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REACTOR DEVELOPMENT PROGRAM PROGRESS REPORT

May 1961

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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 over-all view of progress. The budget classification is followed only in broad outline, and no attempt is made to report separately on each sub-activity number. Further, since the intent is to report only items of significant progress, not all activities are reported each month. In order to issue this report as soon as possible after the end of the month editorial work must necessarily be limited. Also, since this is an informal progress report, the results and data presented should be understood to be preliminary and subject to change unless otherwise stated.

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

The last six reports issued in this series are:

November 1960	ANL-6269
December 1960	ANL-6295
January 1961	ANL-6307
February 1961	ANL-6328
March 1961	ANL-6343
April 1961	ANL-6355

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I. WATER COOLED REACTORS (040101)

A. General Research and Development

1. Irradiation Studies

a. Examination of Irradiated Prototype SL-1 Plate - Examination has been completed on two prototype SL-1 fuel plates removed from the ANL-2 high pressure water loop at MTR. One plate was adjacent to the core of the reactor and the other was in an out-of-pile section of the loop. The plates were 16 in. long, 2 in. wide, and 0.12 in. thick. The fuel was fully enriched Al-18 w/o U alloy which was silicon-bonded on all sides with a cladding of X8001 aluminum alloy.

The plates were operated in the loop for 8 cycles of the MTR at a coolant water temperature of 415 to 425°F with an average pH of 5.9 during the first cycle to 7.0 during the final cycle. Maximum burnup at the hot end of the in-pile plate was 0.92 a/o of the total core atoms. Dimensional and weight measurements were taken on each plate before and after electrolytic descaling in a saturated solution of boric acid. The values obtained for the in-pile plate were compared with those obtained for the out-of-pile plate in an effort to determine the amount of irradiation swelling which occurred irrespective of corrosion. The results of these measurements are given in Table I.

Table I. Measurements on Irradiated Prototype SL-1 Plates.

Plate	Average Change in Length (mils/side)	Average Change in Width (mils/side)	Average Change in Thickness (mils/side)	Average Scale Thickness (mils/side)	Change in Weight (gms)
In-Pile	+10	-6 to +5.5	-3 to -6.5	3.5 to 11	-17.52
Out-of-Pile	-3	-2 to -5.5	-3 to -5.5	1 to 4.5	-10.38

Scale formation on both plates differed in amount, appearance, and tenacity. The scale on the in-pile plate was dark to reddish-brown in color, nonadherent in the first 6 to 8 in. of the plate, and had an average thickness of 3.5 mils per side on the cold end and 11 mils per side on the hot end. The maximum thickness of the scale in the hot end was 14 mils. The scale on the out-of-pile plate varied in color from light to dark blue, was very tenacious, and had an average thickness of 1 to 4.5 mils per side.

The average decrease in thickness for both plates varied from 6 to 13 mils and represents a maximum corrosion rate combined with swelling of 1.2 mils per month per side. A more representative corrosion value, however, independent of swelling, is one based on weight losses.

These amounted to corrosion rates of 7.4 mg/cm^2-mo. for the in-pile plate and 4.4 gm/cm^2-mo. for the out-of-pile plate. This difference in corrosion rate for the two plates results from the flaking of the scale on the in-pile plate and is probably due to the nucleate boiling which occurs on the first four inches of the hot end of the plate.

Thickness changes for the two plates were similar and did not indicate the presence of swelling. Length and width measurements, however, indicated the opposite. The average increase in length and width for the in-pile plate was 20 and 11 mils, respectively, compared to the out-of-pile plate which had an average decrease in length of 6 mils and an average decrease in width of 11 mils. No defects or abnormally swollen areas were visually detected after descaling the in-pile plate.

The increase in length and width and not in thickness of the in-pile plate in comparison to that of the out-of-pile plate indicates that any swelling which occurred in the thickness dimension was masked by accompanying corrosion. This is substantiated by the difference in corrosion rates of the two plates based on weight changes. The exact amount of swelling which did occur will be determined later by metallography.

B. EBWR

1. 100 Mw Modification - Reboilers

a. Repairs - The repair work performed by the vendor on the two reboilers has been limited during the past month since replacement material is not available. Minor operations, such as dismantling, have been done to get the units prepared for final repairs, but future work is contingent upon delivery to the vendor of new tubing and a new tube sheet for the No. 1 reboiler. As of May 18, 1961, the vendor informed the Laboratory that the tubing and the tube sheet material had not been received.

The tube sheet for the No. 2 reboiler, having been ordered some time ago, has been clad and drilled for the tubing. Further work on the No. 2 reboiler will continue when the new tubing is received. It is estimated that fifteen weeks will be required to complete repairs of both reboilers.

b. Reboiler Tubing

Reboiler No. 1 Tubing - Photomicrographs of tube sections which resisted 240 hours attack by 65% boiling HNO_3 show intergranular attack extending inward from the surfaces to a depth of 3 grains, i.e., approximately 0.007 in. In these samples of tubing the grain boundaries are outlined by continuous carbide networks of moderate density.

Reboiler No. 2 Tubing - Tubing samples from top, middle, and bottom portions of the upper pass of the reboiler were catastrophically attacked by 65% boiling HNO_3. Two of the three tube sections were found perforated after a 24-hour exposure. Over 50% of the metal from the tube was lost during the exposure.

Photomicrographs of the as-received reboiler tube sections revealed the reason for their lack of metallic ring (high damping factor). In addition to carbide networks surrounding the austenite grains, the tubing was badly attacked intergranularly. The severe intergranular attack and grain inundation are illustrated in Figure 1.

These investigations show that the tubing used in the two reboilers was defective initially. Subsequent manufacturing operations precipitated the catastrophic failures of all reboiler tubing. Transformation of austenite to martensite took place in rolling of the tube into the tube sheet. Precipitation of carbide networks around the austenite grains occurred, presumably as a result of prolonged thermal exposure at sensitizing temperatures.

2. Component Development

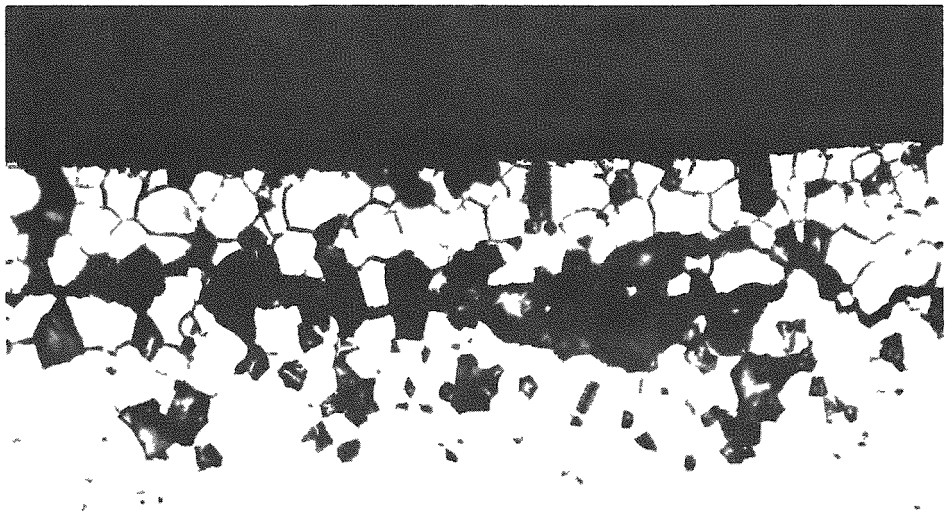
a. 17-4 PH Investigation - The wear tests of a spare rack, pinion and seal shaft, annealed at 1100°F for 4 hr and air cooled, are complete. The test results indicate that the softer material derived from H-1100 heat treatment is satisfactory for EBWR components. After 6000 cycles including 6000 full scrams dimensional checks revealed no wear.

b. Soluble Poison - Boric acid was used as a soluble poison in EBWR during recent cold critical tests and during low power operation. The high pressure addition system must be modified to meet operation requirements. The original design of this system called for a 1-gpm fixed rate pump and a 0 to $1\frac{7}{8}$ gpm variable rate pump. In actual operation at 600 psi the variable speed unit can only supply 0 to $\frac{1}{2}$ gpm. Two pumps, both with a 1.4 gpm fixed rate, will be used to supplement the variable rate unit. (A total rate of $2\frac{7}{8}$ gpm is specified by the Hazards Report* on soluble poison control of EBWR.)

Difficulty was experienced with both the continuous soluble poison monitor and with the manual soluble poison monitor method during reactor down times when the reactor was at atmospheric pressure. The purification system, including the continuous monitor, is under a slight vacuum under these conditions, and samples cannot be drawn from the system for manual monitor analyses. In fact, air is readily drawn in through opened sampling points. This holds up in the continuous monitor cell and contributes a significant error in the output of this device. The possibility

* Hazards Evaluation Report Associated with the Operation of EBWR at 100 Mw, ANL-5781-Addendum (Rev.) (To be published.)

A. Transverse Section



B. Longitudinal Section

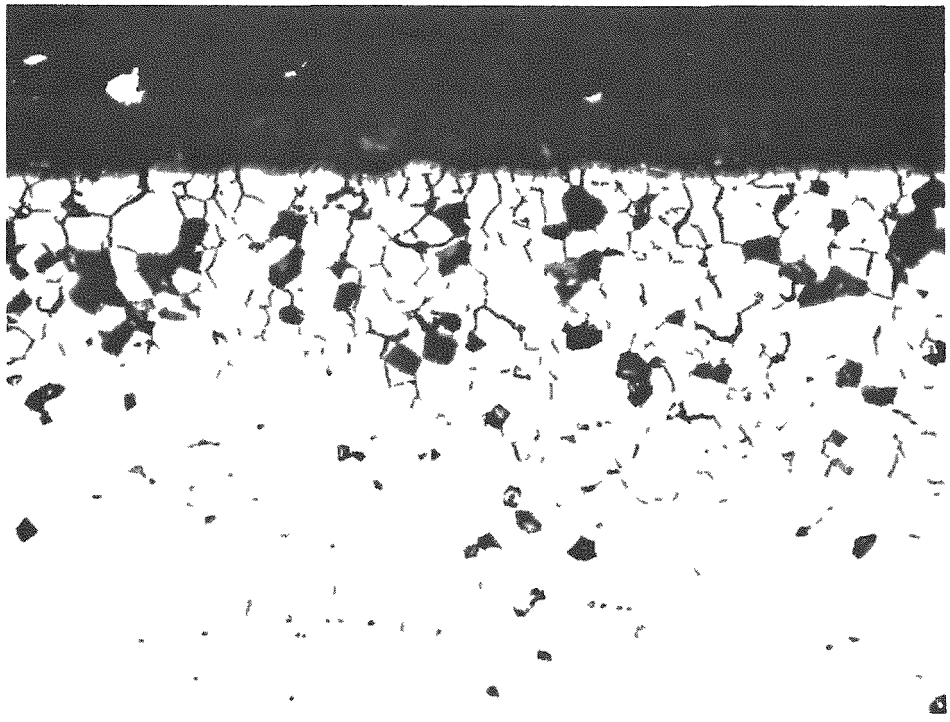


Figure 1

Photomicrographs of As-Received Reboiler Tube Sections Showing Severe Intergranular Attack and Grain Inundation

of relocating the purification system pumps relative to the purification system without affecting the shutdown cooling function of the pumps is being considered. With such a rearrangement, placing the demineralizers and filters downstream of the pumps, this system would be slightly pressurized at all times, allowing sampling operations and preventing in-leakage of air into the continuous monitor cell.

Preliminary laboratory experiments with a small continous electrodialysis unit indicates that the boric acid concentration in water flowing through the unit can be decreased. Further work will be necessary to determine the efficiency and feasibility of the technique prior to application to EBWR soluble poison control problems.

3. Core I-A

a. Zircaloy-2 Follower Control Rods - The expected delivery date of these new control rods is June 15, 1961. Zircaloy-2 angles for the followers have been cold formed to 90 degrees. Cold forming of the 2% boron-stainless steel was attempted; however, the plate broke at an angle of about 15 degrees. It appears that warm forming will have to be used on this sheet material as performed previously on the original Core I-A control rods.

4. Reactor Operation

a. Operating Experience - EBWR was operated at power levels up to 20 Mwt and with varying water levels. The riser or chimney was not in the reactor vessel but the highest water level maintained during these runs would have been above the riser had it been in place. These runs were made to check out the instrumentation placed within the vessel and core for future hydrodynamic studies. Modifications had to be made to some of the instruments and the check-out test will be re-run next month.

Resumption of reactor operation was made primarily for training the reactor operators and to check out the above instrumentation. This also permitted checkouts of other systems and circuits not possible with the reactor in the shutdown condition.

Control rod calibration permitted an estimation of burnup of Core I. This information is required for the IAEA participation in future EBWR programs.

