availability as limited by refueling is 95%.

In the future, more detailed examinations of very large fast reactors may result in a more suitable selection of design conditions, those which

give a lower energy cost, and these examinations may modify the following conclusions, but will probably cause no significant changes. These conclusions are:

- a. The 10,000-MWt sodium-cooled fast breeder reactor is feasible within the context of the ground rules of this study.
- b. The equipment sizes and duties are not excessive by comparison to the probable requirements for nominal 1000-MWe designs.
- c. Although no complete capital costs were estimated, it is not apparent that some normal type of scaling law for the influence of size of sodium-cooled fast breeder cost would not apply; however, no knowledge of scaling effects were gained from this study.
- d. Fuel performance for this concept is in line with other fast breeder studies using carbide fuel, considering the estimated fuel-cycle cost and doubling time.
- e. Successful development of fast breeders of nominal 1000-MWe size will provide the technological base for very large fast breeders with no significant further R&D program required.

In summarizing, one might expect that very large sodium-cooled fast breeder reactors would come about by an evolutionary process and that nuclear power plants utilizing such reactors will be available when electrical systems can accommodate the output, providing the fast reactor research and development program achieves plants of 1000-MWe size in the 1980's.

V. NUCLEAR SAFETY

A. Reactor Kinetics

1. Fast Reactor Safety

a. Coolant (Water) Expulsion Studies. During the past month, the expulsion apparatus was modified in order to study the effects of a flowing coolant. The flow rate of coolant was varied for fixed values of all other parameters; this sequence of runs was repeated for several different rates of energy input and for different initial temperatures. The collection of data for flowing water has been completed, but results have not yet been analyzed.

b. Superheat Experiments. The first shipment of piezoelectric pressure transducers has arrived, as well as the oscilloscope. This equipment is being calibrated. Following this checkout period, final assembly of the system should be started.

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

The design and construction of experiments to study critical flow of different fluids are continuing. Fabrication of individual components for the sodium blow-down loop have been initiated. Also, approximately 75% of the pipeline, valves, and instrumentation (transducers and thermocouples) needed for the sodium loop as well as the argon-cleaning system have been ordered.

An analysis of two-phase, one-component droplet flow has been completed. Critical flow is assumed to occur when the system velocity becomes equal to the propagation velocity of a small pressure disturbance in the system. The propagation velocity is determined from the fluid conditions prevailing at the point of choking. A simple propagation velocity model was formulated by assuming the flow pattern is mist flow. When relaxation phenomena that result from temperature and velocity lags between the phases are included in the analysis (nonequilibrium), good agreement is obtained between predicted values and existing two-phase critical-flow data obtained in the high-quality region.

A preliminary analytical investigation dealing with the speed of sound in low-quality mixtures has been completed. Simplified models were used to show the effect of metastability and slip on the speed of sound. An examination of the literature reveals that for low-quality one-component mixtures, only the propagation velocity of compression waves has been

investigated; no information exists for rarefaction waves, yet the latter type appears to be more important in interpreting the critical-flow phenomenon.

Experimental data are needed to check the model predictions because thermodynamic considerations indicate that the speed of propagation of the two types of waves in one-component mixtures may be quite different, particularly in the low-frequency range.

2. Containment Program

The transient responses of a thick, hollow elastic sphere and an infinitely long, hollow cylindrical shell under the simultaneous action of arbitrary surface tractions and thermal radiation conditions on both surfaces were investigated. By this means the stresses engendered throughout a whole-core excursion from time $t = 0$ to final steady-state conditions in a pressure vessel can be studied. In core excursions which generate shock waves, the pressure vessel is deformed long before the gas bubble acts on the deformed vessel. However, after the initial shock wave, the action follows that of the solved problem. Thus, to a first approximation these solutions also apply to the behavior of the pressure vessel after the shock wave has passed.

The torsional vibrations of a finite, hollow, thick elastic cylindrical shell were also studied. A solution was obtained for surface tractions on all four surfaces, all of which are arbitrary functions of space and time. By this means the torsional oscillations of a pressure vessel caused by outside seismic disturbances or internal sources can be determined. In case of seismic disturbances, the dynamic response of the pressure vessel will serve as a known input to the reactor internals. For pressure vessels of varying thickness, the solution can easily be obtained by dividing the vessel transversely into small sections so that the obtained solution applies to each individual section. By requiring the stress and displacement to be continuous at each interface, the solution for the entire vessel can be obtained analytically. This approach is very similar to computer techniques used for pressure vessels. This approach could also be used for the torsional vibrations of tapered shafts or stepped shafts that arise as control-rod drives. The actual analytical solution of this extension of the problem is underway.

3. Irradiated U-5 w/o Fs

A successful technique has been developed for metallographic and electron-microprobe analysis of irradiated fast reactor fuel materials. The specimen in the form of a wafer (approximately 1 mm thick) is mounted with conductive epoxy resin in a stainless steel holder that is in turn mounted in DIAL conductive mounting compound with the microprobe standards.

A U-5 w/o Fs (0.32% burnup of heavy atoms) mounted in this manner was examined in the ARL microprobe. The radial distribution of zirconium, ruthenium, molybdenum, and carbon was measured. No irradiation induced segregation of these elements was observed.

B. TREAT

1. Operations

A sample of mixed $\text{PuO}_2\text{-UO}_2$ Fast Ceramic Reactor fuel that had previously been irradiated to a burnup of 64,000 MWd/ton was subjected to a transient designed to cause melting of a large fraction of the fuel.

A packaged sodium loop containing a 7-pin cluster of EBR-II pins was subjected to a steady-state irradiation. Fission-rate data from this experiment will be used to establish the reactor transient characteristics required for meltdown tests in flowing sodium.

Steady-state and transient data were obtained from the modified fast-neutron hodoscope equipment. A packaged sodium loop that did not contain fuel was used for the majority of these tests.

Several steady-state runs were made with boron nitride material to investigate possible use in detectors for neutron-imaging systems. In this connection, two neutron-sensitive vidicon tubes were tested with the boron nitride as the active surface material.

2. Large TREAT Loop

Interconnection of the electrical heaters was completed. Connection of thermocouple wiring is in progress. The pneumatic system for control of the main sodium valves was installed, and all valves were operated satisfactorily. The sodium and blanket gas systems have been held at leak-testing pressure for two weeks.

The surge suppressor was received at ANL and is now being radiographed to establish the condition of the welds prior to acceptance.

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

1. Metal-Water Reactions

a. Calculation of the Extent of Metal-Water Reaction during a Loss-of-coolant Accident. Analysis of the effect of the reaction of steam with fuel cladding metal in a loss-of-coolant accident has continued. The loss-of-coolant accident resulting from a break in a primary-system

water pipe has been taken to be the "maximum credible accident" for most of the power reactors built to date. Water loss following the pipe break results in the core being left in contact with steam. The decay energy in the core is sufficient to cause the fuel cladding to reach temperatures in a short time at which metal-steam reaction can occur.

A previous analysis (see Progress Report for April 1965, ANL-7045, p. 67) used the proposed LOFT (Loss of Fluid Test) Reactor as a model, except that the calculations were made for a 25-mil-thick Zircaloy cladding rather than the 15-mil-thick stainless steel cladding initially proposed in the LOFT design.

The present calculations were similarly made for 15-mil stainless steel-clad, UO_2 -core fuel. The major differences in the calculational technique were the assumptions made to account for the stainless steel-steam reactions. Firstly, the assumption was made that the stainless steel cladding did not react until it had reached 1400°C (melting point, $\sim 1370^\circ\text{C}$). Secondly, the stainless steel reaction rate was assumed to be limited only by the steam availability above 1400°C . It was assumed that for complete reaction of the metal, spinel-type compounds (M_3O_4) are formed and that 0.51 liter of hydrogen (STP)/g of stainless steel is evolved. A value of 155 cal/g of steel was used for the heat of reaction, assuming that the molten cladding forms molten oxides.

Fig. 26. Calculated Percent Reaction-Time Curves for Reactor Core with Various Quantities of Steam in Loss-of-coolant Accident Analysis

The principal difference between the behaviors of the two cladding metals is to be attributed to the different heats of reaction. Because of

the very low heat of the stainless steel-steam reaction, the time of core collapse was not greatly affected by the extent of reaction. In the case of

zirconium cladding, the time of collapse was greatly shortened because of the large amount of energy released by the zirconium-steam reaction (see ANL-7045, p. 67).

The extent of stainless steel-steam reaction at the times the core reached average temperatures equal to the melting point of (i) stainless steel cladding (1370°C), and (ii) the urania core (2800°C) is shown in Table XXV. Since no satisfactory model for the collapse of the core exists, the times shown in the table were taken to represent the lower and upper limits of those required for the core to collapse onto the lower support structure.

TABLE XXV. Calculated Time Required for Collapse of Core and Extent of Stainless Steel-Steam Reaction Based on the Average Nuclear Decay and Chemical Energy Content

(Analysis based on LOFT-type reactor utilizing 15-mil stainless steel-clad core.)

Steam Flow	Average Core Energy Sufficient to Melt Cladding Completely (1370°C)				Average Core Energy Sufficient to Begin to Melt Fuel (2800°C)			
	Collapse Time ^a (sec)		Metal Reacted ^a (%)		Collapse Time ^a (sec)		Metal Reacted ^a (%)	
	Case A	Case B	Case A	Case B	Case A	Case B	Case A	Case B
Unlimited	830	350	44	38	2560	1850	80	73
1000 lb/hr	910	400	18	10	2580	1860	65	68
100 lb/hr	918	407	2.0	1.0	2700	1970	20	14
No Metal-Water Reaction	920	410	-	-	2750	2010	-	-

^aCase A: Initial section temperatures 285°C.