Detailed critical experiments are being held up for lack of a final core loading. This will be possible when approval of the revised 100 Mw Hazard Summary Report is received.

b. Void Simulator - A void simulator was inserted in a central enriched thin fuel element with no difficulty. The electrical wiring for the associated air valve was completed and tested.

C. BORAX V

1. Installation of Reactor and Components

Installation and alignment of the repaired main bearings for the turbogenerator is complete and additional maintenance work on the turbogenerator is continuing. Painting in the turbine building is continuing and sandblasting of the walls and floor of the water storage pit is essentially complete. Installation of instruments in the reactor-building instrument room has started. The old core structure used with BORAX III and BORAX IV has been removed from the old reactor vessel and deposited in the burial ground. The measured activity of this core structure was about 250r and the removal and handling operation required detailed planning and preparation. The remainder of the old control rods and the old reactor vessel head have been deposited in the NRTS burial ground.

The purchase order contract with the Babbitt Electric Company for the installation of additional wiring and electrical equipment was completed early in the month.

Minneapolis-Honeywell representatives spent two weeks assisting in the testing and calibration of the process instrumentation and control system. Calibration of this system is continuing.

After the misalignment in the pump and motor shafts of the two feed pumps was corrected, the feed water system was made operational. The auxiliary water system, including the electric preheaters, is operative. The temporary hydrotest studs of the reactor vessel main flange have been replaced with the operational studs. After installation of clean-up screens on the suction side of the two feed pumps, auxiliary pump and the forced convection pump, the system received a preliminary flushing. The initial testing of the forced-convection pump is in progress at month's end. Representatives of the Worthington Co. and the Borg-Warner Co. were present.

The makeup-water demineralizer system has been completely checked out and is being used to furnish water for flushing and operational tests on the process piping system and equipment.

The following BORAX V reactor components (fabricated in the Laboratory's Shops), have been received at the site: all the standard chimneys, all the standard hold-down boxes, the forced convection baffle and forced convection seal rings, the two Belleville springs with upper and lower bearing rings, all the in-vessel instrument thimbles, the pressure vessel dowels, the control-rod-drive-nozzle shielding plugs and the complete boiling core structure including core support plate, shroud, legs, struts, and top ring flange. It is estimated that the rest of the components (core structures, fuel boxes, control rod extension shafts, etc) will all be ready for shipment at various times up to July 15.

2. Procurement and Fabrication

A complete assembly and inspection of the boiling core structure and the central superheater core structure was conducted at the Laboratory's Central Shops prior to shipment of these items. A photograph of the central superheater core structure is shown in Figure 2.

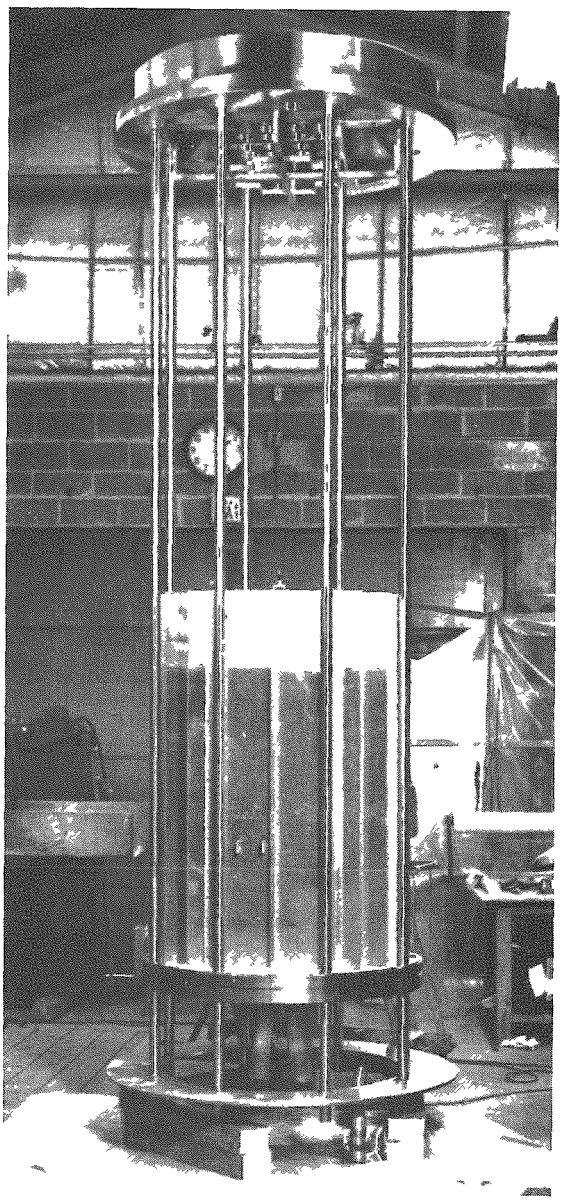


Figure 2

BORAX V Central Superheater Core Structure

At the Idaho Division Shops, the three core-structure storage stands, the core-structure lifting fixture, the forced-convection-baffle lifting fixture, and the boiling-fuel-assembly loading fixture have been completed. An order has been placed with the Davison Chemical Company for a depleted uranium extension section for the existing BORAX coffin. This extension is necessary to give adequate shielding for the superheater fuel assemblies. Material has been ordered for the superheater flood and drain outlet piping, superheated steam drain lines and traps, makeup-water polishing demineralizer system, reactor-water demineralizer system, fuel-storage demineralizer system and the alternate preheat piping system.

Test data have been received from the supplier on the twelve in-vessel thermocouples. A two-pen recorder and two differential pressure cells have been ordered for the instrumented superheater-fuel-assembly flow-measuring system.

3. Design

Detailed design of the instrumented boiling fuel assemblies and pressurized terminal boxes for instrumented fuel assembly leads was completed. Storage boxes and racks for radioactive storage of control rods, chimneys and hold-down boxes in the old reactor pit, and an extension spool for the old reactor vessel to provide an extra two feet of water shielding over stored radioactive superheater fuel assemblies, have also been designed. Design was completed on the superheater flood and drain outlet piping, the drain lines and traps for the superheated steam lines, a layout of instrumentation and sampling lines in the upper reactor pit, a portable cover for the reactor vessel and a working platform for the upper reactor pit. Detailed design of the revised control rods was also completed.

A preliminary design of equipment for measuring the activity distribution of flux wires has been made. Plans have been made for calibration of a cadmium-ratio method for measuring void fractions. A fuel-water lattice using BORAX V boiling fuel rods will be used in this experiment.

A series of RE-147 problems is being analyzed to investigate the validity of the assumption (made in the analysis of the maximum accident) that all heat generated in the boiling zone during an excursion remains in the oxide fuel. Several RE-129 problems have been run and analyzed in order to assess the severity of an accident in which all four intermediate control rods are moved out of the core at high speed rather than at low speed. The resulting excursion can be terminated by a high-flux scram.

A number of PDQ problems have been run and are being analyzed in order to determine the worth of aluminum-clad intermediate control rods. MUFT, SOFOCATE, and DSN problems based on a stainless steel-clad control rod have been run. The purposes of these problems are to determine whether additional PDQ problems on the worth of stainless steel-clad control rods are needed and to furnish input for such PDQ problems if they are needed.

4. Development and Testing

a. Control Rods - Static corrosion tests are being run at 600 psig saturated conditions on Type 304 stainless steel-clad Boral samples simulating the new reference control rod material. Measurements taken on three samples after 11 days of testing indicated that the thickness had been reduced by 0.002 to 0.004 in. It is presumed that this reduction is caused by the external pressure.

Vibration tests on a sample boiling fuel rod in the air-water loop have finally been completed. After the extraneous vibrations of the test stand and mockup boiling fuel assembly box were measured, it was determined that the actual maximum amplitude of vibration of the rod at flow conditions of 20 fps and 50 vol.% in voids is approximately 0.01 in. Vibrations at lower velocities and lower voids is much reduced. The stress induced in the 0.015-in. thick stainless steel cladding by the maximum vibration is low and the possibility of a fatigue failure is very remote. As a result of these tests, the decision has been made not to install a center grid in the boiling fuel assembly.

A preliminary inspection has been made of the four old BORAX X-8001 aluminum-clad Boral control blades with riveted-on hafnium tip and one aluminum-clad cadmium blade. The blades were found to be coated with a uniform reddish-brown deposit. The maximum activity found on the control rods was 1.5r, measured at the hafnium tip and at a point on the extension shaft about 2 ft above the top of the blade. The nominal original thickness of the aluminum-clad Boral section of the blades was 0.375 in. The maximum thickness measured during this inspection was 0.470 in. There was no evidence of severe corrosion or failure of the blades. The extension shaft was cut off one Boral blade and the cadmium blade and these blades will be examined thoroughly.

The wear test on a chrome plated, 17-4 PH control rod extension shaft heat-treated at 1150° F was continued. Inspection of the shaft and seal rings after 750 motor-driven cycles and 1000 scram cycles revealed no significant signs of wear.

b. Superheat Fuel Elements - Twelve additional plates have been received from AI and are now being evaluated. Nondestructive testing of these plates is completed. Plate thickness, squareness, and flatness, and core length is within tolerances in all cases. The side cladding width, as measured from radiographs furnished by AI is satisfactory for the six type HCD plates; however, five of the six type FPD plates appear to have less than the minimum required side cladding. End cladding length, as measured from the radiographs is slightly excessive on one plate. Nine of the plates have very shallow markings on the surface and one plate has a deep pit in the face cladding which appears to have been caused by a foreign particle imbedded during rolling operations. The type HCD plates do not have the specified core length symbol.

No radioactive surface contamination was found on the plates. Nondestructive measurement of core-to-clad and clad-to-clad bond integrity gives no evidence of nonbonded areas. Measurements of gamma energy emission were taken from five locations on each plate. Measurements taken near the ends of the core area are lower than those from other locations, probably because the thickness of the core tends to taper at the ends. Evaluations of homogeneity based on the counts taken from the three less extreme locations indicate that three type HCD plates and one type FPD plate have more inhomogeneity in the core than the maximum specified. Some of the inhomogeneity indicated may be caused by plate and face cladding thickness variations.

Samples have been taken from the plates for destructive testing purposes. End cladding coupons are being Strauss tested for susceptibility to intergranular corrosion. Metallography is being performed on specimens for bond integrity, cladding thickness, and oxide fragmentation and stringering. Face cladding was removed from five 1 in. square samples from each plate and the declad specimens are being analyzed for weight percent uranium.

No estimate of the delivery schedule for the production superheater fuel plates can yet be made.

Static corrosion tests in 540° C steam on brazed samples of Coast Metals alloy 60 and stainless steel Type 304 still show no signs of corrosion after two months.

II. SODIUM COOLED REACTORS (040103)

A. General Research and Development

1. ZPR-III

Experimental work on Assembly 34 was completed. This is a large, dilute carbide assembly containing one column of enriched uranium, two columns of depleted uranium, two of graphite, six of reduced- density aluminum and three columns of stainless steel per drawer. The critical mass of this composition was 503.01 kg of U^{235} and the critical volume 574.5 liters. (In the April 1961 Progress Report, ANL-6355, the enrichment of this assembly was incorrectly reported as about 3 percent. The core was about 31 percent enriched.)

Experiments carried out this month concerned central reactivity coefficients for both fissile and nonfissile materials, radial fission traverses with U^{235} and U^{238} counters, reactivity effect of inhomogeneity of fuel, measurement of the Rossi-alpha, depleted uranium foil irradiation and the distributed worth of the constituents of the core including U^{235}.

Central reactivity coefficients for this assembly are given in Table II.

Table II. Central Reactivity Coefficients of Various Materials in Assembly 34

Material	Sample Wt.	Reactivity Change Due to Sample (Inhours)	Ih/kg
U^{235}	268.42 g U^{235}	28.6	106.8*
	19.36 g U^{238}		
U^{233}	221.2 g U^{233}	45.5	205.6*
	5.3 g U^{238}		
Pu^{239}	186.0 (94.5% Pu^{239})	32.20	182.4
U^{238}	2455 g U^{238}	-13.66	-5.78*
	4.92 g U^{235}		
Al	350.64	0.329	0.937
Al_2O_3	355.12	2.87	8.08
Ag	1366.05	-60.4	-44.2
Be	240.7	20.4	84.8
Bi	1276.2	-0.477	-0.374
B_4C (enriched)	16.087	-20.7	-1286.
C	196.42	7.19	36.6
Cr	439.2 g Cr	-0.413	-0.603**
	102.4 g SS†		
Fe	1028.0	-1.36	-1.32
Hg	1383.4 g Hg	-7.30	-5.07**
	197.3 g SS		
Li	54.9 g Li	-4.77	-81.8**
	195.1 g SS		
Mo	1279.8	-12.14	-9.49
Nb (Cb)	489.6 g Nb	-7.98	-16.0**
	102.4 g SS		
Na	91.2 g Na	0.37	7.17**
	195.0 g SS		
Ni	1150.4	-2.78	-2.42
Pb	1474.9	-0.14	-0.094
Ph-I	232. g Ph-I	-3.67	-14.8**
	168 g SS		
Ph-II	210 g Ph-II	-3.21	-13.4**
	275 g SS		
Stainless Steel	1017.2	-1.48	-1.45
Ta	1004.4 g Ta	-28.9	-28.7**
	102.4 g SS		
Th	1488.47	-18.9	-12.7
Y	582.1	-8.69	-14.9
Zr	846.0	-1.03	-1.21

* Reactivity of principal isotope only

** Corrected for effects of stainless steel can

† Stainless Steel

Since these central reactivity measurements require opening and closing the halves of the reactor, the reproducibility of the values given for the reactivity change due to the sample is about ±0.5 inhour. As a result, the inhours/kg values given for some materials, such as aluminum and sodium, are questionable. On the whole, the central reactivity coefficients measured in Assembly 34 are quite similar in both sign and magnitude to those measured in Assembly 29, the large dilute, oxide core.

The radial fission traverse results are given in Table III.

Table III. Radial Fission Traverse Results in Assembly 34

	Normalized Count Rate	
Distance From Center (in.)	U^{235}	U^{238}
0	1.00	1.00
1	0.981	0.965
2	0.969	0.973
3	0.994	0.956
4	0.935	0.951
5	0.936	0.948
6	0.915	0.909
8	0.854	0.842
11	0.763	0.721
14	0.609	0.575
15.2 (core-blanket interface)		
17	0.468	0.387
20	0.272	0.113
24	0.0967	0.0187
28	0.0475	0.004

The unexpectedly high count rate at position 3 for the U^{235} counter is probably due to noise.

Several normalized count rates were obtained from the neutron flux calculated by the DSN code using the 16-group cross section set of Okrent and Yiftah. Agreement between the experimental and calculated values is satisfactory except near the core-blanket interface.

The inhomogeneity effect of the fuel was measured by bunching the fuel in a twenty-drawer, wedge-shaped section of the core. The enriched uranium was removed from 10 drawers and placed next to the enriched uranium in the other 10 drawers. The assembly gained 21.8 inhours of reactivity. A linear extrapolation to the entire core indicates that a gain of 475 inhours would result from placing the fuel in $\frac{1}{4}$ in. columns rather than the $\frac{1}{8}$ in. columns. Similarly, in the same twenty-drawer wedge section, the fuel

was separated into two $\frac{1}{16}$ in. columns per drawer. The assembly lost 9.83 inhours. A linear extrapolation to the entire core indicates a loss of 213 inhours by separating all the fuel into $\frac{1}{16}$ in. columns.

The normal arrangement of uranium and graphite in Assembly 34 was depleted U, graphite, enriched U, graphite, and depleted U. By interchanging the position of the graphite and depleted uranium plates in the twenty-drawer wedge section of the core, the assembly gained 7.07 inhours. When extrapolated to the entire core, a gain of 154 inhours is indicated.

Radiochemical analysis of depleted uranium foils placed at the center of the core indicate a U^{238}:U^{235} fission ratio of 0.04. As measured by fission chambers, this ratio is 0.034. The ratio of U^{238} capture to U^{235} fission was 0.12.

Rossi-alpha measurements give a value of $\alpha = 2.92 \pm 0.07 \times 10^4\ \text{sec}^{-1}$.

Core material substituted for blanket at the edge of the core is worth 17.8 inhours per kilogram of U^{235}.

The distributed worths of the materials in the core are listed in Table IV. These values were obtained by changing the quantity of materials in a wedge section of the core.

Table IV. Distributed Worths of Materials in Assembly 34

Material	Inhours/kg
U^{238}	-0.846
U^{235}	44.08
Aluminum	4.65
Stainless Steel	1.66
Graphite	16.3

Multigroup calculations using the 16-group Okrent-Yiftah set give a k value of 1.046 for the just-critical core. The central fission ratios calculated from the multigroup fluxes are larger than the corresponding measured values. Table V gives both the experimental and calculated ratios.

Table V. Calculated and Experimental k Values in Assembly 34

Ratio	Measured Value	Calculated Value
$\sigma_{238}/\sigma_{235}$	0.03391	0.0396
$\sigma_{234}/\sigma_{235}$	0.2467	0.3035
$\sigma_{233}/\sigma_{235}$	1.4544	1.5384
$\sigma_{236}/\sigma_{235}$	0.08035	0.0968
$\sigma_{239}/\sigma_{235}$	1.0667	1.1335
$\sigma_{240}/\sigma_{235}$	0.2709	0.3027

2. ZPR-VI and ZPR-IX

a. Building - Construction of Building 315 is now essentially completed.

In conjunction with replacement of the thrust bearings on the cell doors, adapter rings were installed which will apply the door load more evenly to the bearings. Also a roller thrust bearing having a larger thrust capacity is now being used instead of the ball thrust bearing.

Further leak testing of air locks and cells is being delayed pending delivery of an inflatable seal. Meanwhile pipe and conduit penetrations into the cell are being sealed.

b. Procurement

(1) Bed and Table Assembly - The bed and table assemblies for ZPR-VI and ZPR-IX were received in early May. Both beds are being installed in their respective reactor cells. The tables will be stored until the cells have been pressure tested and accepted from the contractor. Approximately 50% of the stainless steel square tubing to be used as the matrix has been received from the vendor. All of the tubes received seem to be satisfactory. The vendor for the aluminum tubing is having difficulty meeting specifications and has suggested alternate specifications which are now being studied.

(2) Control and Safety Rods - A vendor has been selected for the fabrication of the drive mechanisms for the dual purpose control and safety rods. Delivery will be approximately four months after the order has been placed with the vendor.

Bids are being invited from prospective vendors for the fabrication of the blade-type insertion safety rod drive mechanisms.

(3) Control Console - A meeting was held with the vendor to review the layout of the control console. Details concerning the interlock circuitry were also clarified during the discussions.

(4) Miscellaneous - Bird cage-type storage units are being designed for the storage of the highly enriched uranium fuel. Approximately 58% of the U^{235} fuel plates have been received from the vendor.

Depleted uranium for use as dilution plates and blanket are being coated with a KEL-F-800 plastic coating. Approximately 50% of the depleted uranium has been processed by the coating machine.

All the fabricated components for the working or loading platform are being held in storage by the vendor. A delay in receiving the motors to drive the platform will cause an over-all delay of about three months.

c. Hazards Summary Report - A revised draft of the Hazards Summary Report for ZPR-VI has been submitted for review by the Laboratory's Reactor Safety Review Committee. It is scheduled for review early in June.

The meltdown accidents discussed in the report have been carefully re-examined and re-calculated during the last few months. The actual power distribution across the reactor core was substituted for the previous uniform melting hypothesis. It was also recognized that trapped air, largely from the holes in the low density aluminum, will restrain the degree of initial collapse of the core upon melting. A very important conclusion resulting from this approach is that mockups of power reactor systems on ZPR-VI can collapse to prompt critical upon meltdown only under very restricted conditions.

The effect of core size on burst energy yield was examined by comparing a meltdown accident for a 400 liter fast ZPR-VI core to the same accident previously calculated for the 1200 liter core when both cores have the same composition (except for enrichment). The smaller core had a burst yield which was smaller than that of the 1200 liter core by a factor of five.

d. Experimental Program

(1) Perlow Spectrometer - In previous work with the Perlow counter, the accidental counting rate has been estimated by brief operation at a power level sufficient to cause dominance of accidental coincidences over true events. Theoretically it should be possible to determine the accidental rate accurately by delaying the pulse from one counter by 3 to 4 microseconds relative to the other. To test this idea, a circuit with a delay of approximate 3- microsecond is being constructed.

(2) Methane-Type Recoil Spectrometer - The new filling system (see April Progress Report) to be used in connection with the recoil spectrometer has been assembled and checked for tightness and cleanliness. The counters prepared earlier for tests on CP-5 have been refilled with a mixture containing two atmospheres of methane and traces of nitrogen for

calibration purposes. Some data have been collected showing the degree of resolution of the counter at elevated gas pressures. Since the tests were carried out at the ATSR, it is likely that fast recoils were observed along with signals from the $N^{14}(n,p)C^{14}$ reaction, forming a smooth background above which the signal from the 600 kev protons from nitrogen was well visible. In order to eliminate this interference the counter is now being readied for a pure nitrogen filling. These measurements were made using neutrons from a natural uranium converter placed in front of ATSR.

(3) Miniature Fission Counters - Miniature fission counters containing small amounts of Pu^{240}, Th^{232}, and Np^{237} have been completed. These will be used to determine spectral indices in assemblies studied on ZPR-VI.

(4) Miscellaneous - Procurement of small quantities of various elements for fabrication into danger coefficient measurement samples for ZPR-VI has been started.

Since the pile noise technique is inadequate for prompt neutron lifetime measurements for cores containing large amounts of U^{235}, a study is under way to develop electronic equipment for Rossi-α measurements.

B. EBR-I

1. Fabrication of Mark IV Core

All Inconel-X and stainless steel hardware for noninstrumented rods has been finish machined. Commercially procured Zircaloy-2 wire, $\frac{1}{16}$ in. diameter, was accepted for the balance of the jacket tube rib wire. Straightening, eddy current inspection and acid pickling operations were performed on 2100 feet of this wire. All Zircaloy hardware other than jacket assemblies has been finish machined. Jacket assemblies and blanket uranium slugs required for assembly of 73 blanket rods were completed in May.

Instrumented fuel and blanket assemblies require thirty feet of 0.080 in. O.D. x 0.050 in. I.D. Zircaloy-2 tubing for thermocouple rods. Commercial fabricators have not been able to supply this tubing. The major difficulty in producing acceptable tubing of this size is due to the generation of severe I.D. cracks as the tube is reduced below about 0.120 in. outside diameter. It is believed that cracking is caused by a combination of compression shear stresses and the presence of stringers and other inclusions. Adequate intermittent annealing during cold reduction should eliminate failure from compressive stress during sinking operations. Microscopic examination of numerous samples has revealed that stringers or inclusions do not diminish in size, proportionately, as tube reduction takes place and may therefore lead to gross inhomogeneity in the final product.

In view of the importance of starting with material as free of defects as possible, several lengths of sound 0.299 in. O.D. x 0.020 in. wall Zircaloy-2 tubing that had been eddy current inspected were drawn down to the desired finish size of 0.080 in. O.D. by 0.015 in. wall. Inspection of this tubing revealed cracks on the inside surface extending to a maximum depth of 0.005 in. In general, however, the tubing appeared to be of higher quality than that obtained commercially. A sufficient amount of acceptable 0.299 in. O.D. tubing is on hand to produce the required footage of 0.080 in. tubing.

The embrittling effect of hydrogen on zirconium is well known. Heat treatments designed to lessen hydrogen embrittlement have been conducted on one ANL extruded tube and on two commercially supplied lengths of tubing. There is some indication that cracking can be substantially reduced by this means.

2. Properties of Plutonium-Aluminum Alloys

A series of Pu-Al alloy specimens (composition range 2.25 to 21.63 a/o Al) whose expansion coefficients were measured in the as-cast condition have been homogenized at 450°C under vacuum for 145 hours. Their expansion coefficients in the homogenized condition will be determined.

Four additional slump tests were conducted as part of the program to evaluate certain engineering and physical properties of the plutonium rich plutonium-aluminum alloys. The Pu-1.25 w/o Al (10.07 a/o Al) composition was selected for these tests, since it is the fuel alloy for the Mark IV loading of EBR-I. It was the purpose of these tests to determine the slump behavior of this composition on repeated heating and cooling while under a static load.

The four specimens selected were identical in size (0.235 in. in diameter by 1.500 $\pm$ 0.001 in. long) and as identical in composition as the injection casting technique is capable of producing. Two of the specimens, in the as-cast condition, were subjected to four heating and cooling cycles while supporting static loads of 7.2 psi and 20.3 psi respectively. The remaining two specimens were homogenized at 450°C under vacuum for 145 hours before being subjected to five heating and cooling cycles under the same load conditions. Each cycle was approximately of ten hours duration. The results of these tests show that perceptible slumping ($\Delta L > -0.50$ mils) for the Pu-1.25 w/o Al (10.07 a/o Al) composition in the as-cast condition will occur during a three hour holding period at 400°C on the first heating cycle. An appreciable increase in slumping can be expected in the temperature range 425-500°C for comparable holding periods. However, the amount of slumping which occurs at any given temperature in this range for a specified holding time will decrease regularly on successive heating cycles. The maximum slumping observed under a static load of 7.2 psi occurred on the second heating cycle during a $3\frac{1}{4}$-hour holding period at 450°C, and amounted to 1.3 mils. Under a static load of 20.3 psi a similar specimen slumped 1.2 mils on the second heating cycle during a $3\frac{1}{4}$-hour holding period at 425°C.