Case B: Initial section temperatures are those at operating power (50 MW).

Even though the core collapses, the chemical reaction between cladding and steam will continue until either the temperatures are lowered or the supply of steam is depleted. The collapse and chemical reaction that would occur for the zirconium cladding were previously discussed.²² Similar behavior would be expected for the stainless steel cladding. Metal-steam reaction will proceed unless the core collapses into a pool of water; in this case, a quenching reaction could occur that would be terminated by cooling of the debris. The extent of reaction occurring in the quenching reaction can be estimated from the condenser-discharge and TREAT experiments. Results of stainless steel-water reaction studies by both methods indicated that the extent of additional reaction would be limited to

²²Chemical Engineering Division Semiannual Report, July-December, 1964, ANL-6925, p. 222; Reactor Development Program Progress Report, April 1965, ANL-7045, p. 67.

about 35%. If, however, the eventual collapse of the core should occur into an empty vessel, the reaction could continue with either steam or air, depending only on the availability of the oxidizing media.

b. Calculations of Heat and Mass Transfer in the Loss-of-coolant Analysis. In the analyses of loss-of-coolant accidents described above, the effects on reaction rates and core temperature of (i) local heat transfer between the core and flowing steam (and hydrogen), and (ii) local changes in the concentration of hydrogen in the steam have been neglected. Improved calculational techniques are being developed.

In a loss-of-coolant accident, the expected flow rates of steam through the core after blowdown are relatively low. The 1000-lb/hr flow rate used in the calculations for the LOFT core corresponds to a rate of about 0.3 lb/hr per fuel rod, a rate that would result in a laminar flow.

Equations for heat and mass transfer coefficients for laminar flow are usually functions of the distance from the entrance (in the direction of flow) until steady temperature and velocity profiles are developed. However, for gas flow in channels of small width, the flow profiles are quickly developed.²³ This allows the use of heat transfer coefficients that are functions only of the conductivity of the gas and the channel cross-sectional geometry. Similarly, the mass transfer coefficients are functions only of the gas conductivity and channel cross-sectional geometry. Thus the Nusselt Number hL/k and the Sherwood or Taylor Number (Nusselt Number for mass transfer), h_dL/D_g , are equal to each other and are functions only of the channel cross-sectional geometry. Here,

D_g = diffusivity of steam in the gas, cm^2/sec

h = coefficient of heat transfer, $\text{cal}/(\text{cm}^2)(\text{sec})(^\circ\text{C})$

h_d = coefficient of mass transfer, cm/sec

k = thermal conductivity of the gas, $\text{cal}/(\text{cm})(\text{sec})(^\circ\text{C})$

L = characteristic length, cm .

For heat or mass transfer to a gas in laminar flow from a tube wall of relatively small diameter, D ,^{24,25}

$$\frac{hD}{k} = \frac{h_dD}{D_g} = 3.65,$$

²³Eckert, E. R. G., and Drake, R. M., Jr., Heat and Mass Transfer, 2nd ed. McGraw-Hill Book Co., Inc., New York (1959), pp. 197-199.

²⁴Ibid., p. 194.

²⁵Jakob, M., Heat Transfer, Vol. 1, John Wiley and Sons, Inc., New York (1957), p. 459.

and for heat or mass transfer to a gas in laminar flow between flat plates a distance L apart,²⁶

$$\frac{hL}{k} = \frac{h_d L}{D_g} = 3.75.$$

These equations are based upon the wall temperature and cup mixing temperature (bulk average temperature) or average concentration of steam in the gas.

Heat and mass transfer coefficients have been computed for gas in axial laminar flow through 0.39-in.-diameter fuel rods spaced on 0.58-in. rectangular centers (LOFT core design) or for fuel rods 0.375-in.-diameter spaced on 0.60-in. triangular centers. In each of these cases, the ratio of the cross-sectional area of the gas space per rod to the cross-sectional area of the rod is 1.81. The coefficients are substantially the same in both cases:

$$h = 6.5 k [\text{cal}/(\text{cm}^2)(\text{sec})(^\circ\text{C})];$$

$$h_d = 6.5 D_g [\text{cm}/\text{sec}].$$

The same value of heat transfer coefficient was also obtained from the recent work of Sparrow et al.²⁷

The temperature and velocity profiles would be essentially fully developed in an axial distance which is less than the distance between fuel-element surfaces; in the cases calculated above, this distance would be less than about 1 cm. The effect of a rod spacer or support would be to disturb the developed profile; however, the profile would be redeveloped in less than a centimeter and, unless the spacers were closer than about one foot, their effect on the heat and mass transfer coefficients would be slight.

²⁶Eckert, op. cit., p. 195.

²⁷Sparrow, E. M., Loeffler, A. L., Jr., and Hubbard, H. A., Heat Transfer to Longitudinal Laminar Flow between Cylinders, Journal of Heat Transfer, Transactions of ASME, November 1961, p. 415, Series C, Vol. 83.

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