Specimens of this composition which had received the 450°C homogenizing heat treatment showed no perceptible slumping below 475°C. Up to 500°C the amount of slumping observed during a three hour holding period is of the order of 1 mil. The amount of slumping occurring at 500°C for specific holding times decreases on successive heating cycles. An appreciable increase is noted in the amount of slumping which occurs at 550°C for a comparable three hour holding period, and the rate of decrease in slumping at this temperature on successive heating cycles is noticeably less. At 600 and 640°C the amount of slumping observed during a one to two hour holding period varied with the static load and ranged between 23 and 40 mils.

On the basis of these results it appears that the load-carrying capability of the Pu-1.25 w/o Al (10.07 a/o Al) alloy can be enhanced by a homogenizing heat treatment at 450-500°C. Although the lower limit of the holding time for such a heat treatment has not been established, a holding period of 4-5 hours seems reasonable. This is predicated on the fact that the amount of slumping observed in specimens in the as-cast condition at temperatures below 500°C decreases noticeably on successive heating cycles using 3 hour holding periods at temperature.

C. EBR-II

1. Construction

The approximate status of construction contracts as of May 18, 1961 was as follows:

Building	% Completion
Power Plant (Package 2)	100
Reactor Plant (Package 2)	100
Sodium Boiler Plant	95
Fuel Cycle Facility	90

a. Sodium Boiler Plant - The X-ray examination of the secondary sodium piping referred to in the Progress Report for April (ANL-6355) is complete; approximately 1400 negatives were made. About three hundred negatives indicate unacceptably dirty conditions. These have been transmitted to the Idaho Operations Office for information and action.

It was indicated in the April Progress Report that chemical analyses of three piping samples had been received from the Contractor and that these analyses indicated that the samples were SAE-4130 rather than the $2\frac{1}{4}$% chromium-1% molybdenum specified. The contractor has now conducted an investigation of this matter and reports that the original analyses were erroneous owing to a "mix-up of samples" by the analytical laboratory, and that reanalysis by the original laboratory, and referee

analyses by a second laboratory, indicate that the piping material actually does meet specifications ($2\frac{1}{4}$% chromium-1% molybdenum). He has also submitted chemical analyses on seventeen additional samples, one from each of seventeen heats of piping material sampled at start of shop fabrication (about 10 months ago) These analyses indicate sixteen of the samples to be $2\frac{1}{4}$% Cr-1% Mo and one to be $1\frac{1}{4}$% Cr-$\frac{1}{2}$% Mo, all as specified.

Submittal by the Contractor of chemical analyses for the secondary system piping is still considered unsatisfactory. In addition to the "mix-up" on the first three samples, it is noted that a second "mix-up" occurred on the single sample of the second submittal referred to above. The report from the analytical laboratory to the Contractor first indicated this to be $2\frac{1}{4}$% Cr-1% Mo but then subsequently was changed, before submittal to the Commission, to show it to be the required $1\frac{1}{4}$% Cr-$\frac{1}{2}$% Mo and the same explanation of "mix-up" quoted. These circumstances, with no detailed explanation of them yet available, engender some doubt as to the probable accuracy of the submittals. In addition, the first three samples remain unidentified; the Contractor has not stated to what piping lots or to precisely what locations in the piping system they relate. The seventeen samples of the second report have been identified by heat (in this case, equivalent to lot); however, these plus the first three samples do not represent the total required by the specifications to be analyzed and submitted (one from each piping lot or about thirty). In addition, the contractor apparently is unable to identify precisely the locations in the piping system to which these seventeen samples relate, thereby eliminating the possibility of resampling the pipe for checking. In view of these factors, it is necessary to assume that, until these problems are resolved, conformance of the installed piping with the specifications is uncertain.

In order to determine with a higher degree of assurance whether non-specification material is in the sodium-boiler plant piping, development of a non-destructive testing device which may be capable of identifying $2\frac{1}{4}$% chromium-1% molybdenum material has been initiated. Hopefully this instrument may be useful in checking weldments. Each piece of pipe, fitting, etc., in the piping system will be inspected if and when the device can be used with confidence.

2. Installation of Equipment - Package 4

All Package 4 pre-dry critical installation work by the contractor was completed in the reactor building May 12, 1961. Preparations for the dry critical experiments were then started by Laboratory personnel.

Figure 3 is a photograph taken in the reactor plant at the conclusion of the contractor installation work. Visible in the center of the photo are the control rod drive mechanisms, the reactor vessel cover lifting mechanism upper structure, portions of the gripper and hold-down mechanisms,

and the drive motor for one of the primary pumps. The structure at the right is the storage basket elevating mechanism; in front of it is the fuel transfer arm. Descriptions of the design and functions of all these devices have been provided in earlier Progress Reports.

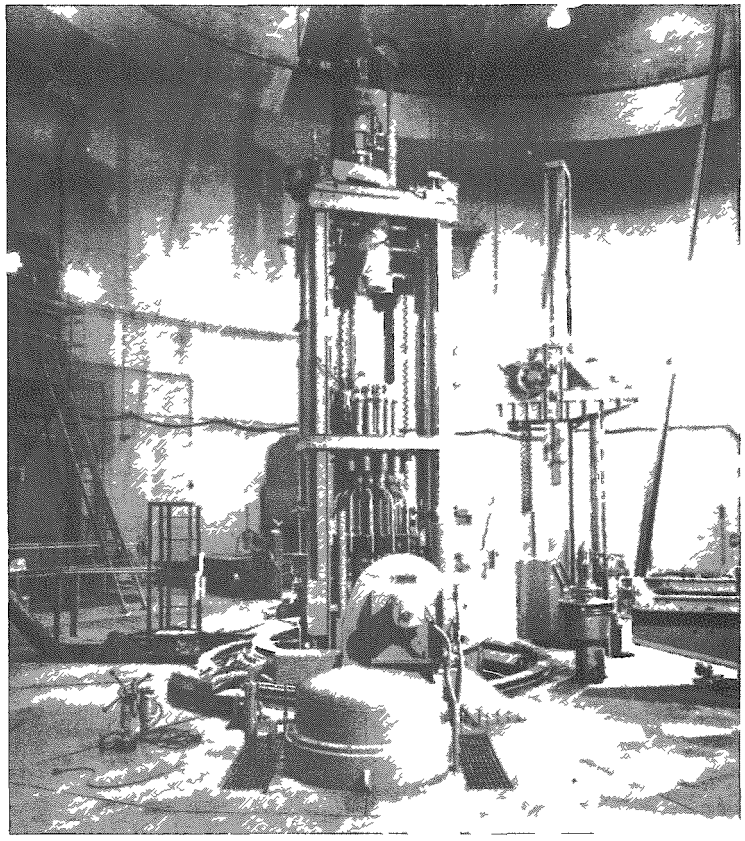


Figure 3

EBR-II: Interior of Reactor Plant

The only means of personnel access to the inside of the primary tank now available is through the intermediate heat exchanger nozzle. A ladder extending through this nozzle may be seen in the figure. At various times during the dry critical experiment period, this ladder will be removed and a shielding plug equipped with a large air fan installed in the nozzle. The shielding is required whenever the reactor is to be made critical. The purpose of the fan is to circulate primary tank air through the reactor to aid in establishing the isothermal temperature conditions desired during certain experiments.

In both Figure 3 and Figure 4 the checkered plate which forms the operating floor directly over the primary tank and the other depressed areas may be seen in place.

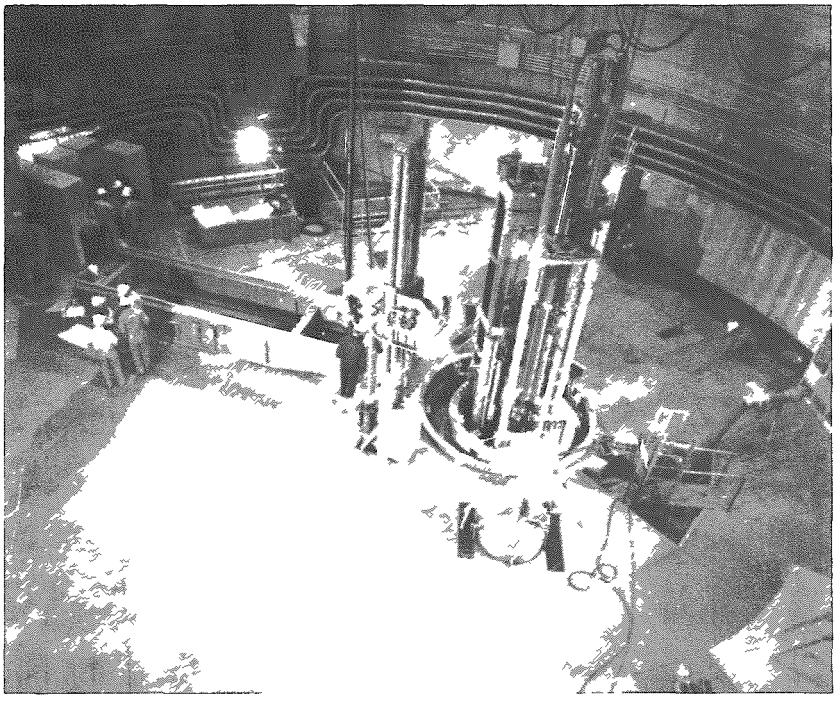


Figure 4

EBR-II: Over-all View of Center Area of Reactor Plant

Photographs of the areas under these plates containing the cables, conduit, junction boxes, piping, valves, motors, etc., required for operation of the mechanisms and equipment in and on top of the primary tank have been presented in previous Progress Reports.

To the left of the storage rack mechanism in Figure 4 may be seen the beam supports and rails for the fuel unloading machine and the Dry Critical subassembly loading rig. The latter is a temporary, manually operated device which is to be used to expedite fuel handling operations during the Dry Critical Experiments. It bypasses the storage rack by loading directly to the transfer arm. The fuel unloading machine, which is to be a permanent part of the plant and is required for fuel loading operations after sodium is introduced into the primary system, will be installed after the Dry Critical Experiments have been completed.

Figure 4 also shows one of the two festoon cables mounted on the wall. These cables contain instrumentation, power, and control leads for those mechanisms mounted on the rotating plugs which must remain operable at all times irrespective of plug rotation. The festoon arrangements permit permanent connection of the cables to the plugs with plug rotation accommodated by winding or unwinding of the cables around the peripheries of the plugs. To the left of the fuel unloading machine rails is the console for the fuel handling center. It is from this console that the various mechanisms required for fuel handling are operated.

Directly in front of the man at the extreme right of Figure 4 may be seen the handle of the manually operated switch for the auxiliary pump. The pump is located in the primary reactor outlet piping and is submerged in the bulk sodium in the primary tank. The purpose of the switch is to disconnect the pump from its power supply. The standby batteries for the auxiliary pump power are located below the floor grating and along the wall. Barely visible next to the gas welding rig by this grating is Instrument Center No. 1 (for the auxiliary pump). The cable trays and conduit all along the wall and the electrical cabinets behind the floor grating over the standby batteries contain terminal blocks for the control and power conductors and cables running away from the primary tank. The other cabinets to the right of the floor grating constitute Instrument Center No. 2, associated with the nuclear and process instrumentation in the primary tank.

3. Procurement

A number of shipments of reactor fuel and blanket subassemblies have been made to the site in the past month. The entire outer blanket has been shipped. Also, most of the special in-core hardware such as dummy subassemblies, safety rod and control rod guide tubes, the central thermocoupled subassembly, etc., have been shipped. It is expected that the core subassemblies and inner blanket subassemblies will be shipped by early June. All components for assembly of the neutron sources required for Dry Critical have been shipped, and the antimony rods have been irradiated at ETR and sent to the EBR-I site for assembly.

All special out-of-core devices required for Dry Critical have been shipped except the rod oscillator. This includes such devices as the periscopes, lights, mirrors, closed circuit TV, hardware, fans, etc.

4. Component Development - Instrumentation

a. Control Rod Reactivity Generator - An analysis was begun to determine the requirements for achieving a reduction in the input time constant and stability of the amplifier in the nulling circuit when low power transfer functions are measured.

The drawings of the oscillator rod have been completed and fabrication has begun. Layouts of the oscillating mechanism have been started. A mechanism which is essentially a "Scotch Yoke" will provide a $\pm$4 in. vertical stroke up to 2 cycles per second.

b. Automatic Control System - Transport delay times were determined for the steam from the steam drum to the turbine, and an analysis of the steam loop controls for transient response times was initiated.

The transfer function approximation to the temperature feedback (Progress Report for April, ANL-6355, p. 26) was used to establish a set of operating lines for the reactor. These show how the gain constant of the temperature feedback path varies with primary coolant flow rate and power level of the reactor.

A "normal operation" gain constant line was established and was found to be approximately linear with power levels from 10% to 100% of reactor power. It was found that the time constants in the temperature feedback path could be approximated with one non-linear time constant in the denominator dependent on the flow rate only.

The use of the above model on an analog computer will facilitate the study of reactor operation upon a change of steady state power level, using manual or automatic control.

5. Component Development Fuel Reprocessing Facilities

The Fuel Cycle Facility that is being constructed as a part of the EBR-II project is 90 percent completed. Installation of ANL-furnished equipment is continuing together with installations of building services.

Much of the processing equipment for the Air and Argon Cells has been purchased. Two melt refining furnaces are being manufactured; progress in their manufacture is considered satisfactory. The purchase order for service sleeve feed-throughs has been placed. Design of the transfer locks has been completed and drawings have been released for bids.

The defects that were found in the toggle switch control boxes (see April Progress Report) for remote control of cranes and manipulators have been corrected. The toggle switch boxes are now considered satisfactory and are being prepared for shipment to Idaho. Manufacture of manipulator carriages is progressing satisfactorily; inspection of the carriages in the General Mills' shops has been scheduled.

Bearings lubricated with radiation-resistant greases have been alternately irradiated and run at 40 rpm under 800 lb axial load in an air atmosphere. The results of this test indicate that the useful life of the bearings in the manipulator bridges in the Air Cell will be one year. The more rapid deterioration of the grease is attributed to the manner of its irradiation. The grease was irradiated as a thin film and not as a bulk sample. Thin films deteriorate more rapidly under irradiation, particularly in an air atmosphere. Since the grease retainers on the bridge bearings will contain a larger quantity of grease, it is anticipated that the operating life of the bearings will be considerably greater.

Upon heating crucibles in the melt refining furnace, large temperature gradients between the crucible wall and bottom may produce thermal stresses which are sufficiently severe to crack the crucible. The incidence of cracking of the crucibles may be reduced by applying power to the furnace in small steps, thereby substantially reducing the temperature gradients in the furnace.

Design work has been started on process equipment for the skull recovery process.

The design of a prototype subassembly dismantler has continued. The dismantler will be used to remove the fuel elements from an EBR-II fuel subassembly. Some operations are being tested with functioning experimental models while the over-all arrangement is being checked out on a non-operational model. Fabrication has started on one of the cutting devices and about half of the complete dismantler should be released for fabrication during June.

One of the principal units of the operational fuel element decanner, the Spiral Decanning Unit, has been ordered with a scheduled delivery date of July 20, and it is planned to place orders for the remainder of the decanner during June.

6. Process Development

The initial core loading of EBR-II, a 50 percent enriched uranium-fissium alloy, will be reprocessed by melt refining. A supplementary reclamation (dragout) process involving a liquid metal system will be used to recover uranium and plutonium from the melt refining skulls so that the proper concentrations of noble fission products may be maintained in the recycled fuel. A second liquid metal process will be used to concentrate the plutonium in the reactor blanket so that it can be used for replenishing the reactivity of the core alloy.

Studies are being made toward the development of pyrometallurgical process flowsheets. These investigations include the determination of chemical and physical properties of substances of process interest, the development of engineering operations into workable plant-scale techniques, and demonstrations of the processes under levels of radioactivity anticipated in plant usage.

a. Melt Refining Process Technology - In preparation for the fourth melt refining experiment with highly irradiated fuel, ten percent enriched uranium-fissium pins irradiated to an estimated burnup of one total atom percent were discharged from MTR during the latter part of the month. The melt refining experiment is tentatively scheduled for the middle of June.

A brief study was made to determine the quantity and composition of the gas evolved from a zirconia melt refining crucible under various outgassing conditions. Only about 2.5 cc of gas (S.T.P.) was evolved from 100 grams of crucible material as the temperature was raised to 1100°C in an evacuated system. The identification of the gas samples collected at various temperatures has not yet been completed. Determinations of the surface area of the crucible material indicated a nonporous structure. This suggests that fine pores or microstructures are being effectively sealed by the sintering process during manufacture.

Blank runs (no uranium present) that simulated a melt refining operation were carried out in order to determine the degree of purity that could be maintained in the argon atmosphere of the melt refining furnace during the course of the run. Crucible temperatures were maintained at 1400°C for three hours in a high vacuum furnace. Samples of the argon atmosphere in the furnace were found by mass spectrometer analysis to have a nitrogen concentration of 0.10 percent and an oxygen concentration of 0.02 percent. The maximum amount of uranium consumed by complete reaction with these contaminants at the above concentration levels is negligible (about 0.01 gram atom).

b. Processing of Melt Refining Skulls - Work was continued on the development of a process for reclaiming the residual material (skull) remaining in a zirconia crucible after a melt refining operation. In the first step of the process, the skull material is freed from the crucible by controlled oxidation at 800°C which converts the skull material to a free-flowing powder. In the second step of the process, the noble fission product metals are extracted into zinc from a slurry of skull oxides in a molten salt flux. Work on this phase of the process was directed mainly toward the development of methods for the separation of the flux and metal phases. Thus far, the easiest and most reliable procedure has been the pressure siphoning of the zinc phase after freezing the salt phase. The pressure siphon tube has been kept clear during the freezing of the salt by passing a slow stream of argon through the tube. On a one-kilogram scale, zinc transfers have been consistently about 90 percent. A larger zinc charge would yield a higher percentage of transfer, since the zinc heel left in the crucible after the transfer would weigh the same as the heels obtained in transfers of smaller charges.

The third step of the skull reclamation process consists of reducing uranium oxide by means of a magnesium-zinc solution in the presence of a molten salt flux. In work concerning this step, it has been found that graphite crucibles appear to be unsuitable for handling solutions of uranium in zinc containing high concentrations of magnesium. It is suspected that the activity of uranium is increased by high magnesium concentrations, and this, in turn, results in the precipitation of uranium carbide. This problem can be circumvented by the use of metal containers fabricated from materials such as tantalum, tungsten, molybdenum, or alloys of these elements.

It has been found that interactions of uranium and ruthenium in zinc containing high concentrations of magnesium are the underlying cause of the unsatisfactory ruthenium removals that have been obtained in the past. This problem has been overcome by the inclusion of the noble metal extraction step in the skull reclamation process, since ruthenium is effectively removed in the extraction step.

After the uranium oxides have been reduced, two successive precipitations of uranium are carried out to provide additional separation of the uranium from the fission products. These precipitations are followed by a final retorting step to remove residual metal solvents from the uranium. Some difficulty is being encountered in finding a suitable material for this final step. The material that is being sought must permit the ready removal of the uranium product. Plans are being made to test beryllia for this purpose.

c. Plutonium Recovery Processes - Additional exploratory studies were made on possible methods of separating uranium, plutonium, and fission products from the second EBR-II core fuel. The separation of cerium and plutonium by means of selective reduction with a magnesium-magnesium chloride system is being studied. It has been found that about 80 percent of the plutonium oxide is reduced by the magnesium at 800°C in about two and one-half hours. The fate of cerium in this experiment is not yet known, but other experiments with cerium alone indicate that the cerium will be extracted into the flux phase. There is, therefore, a good possibility that the plutonium-cerium separation can be made by this procedure.

d. Fused Salt Studies - Experiments were continued on the chlorination of uranium oxides to render them soluble in fused salts. It was found that the uranium oxide in an oxidized melt refining skull was not affected by treatment with a carbon monoxide-chlorine mixture at 800°C in a lithium chloride-magnesium chloride-magnesium fluoride flux. In a sodium chloride-potassium chloride flux, however, the uranium oxide was converted to soluble uranyl chloride in 30 to 60 minutes.

e. Materials and Equipment Evaluation - Tests of the corrosion resistance of type 304 stainless steel in cadmium-magnesium-zinc systems at 750°C have shown that little or no corrosion occurs at zinc levels below 15 atom percent zinc. Varying the cadmium-magnesium ratios did not affect these results. Above a zinc concentration of 15 atom percent, the rate of corrosion increases sharply as the zinc concentration increases. In this same system, at 850°C, tantalum showed good corrosion resistance up to zinc concentrations of about 75 percent. At this concentration slight attack was noted. At a 98 weight percent zinc concentration, the attack on tantalum was severe. In a 95 percent zinc-5 percent magnesium system, sintered molybdenum was severely attacked. Tungsten was not attacked under similar conditions.

f. Liquid Metal Distillation - The pilot plant-scale (100 kg/hr distillation rate, 750 kg feed capacity) cadmium distillation unit was placed in operation during the past month. In general, the initial run was satisfactory and indicated that the design distillation rate can be easily met and that good control of the distillation can be maintained. The operation of the equipment will provide design data pertinent to fuel reprocessing equipment. The distillation unit is shown in Figure 5.

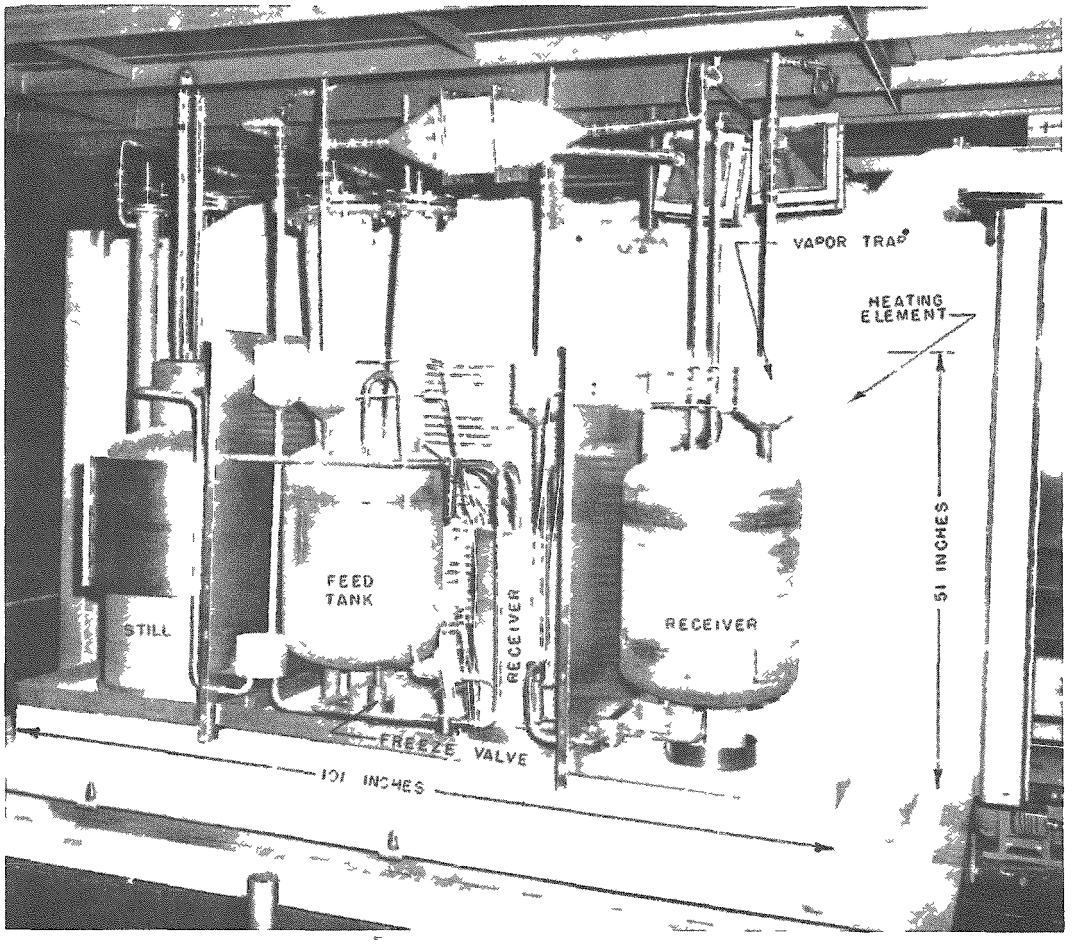


Figure 5

Cadmium Distillation Unit. (Front and side panels of furnace removed; condenser located behind rear panel).

7. Fuel Development and Fabrication - Core I

a. Fuel Prototype Irradiation - The two prototype elements, which were unloaded from the CP-5 reactor when they began showing erratic temperature behavior (see April Progress Report) were removed from

their respective capsules for examination. The damage to each element was very localized and almost identical in nature. The damage in both instances was limited to the top of the uranium-containing region. One suffered a penetration approximately 2.5 cm long while the other was breached over a length of 6 cm.

The recurrence of this type of damage in these and many other cases, coupled with the observation that the erratic behavior seems to coincide with a reactor shutdown, leads to the following hypothesis. When the reactor is scrammed and the fuel ceases to generate power the sodium surrounding the outer periphery of the cladding freezes. The resultant shrinkage of the sodium produces a void which may extend downward for some distance, eventually presenting a path of resistance to heat flow at the top of the uranium region. When the reactor is restored and brought rapidly to power, the fuel may reach prohibitively high temperatures before the entire bond becomes re-established. Under these conditions, the uranium can melt and penetrate the cladding, and may even cause local boiling upon contact with the sodium, further aggravating the situation.

The possible solutions to the problem are: (a) to raise the reactor power slowly; (b) to provide a sufficient head of sodium above the bond region; or (c) to use a liquid metal which does not freeze at room temperatures. The last solution has been chosen for the next capsule, in which NaK will be used as the bonding agent.

8. Core II Fuel Development

a. Fast Reactor Fuel Jacket Development - Arc melted buttons of seven ternary and quaternary niobium base alloys have been prepared to screen fabrication procedures for the candidate cladding materials. The buttons are in process of being jacketed for hot rolling.

Fourteen samples of various vanadium base alloys and one nickel base alloy (René 41) were prepared for sodium compatibility tests.

Commercial tantalum tubing measuring 0.188 in. O.D. x 0.162 in. I.D. was reduced to 0.175 in. O.D. x 0.156 in. I.D. by cold drawing. A total of 61 in. of tubing was fabricated. This material was satisfactory for jacketing irradiation test specimens. An additional 10 feet of the 0.188 in. O.D. tubing will be drawn to size when longer 0.156 in. diameter mandrels are received. A total of 56 in. of tantalum rod for end plug material was fabricated by swaging 0.188 in. diameter rod to 0.175 in diameter.

Several short lengths, a total of 33 in. of Ta-0.1 w/o W alloy tubing were fabricated to 0.175 in. O.D. x 0.156 in. I.D. using the same procedure reported in the December, 1960 Progress Report (ANL-6295). All material was leak checked with a helium mass spectrometer and no defects were found. A 0.450 in. diameter Ta-0.1 w/o W alloy bar was cold rolled and swaged to 0.175 in. diameter x $30\frac{1}{2}$ in. long for end plug material.

III. STUDIES AND EVALUATIONS (040116)

A. Improved Fast Reactors for Central Station Power

A study has been started to identify thos particular areas which offer the greatest possibility for cost improvement, and to attach some magnitude to the value of the improvements. These results will serve as a guide for new fast reactor designs for cheaper central station Power. It appears that current design overemphasizes the value of breeding based on prices established under present ground rules, and perhaps under-emphasizes the potential capabilities of liquid metals coolants, with resultant cost penalities.

The large number of interrelated variables involved makes it impossible to attach accurate numerical values to benefits from specific performance and design changes, especially before new and complete experimental data become available. In particular, future fuel cycle developments will be a major factor in establishing design and cost performance. However, operational analysis techniques under consideration may help to establish design objectives and performance ceilings.

B. 50-Mwe Prototype Organic Power Reactor (POPR) Evaluation

The objectives of the Prototype Organic Power Reactor (POPR) evaluation were to check and review the conceptual design of the POPR and to determine the areas requiring improvement. Also, suggestions for improving the POPR design were desired.

The conceptual design of the 50-Mwe POPR appears to be satisfactory. No basic inadequacies or difficulties were found. In one respect, the POPR is not a prototype for a large (300 Mwe) power producing organic-cooled reactor. The POPR uses downflow with no coolant phase change while the trend is toward upflow with a phase change (local boiling) for high performance.

In view of the short time available for this evaluation, only spot checks of the nuclear constants were made. Our estimates of the thermal utilization, resonance escape probability, fast fission factor, and control rod worth are in agreement with the Atomics International values. No check calculations have been made of the burnup results.

IV. REACTOR SAFETY (040117)

A. Thermal Reactor Safety Studies

1. Fuel-Coolant Chemical Reactions

Knowledge of the nature and extent of chemical reactions with nuclear reactor core metals that may occur in pressurized water or steam is essential to safe operation of reactors. The principal laboratory procedure uses a condenser discharge to provide almost instantaneous heating and melting of metal wire in water or steam. The energy input to the wire indicates reaction temperature; the transient pressure measures reaction rate; light emission indicates time-temperature behavior; hydrogen generated gives extent of reaction; and particle size of the residue indicates the surface area exposed to reaction. A second method consists of heating the metal inductively and then subjecting it to a steam pulse to induce a metal-steam reaction.

The reaction of stainless steel with uranium oxides is being studied by use of differential thermal analysis.

Studies of the kinetics of metal-water reactions under reactor incident conditions are being made in the TREAT reactor.

A series of condenser discharge runs with uranium wires in heated water was begun. Preliminary results indicated that more extensive reaction occurred in heated water than in room temperature water. These results are similar to those reported with zirconium in the April Progress Report.

Studies were begun of the uranium-steam reaction by the pressure-pulse method. Runs were made over a temperature range from 1200° to 1600°C. Preliminary results indicated that uranium is much more reactive than aluminum under similar conditions.

Further results have been obtained by the levitation melting method. Aluminum spheres, one-quarter inch in diameter, were heated to between 1800° and 2000°C while suspended in air. Ignition occurred in air at these temperatures. Vigorous burning continued even when the metal was dropped into water. Burning resulted in complete consumption of the metal.

Preparations have been made for another series of in-pile tests in the TREAT reactor. Four experimental assemblies with UO_2 core, stainless steel clad, fuel pins with varying annular gaps between core and cladding (2, 3, and 4 mils) and one containing a U-Zr-Nb alloy pin, with gold foil and tungsten wire flux monitors, have been assembled and shipped to the TREAT Reactor in Idaho. Two transients have been completed. The detailed results are not yet available.

2. Kinetics of Oxidation and Ignition of Reactor Materials

Studies are being made of the oxidation and ignition kinetics of the metals uranium, zirconium, and plutonium in order to provide information leading to an understanding of the reactions. This knowledge should make it possible to minimize the hazards associated with handling these nuclear reactor materials. Isothermal oxidation on microscope stage, shielded ignition, burning curves, rate of propagation of burning foil, and burning temperatures are the techniques being used. In the continuing study of ignition and burning of uranium, zirconium, and plutonium, more emphasis is being placed on the burning process. Burning propagation rate studies provide a useful tool to observe the effects of many variables. The effect of the presence of halogenated hydrocarbons on the burning of uranium foil in air is being investigated.

Construction of apparatus for studying the ignition of zirconium powders by the shielded ignition method was completed and shakedown tests have begun. Spherical zirconium and Zircaloy-2 powders have been received.

The effect of halogenated hydrocarbons on the burning propagation velocity and maximum burning temperature of uranium foil (0.13 x 3.0 mm) burning in air has been examined using a photoelectric pyrometer The addition of four percent $CHCl_3$, CCl_4, or CF_3Cl, or CF_2Cl_2 lowered the burning velocity over 30 percent from 0.52 cm/sec and lowered the burning temperature by 200° to 300°C from the burning temperature of 1335°C in pure air.

B. Fast Reactor Safety Studies

1. Core Meltdown Studies - TREAT Program

In-pile meltdown experiments are being performed in the TREAT reactor in order to survey types of fast reactor fuel element failure and the associated movement of fuel element materials, as well as to determine the mechanisms producing such phenomena.

Results of the transient meltdown experiments have been reviewed and compared with simple cladding failure calculations. The experimental mechanisms of sample failure appear to be in reasonable agreement with estimates of the roles played by possible failure mechanisms.

a. EBR-II, Mark-I Failure Calculations - Failure of cladding on an EBR-II Mark I specimen might be assumed to be due to one or more of the following effects: bursting from radial thermal expansion of the fuel, bursting from internal gas pressure, and dissolution of cladding by the fuel alloy.

Estimates of the relative thermal expansion of the fuel and the cladding for typical sample temperature distributions rule out failure from radial expansion. Bursting from internal gas pressure can also be

eliminated for most temperatures of interest. At 1000°C, for example, the estimated cladding bursting pressure is about six times the total internal pressure calculated for an isothermal sample. However, at 1230°C the internal pressure is equal to the bursting pressure. Hence, internal pressure could become significant in conjunction with cladding flaws and/or appreciable cladding dissolution.

Time of penetration studies (see April Progress Report) on attack of steel and iron by molten uranium or uranium-5 w/o fissium alloy indicate low Type 304 stainless steel penetration rates by uranium-fissium for temperatures near 1100°C. No data are available on the effects of contact pressure on the attack of steel by uranium at the temperatures of interest here. However, measurements at 750°C by Nuclear Metals, Inc.* (under an ANL subcontract) showed rate increases of a factor of two by increasing contact pressure from 0.6 atm to 1.05 atm. A further increase of about 70% was obtained by increasing the 1.05 atm pressure to 2.1 atm. Internal pressure at 1000°C is estimated to be about 9 atm absolute. Hence, dissolution of cladding unprotected in localized areas by the oxide film on the fuel pin, possibly assisted by internal pressure, is the most likely theoretical cause of cladding failure for temperatures of about 1100°C.

b. EBR-II, Mark-I Pins Clad with Refractory Metal - The same three mechanisms considered for the steel clad EBR-II fuel pins are of interest in the case of refractory metal clad pins.

Again, radial thermal expansion may be eliminated. For niobium or tantalum cladding, calculations indicate bursting due to internal pressure at sample temperatures of 1500°C. Data are available on the time for molten uranium to penetrate 0.054 cm thick cans of niobium, tantalum, and zirconium. If the penetration mechanism is diffusion controlled, the thickness of metal dissolved, t, by the uranium in a time, τ, may be described by the equation:

$$t(\tau) = k\sqrt{\tau} \ ,$$

where k is a constant determined by materials, geometry, and temperature.

Using the equation to correct for the different cladding thickness, and extrapolating the penetration data to shorter times than reported, it is possible to estimate the time at maximum temperature required for complete cladding dissolution. Times for complete penetration in the range 1300-1400°C are typically in the hundreds of seconds. Hence, failures at temperatures of about 1400°C should be typical of bursting due to internal pressure.

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*R. D. Jenkins, Progress Reports to ANL, NMI-4815 and NM-4816 (1960)

at about 1500°C penetration times are of the order of a few seconds, and cladding dissolution may be significant, but the available data are not sufficient to indicate clearly how important it is because of the necessity to extrapolate data well out of the experimental range.

c. Fermi-I Samples - For the metallurgically bonded zirconium-clad Fermi-I samples, cladding dissolution by the molten fuel core is the mechanism of interest. Using extrapolations of the zirconium penetration data and the time-thickness dissolved relation, it can be estimated that cladding dissolution becomes significant in short temperature excursions like those of the TREAT experiments for sample temperatures ~1400°C. It is estimated that 40 sec is required to penetrate the cladding for a sample held at 1294°C, but that this time is decreased to 4 sec for a sample at 1370°C.

d. Oxide Sample Experiments: TREAT Program - Preparations have begun for the first meltdown experiments on uranium oxide samples. The oxide cylinders to be used are about 80% of theoretical density and are 10.9% enriched and 0.381 cm in diameter. They will be clad with stainless steel EBR-II tubes for the first oxide meltdown series. Higher density samples ranging up to 96% of theoretical have been produced. Meltdown experimentation planning calls for tests on both relatively high density and relatively low density in order to check density effects on such phenomena as fuel "slumping," sintering, etc.

The first test planned is a preliminary experiment with a miniature sample to check possible gas evolution during heating and meltdown. Following it there will be four experiments on samples of the same length as EBR-II elements in which each sample is scheduled for a higher energy input than the previous one. These will cover the approximate temperature range of 1200°C to 2750°C. Samples will be instrumented with fast response Pt-Pt 10% rhodium thermocouples welded to the cladding, and will be contained inside the standard opaque graphite-lined stainless steel meltdown capsule.

e. EBR-II Elements in Stagnant Sodium - Experiments are being performed on meltdown characteristics of samples contained in stagnant sodium in order to survey the effects on element failure of changes in sample heat transfer caused by the sodium, and of the influence of sodium on meltdown product movement. A series of five meltdown experiments on EBR-II, Mark I elements is being prepared. One sample will be instrumented with an internal tantalum-sheathed, tantalum-molybdenum thermocouple of the type tested previously in a dry environment (see March Progress Report). The remaining four samples will be run under conditions set to investigate the possibility that gross sodium movement may occur during the transient heating of samples exposed previously.

2. Sodium Vapor Pressure Furnace

The lack of experimental data led to the design of an experiment to measure the vapor pressure of sodium above 1406°K. A special furnace was constructed and operated to determine its temperature capability. It successfully reached a temperature of approximately 1760°C. The furnace is now being modified to permit vapor pressure measurements to be made.

V. NUCLEAR TECHNOLOGY AND GENERAL SUPPORT (040400)

A. Applied Nuclear and Reactor Physics

1. 3.0-Mev Van de Graaff

a. Elastic and Inelastic Fast Neutron Scattering - The electromagnetic fast pulsing system, whose design and installation has been described in various progress reports, has been utilized in a most productive manner. The angular distributions of both inelastically and elastically scattered neutrons from iron, niobium, zirconium, carbon, and thorium were examined in detail at incident energies ranging from 550 kev to 1.6 Mev. The experimental results are now being analyzed and corrected for small effects due to multiple scattering within the samples. The measurements are of interest to the fast reactor program and help fulfill long standing needs of the reactor physicist for precise microscopic scattering cross sections as set forth in the publication, "Current Outstanding Reactor Physics Problems," TID-8210, p. 24.

From the point of view of the experimental physicist actually carrying out the microscopic measurements, the electromagnetic pulsing system is a most effective tool. The device produces extremely intense millimicrosecond neutron bursts that enable optimum use to be made of the fast pulse technique.

b. $\overline{\nu}$ Measurements - Measurements of $\overline{\nu}(U^{235})$ at 0.21, 0.61, and 1.58 Mev and of $\overline{\nu}(U^{238})$ at 1.58 Mev incident neutron energy were completed. The results for U^{235} are shown in Figure 6 where $\overline{\nu}$ thermal (U^{235}) = 2.43 was taken as a standard reference point.

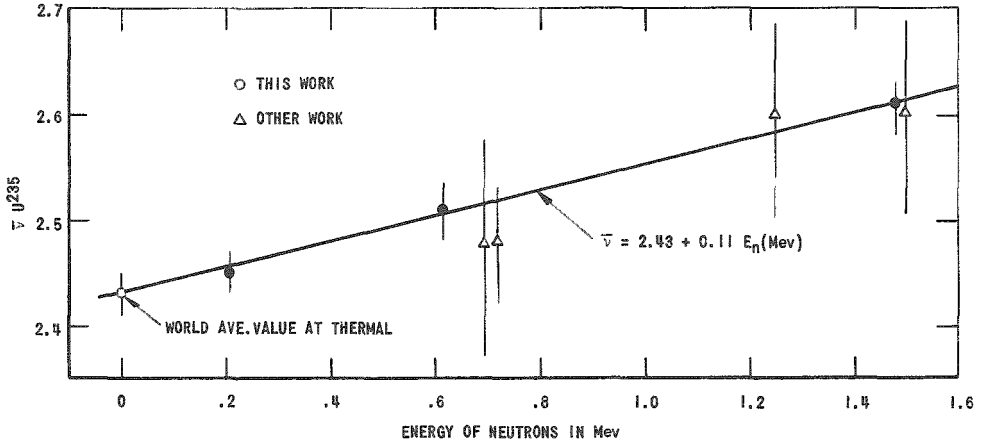

Figure 6

$\overline{\nu}_{U^{235}}$ as a Function of Energy

The value of $\overline{\nu}(U^{238})$ at 1.58 Mev, also referenced to $\overline{\nu}$ thermal (U^{235}) = 2.43, was measured to be 2.605 ± 0.04 in. in good agreement with previous results. The figure yields, by graphical analysis, a slope $d\overline{\nu}/dE(U^{235})$ = 0.11 Mev^{-1}.

This value is lower than the value of about 0.139 obtained by objectively fitting previous measurements. It is concluded that the previously accepted postulate of a linear dependence of $\overline{\nu}$ on incident energy is not correct and that $\overline{\nu}(U^{235})$ depends on the incident neutron energy in a more complex manner. This conclusion has been suspected for some time by theoretical reactor physicists as a result of their analyses of fast reactive assemblies, but the previous microscopic measurements were not of sufficient accuracy to justify other than a linear assumption.

The measurements are continuing in order to establish $\overline{\nu}(U^{235})$ and $\overline{\nu}(U^{238})$ with high precision at other incident neutron energies. In addition, $\overline{\nu}$ of other fissile materials will be examined, Th^{232} in particular.

c. Fast Neutron Capture Cross Sections - The relative activation cross sections of the Rh^{104} ground state and metastable state were determined as a function of incident neutron energy. The results are shown in Figure 7.

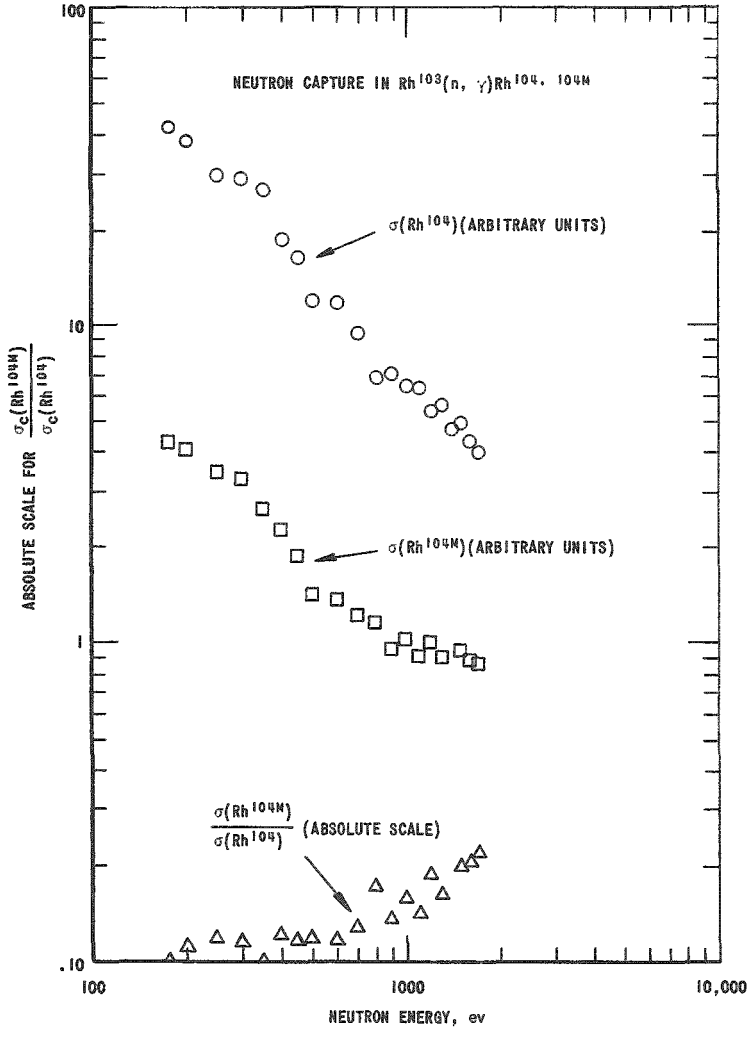

Figure 7

Relative Activation Cross Sections of the Rh^{104} and Rh^{104m} vs Neutron Energy

The technique employed utilized short neutron irradiations and multichannel time analysis to determine the two components. The results are

a portion of our general program of fast capture cross section measurements, the ultimate objective of which is a full understanding of this important type of fast neutron reaction. In isotopes of the Rh^{104} type, where capture to a metastable state is involved, the experimental results are of particular interest from the theoretical standpoint.

d. Performance - Several months ago extensive Van de Graaff modifications involving vacuum and gas handling systems were briefly outlined. Such work is tedious and necessary but not always immediately productive. However, this effort is now producing a beam six times the the manufacturer's specifications and 10% over the design energy specification. Such performance from the basic accelerator is an immense aid in carrying out the physical measurements.

2. Argonne Thermal Source Reactor - ATSR

a. Modifications - The design of the Argonne Thermal Source Reactor system was derived from experience with critical assemblies constructed and operated in the early 1950's. Hence, the underlying philosophy of the control system and instrumentation was heavily influenced by these earlier critical facilities. The change in designation of this facility from ZPR-IV' to ATSR reflects the growing use of this system for experiments requiring a high neutron flux, and recently the authorized power level was raised to 10 kw. Consequently, a systematic review of the design of this system has been performed to determine what changes are desirable in the light of the new role of this facility.

Steps taken to increase the effectiveness of the shielding have been reported previously. Currently, a renovation of the instrumentation and control system is underway. The current-voltage-flux characteristics of each of the detecting chambers have been determined, and replacement of several of these devices by instruments useful over a wider flux range is planned. A corresponding extension in the useful range of the associated circuitry is also a part of the modifications. The result of these changes will be an increase in the overlap of the ranges of the various instrument channels used for following the flux level from that associated with the source to operation at 10 kw. An area monitoring system will be installed as a supplement to the periodic surveys of the radiation level in the vicinity of the system as performed by radiation safety personnel. The gravity-dependent steel tape drives used on the control rods will be replaced by rack and pinion drive arrangements.

The two safety rods on the ATSR facility are inspected periodically as a part of the routine preventive maintenance procedures. In the most recent inspection a number of defects were noted. The cadmium sheets held between stainless steel plates by aluminum rivets had expanded laterally beyond the edges of the steel plates by as much as $\frac{1}{4}$ in., probably due to

cold flow of the cadmium under the pressure of the riveted jackets. A thick layer of hydrated cadmium oxide had formed on the cadmium surface. Excessive warping and buckling of the cadmium had occurred over the top quarter of each rod.

This information is reported with the thought that it may be of use to others in the design of similar control rods. On the ZPR-VII facility the cadmium is waterproofed by heliarc welding the edges of the aluminum plates which provide structural support for the cadmium sheets. Venting was not considered necessary, since high temperature operation does not occur, but provision for venting has been incorporated in control rods used in other reactor systems. It is possible that a breathing effect introduced by temperature changes would allow water to make contact with cadmium in these other rod designs, thereby resulting in the same type of corrosion observed in the ATSR safety rods.

An attempt is being made to provide a protective coating on cadmium by painting with a commercial product known as Heat Rem H-170. A strip of aluminum was painted with this material, air-cured for three hours at 450°F and then held at 1050° for four hours. No damage to the paint surface was apparent. Similar tests were made with cadmium samples which had been rubbed with steel wool to remove the oxide coating. Immersion in boiling tap water for 96 hours produced no signs of oxide formation or scaling of the aluminum paint for several samples, but two did exhibit flaking. These tests will be repeated with cadmium samples which have been acid-etched to remove completely the oxide layer before painting.

b. Preparations for Resonance Integral Measurements - In the Progress Report for February, 1961 (ANL-6328), a difference of a factor of two in the neutron flux per logarithmic energy increment in ATSR was reported, on the basis of activation of detector materials having a single dominant resonance. Self-protection effects were indicated, and plans for repeating this determination with thinner foils were mentioned. Some of these measurements have now been performed with thinner foils and a different technique. The procedure used initially involved a radiochemical method for the measurement of the activation of detectors placed inside a cadmium-lined tube at the center position of ATSR. The current procedure involves conventional counting of bare and cadmium-covered foils.

Thin foils of indium, gold, and manganese have been irradiated in the ATSR and the cadmium ratios determined. From these data and the "known" resonance integrals and thermal cross sections, the resonance flux per log energy decrement has been determined. The results are as follows:

Detector	Cadmium Ratio	Resonance Integral	Thermal Cross-Section	Resonance Flux
Gold-197	2.80	1558 b	98.8 b	5.07×10^8
Indium-115	2.85	3500	191	4.33×10^8
Manganese-55	23.3	7.8	13.2	10.9×10^8

It is readily seen that the indium and gold determinations agree fairly well but that the manganese value for the flux is greater by a factor of about 2. The cadmium ratio has been determined with extremely thin foils and the value used was obtained by extrapolating the cadmium ratio vs foil thickness curves to zero thickness so that self protection in the foils should be taken into account. Since the resonance integrals of gold and indium are known fairly well, it would seem that there are two possible contributions to the differences in measured flux. The first is that the manganese resonance integral is in error by a substantial amount and the second is that the flux in ATSR deviates from 1/E behavior in the region of the principal resonance peak of Mn^{55}, about 260 ev. This study is to continue. Cobalt and molybdenum foils will be irradiated and a further comparison in the higher energy resonance region will then be possible.

Performance testing of the autorod system on ATSR was reported in the April Progress Report, and it now appears to be working satisfactorily. It is planned to make a power measurement in the near future utilizing pile noise observations with the autorod in order to check the system further. Two less absorbing autorods have been constructed and both have operated satisfactorily. One rod is made from $\frac{1}{8}$ in. thick 2% borated steel and has a total worth of approximately 40 inhours. Its total control movement is about two inhours. The second rod is $\frac{1}{8}$ in. thick Type 304 stainless steel and has a total control movement of about 0.7 inhours. With this stainless steel rod, reactivity changes with errors of the order of 10^{-8} $\Delta k/k$ have been measured.

3. ZPR-VII - High Conversion Critical Experiments

Since AEC hazards approval still has not been granted for operation of the Hi-C experiment, emphasis remains on improvement and rehearsal of measurement techniques to be used in the experiment. Although neutron spectral conditions to be encountered in the Hi-C cores are not duplicated elsewhere, foil exposures were made in ARGONAUT, ATSR, and CP-5 to gain experience in measuring activities and yields of fission products and Np^{239}. These measurements are necessary in the determination of the conversion ratio, resonance capture, fast effect, and resonance escape probability.

Measurement of the effective neutron temperature by the use of lutetium detectors is planned. The technique will be similar to the procedure

developed at Hanford;* however, modifications are necessary to account for the large epithermal flux in the Hi-C cores. The formulae have been refined and corrections terms have been calculated using the LGP-30 computer.

A cadmium-lined stainless steel startup source holder was installed, replacing the aluminum holder. The new fixture has greater strength, a higher melting point, and provides about 90% shielding of the contained plutonium (32 gm) from leakage flux from the reactor. The change affords source containment up to the point where core meltdown might be anticipated during a startup accident. The source is normally withdrawn from the reactor into a shielded pit after startup.

The cadmium lining was electroplated onto the source holder, then bored to size, leaving a minimum wall thickness of 0.025 cm (0.010 in.). During the plating operation it was found that the cadmium shells did not bond to the stainless steel unless unusually low deposition rates were used. It appears possible to produce thin, intricately shaped cadmium shells by this technique since the cadmium may be stripped free. The use of carbon steel resulted in strongly bonded plated surfaces for the same conditions which produced strippable coats on stainless steel.

4. JUGGERNAUT

The JUGGERNAUT is a water-cooled and graphite-moderated reactor of the ARGONAUT type. It is to operate at power levels up to 250 kw and provide a versatile facility for nuclear research and component development. Construction of the facility is complete and preliminary tests are in progress. The Hazards Report is being reviewed by the Commission, and the Operating Manual is in press.

a. Engineering Tests - The primary water flow rate was found to be below 125 gpm. A little reworking of the throttle valve increased the maximum flow rate to 135 gpm.

The control rods were adjusted to reduce the fall time which now is running between 0.35 sec to 0.40 sec for all rods. Some rework and adjustments are being made on the console. The cooling tower is being cleaned of debris and fine screens are being provided to reduce the accumulation of dirt.

The resin was changed in the cleanup system. The water is being maintained at 1.5 megohm and a pH around 7. A collimator was placed in the northeast beam hole. The center rotating plug was raised and the inside of the vessel checked for cleanliness. Some dirt was present; it was cleaned up and the plug replaced.

*W. P. Stinson and L. C. Schmid, Lutetium as a Spectra Index Detector, HW-66319 (1960).

b. Critical Experiment Preparations - Control rod calibration procedures have been revised to include calibration using a rod-insertion method. The power trace as a function of time (obtained using automatic counting and timing circuits) will be used in conjunction with the code RE-138 on the IBM-704 to give reactivity versus rod position. Using a perturbation method, it has been calculated that the effective mean distance of each rod from the outer periphery of the core is slightly greater than that (4 cm) assumed in previous rod worth calculations. The resulting decrease in the calculated rod worth is less than 10%.

5. Theoretical Reactor Physics

a. Resonance Integral and Neutron Age Compilation - The first ANL newsletter on "Resonance Integrals" and "Age of Fission Neutrons" is now undergoing review.

b. Cross Section Collapsing Code - A code has been written using FORTRAN for collapsing a given many-group cross-section set (using a given many-group spectrum) to an equivalent few-group cross-section set. A typical problem involving the collapsing of 13 groups to 4 groups and for two materials requires less than 10 sec of IBM-704 machine time after the program and data are read in.

The code is designated as AMD No. 1172 and will accept up to 20 groups and 20 materials. Down scattering is allowed to all lower groups both with the input and the output cross sections.

B. Reactor Fuels Development

1. Corrosion Studies

a. Corrosion of Sintered Aluminum Powder Product Tubing - Samples of impact-extruded Alcoa powder metallurgy tubing made from "modified" powder are still being corrosion tested. Except for the microblisters reported last month the corrosion appears normal for resistant alloys.

Armour Research Foundation powder tubing extruded through a bridge die has been exposed to water for 65 days at 290°C and 71 days at 360°C. Most of the surface of the tubing looks very satisfactory but increased attack continues at the junctions where the metal streams joined during extrusion.

The eutectic bonded (A.I.) plus "motor arc" welded tube end closures show no sign of penetration after 17 days at 290°C.

b. Lightweight Alloy for Liquid Mercury - The survey of the corrosion resistance of commercially available titanium alloys (see March Progress Report) has been continued in mercury at 370°C. Thus far, encouraging alloys have not been found for use with mercury.

Preliminary data show that anodized titanium and some alloys are immune to attack by mercury during two weeks of continuous exposure, whereas unanodized specimens are corroded considerably. The nature of the anodized films and their critical thickness for mercury resistance have not been determined. It is also not certain that these films would resist high temperature mercury for a much longer period of exposure or under dynamic test conditions.

Stress-relief for as-received alloys offers some beneficial effect in reducing the cracking which occurs during exposure to mercury.

2. Irradiation Studies

a. Examination of Irradiated ZrO_2-UO_2-CaO Pellets (EBWR Core IA-Type Fuel) - Pellets of the EBWR Core IA spike composition (81 w/o ZrO_2-10 w/o UO_2, 93% enrich.-9 w/o CaO) were irradiated for about 8 months in the MTR in an unperturbed thermal flux ranging up to 2.7×10^{14}. Nine capsules were irradiated each containing a single unclad pellet 0.219 in. in diameter and 0.6 in. long. The capsules all contained NaK in order to facilitate heat transfer. The pellets were of three different geometric densities (5.22, 4.73 and 4.50 gm/cc) corresponding to 92, 83 and 79% of theoretical. Measurements have been made of the amount of fission gas released and the data are given in Table VI. These preliminary results are based on thermal flux measurements determined by a flux monitor. Final results will be calculated on the basis of mass spectrometric analyses of burnup.

Table VI. Effects of Irradiation on ZrO_2-UO_2-CaO

Spec. No.	Geometric Density, gm/cc	Estimated Burnup, Mwd/T	Max. Heat Output, Watts	Max. Surf. Heat Flux, Btu/hr-ft^2	$\int kd\theta$, w/cm	Fission Gas Release, % Theoretical
ANL-35-76	4.73	11,300	611	560,000	25	0.83
-71	5.22	13,700	840	850,000	37	0.30
-79	4.50	14,200	842	800,000	35	1.22
-78	4.50	15,000	939	900,000	40	0.98
-75	4.73	15,600	1014	910,000	40	1.24
-72	5.22	16,100	1010	1,050,000	46	1.21
-77	4.50	16,600	1162	1,040,000	46	1.88
-73	5.22	16,600	1185	1,090,000	48	1.32
-74	4.73	17,200	1131	1,080,000	47	1.94

Five of the nine pellets were centered in their capsules with a doughnut of stainless steel wool. The remaining four pellets were held loosely in a Zircaloy holder. Using a pinhole camera technique, each

capsule was autoradiographed before being opened. The results indicated that all nine pellets were fractured to varying degrees. The stainless steel wool, however, was more successful in maintaining the pellets in their original geometry than was the Zircaloy holder. The pellets in the stainless wool were removed from their capsules and, after removal of the NaK, were mounted in a cold-setting epoxy resin for metallographic examination.

Specimen ANL-35-78 appears to have melted after fracturing had occurred, although this was not the hottest specimen nor did it release the greatest amount of gas. It was, however, the only specimen to show signs of melting. The melting point of this material is about 2500°C. A piece of the melted ceramic has been mounted for metallographic examination.

The values in Table VI for surface heat flux and $\int k d\theta$ were calculated for initial conditions and give an indication of the maximum severity of the irradiation. After the pellets fractured, the pieces separated and the original geometric conditions that were assumed for calculating these values no longer existed. Because the cylindrical geometry was destroyed, it was not possible to calculate an irradiation temperature at the center of a pellet. The heat output at any time, however, is primarily dependent only upon the U^{235} content and the thermal neutron flux, and is relatively independent of geometry.

The range of fission gas release and burnup is too limited to infer that there is a correlation between them, although one should exist. Likewise, it is not possible to draw any conclusions from the heat ratings and gas release, and density and gas release. The fact that the pellets fractured and subsequently were irradiated at reduced temperatures undoubtedly was a factor in the relatively low fission gas release. Examination of the microstructure may indicate the importance of this factor.

The average ratio of Xe/Kr for the nine specimens is 4.22. Since the ratio of fission yields for the isotopes measured is 7.19, the low value indicates that either the xenon diffusion is enhanced or the krypton diffusion is retarded in the ZrO_2-UO_2-CaO lattice.

The gas release from capsule ANL-35-71 is significantly out of line with the rest of the group. This has also been established from the argon-39 analyses of the gas released from each capsule. Argon-39 is formed from an n-p reaction with potassium-39 in the NaK and is released in direct relationship to the integrated flux to which the capsule is exposed. This fact has been used as an independent rough check on the gas collection apparatus, the sampling technique, and the flux indicated by the monitor. The argon-39 fraction is independent of the xenon and krypton release values because it does not depend on any of the factors which govern the release

of fission gas from the fuel. The volume of argon-39 released from capsule 35-71 is significantly low and therefore it is probable that the fission gas release value is in error.

3. Nondestructive Testing

a. Ultrasonic Techniques - In the February, 1961, Progress Report (ANL-6328) attenuation measurements of various Lamb wave modes was mentioned. The specimens used were 30 mil heat treated brass plates. No consistent relationship between attenuation and grain size was obtained. The experimental method of measuring the attenuation has been modified. A definite difference has now been noted indicating that there is a relationship between the ultrasonic attenuation and grain size for various Lamb wave modes. In order to get more consistent results the equipment has been modified to give better wave shapes.

The investigation of the sound propagation in small steel (SA-212B) cylinders is being continued. In order to prevent overloading of the amplifier a two-crystal technique is being used. One crystal transmits and the other receives the 15 MC ultrasound. A half microsecond pulse is being used. In order to prevent electromagnetic pickup from the transmitter, it was necessary to put the transmitter in a heavy shielded box.

b. Neutron Techniques - Image resolution studies in progress have indicated that appreciable gains in image sharpness can be obtained by the use of a single metal converter screen. This causes some decrease in speed. The thickness of single screen and the film orientation with respect to the neutron beam which would yield the best image have been determined and their relative speed measured. These speed results are given in Table VII.

Table VII. Photographic Speed and Screen Thickness for Best Image Resolution

Screen Thickness (mils) and Film Location	Photographic Speed (relative to 10-20 cadmium)
10-10 rhodium	1.4
20-30 indium	1.1
10-20 cadmium	1.0
1-2 gadolinium	0.9
18-18 silver	0.8
10 cadmium - front	0.67
1 gadolinium - front	0.63
10 rhodium - front	0.62
20 indium - back	0.5
15 silver - back	0.35
KK film - no converter	0.03

The screen thickness is given in mils in the table with the first number indicating the screen thickness toward the neutron beam. For the double screens the film was sandwiched between the screens. For the single screens, the film location designated "front" indicates that the film was on the neutron side of the screen. All results reported are for direct exposures and allow 3 half-life decay of radioactive screens in contact with the film. The speed numbers are arbitrarily based on 10-20 cadmium equal to 1.0, and are all based on several minute exposures to a thermal neutron beam intensity of 3×10^5 n/cm^2-sec. Radioactive transfer time is not considered in the speed values. An indication of over-all sensitivity can be found from the fact that a total density of 1.5 can be obtained using KK film with 10-20 cadmium screens for a total neutron exposure of about 2×10^7 n/cm^2.

The speed values cover a range of about four for the metal screens shown. On a speed basis there is relatively little to be gained by the use of one screen configuration over another. However, for over-all image quality, the use of a ten mil rhodium screen with the film located on the neutron side of the screen would be a highly recommended technique. This technique sacrifices only a factor of about 2 in speed but appears to yield the best combination of image sharpness and film density range. For neutron beams having a large neutron/gamma ratio, this would be a preferred method for obtaining high quality neutron radiographs.

Further neutron radiographic studies have been planned using the JUGGERNAUT reactor. A beam collimator, designed for use at this facility, was placed in the reactor beam tube this month. Most of the other equipment needed for these studies is being assembled.

C Reactor Materials Development

1. Pressure Vessel Steel SA-212B

The metallurgical stability of the SA-212B steel used in the fabrication of the EBWR pressure vessel was investigated. The mildly radioactive plugs, removed from the pressure vessel wall for the installation of the new steam and feedwater nozzles, were prepared into V-notch Charpy and standard (0.250 in. diameter by 1 in. gauge length) specimens (see Progress Report, January 1961, ANL-6307, p. 55). The tensile strength, impact resistance, and ductility data from these specimens were almost identical with the results obtained from control specimens prepared from a 4 in. thick reference plate used in the fabrication of the lower head itself. The shell and head plates were rolled from the same heat of aluminum-treated steel.

The 15 ft-lb NDT temperature for both groups of samples was found to be +5°C. The areas under the stress-strain diagrams were identical within the limits of experimental error. Ductility as determined by elongation and reduction of area at fracture were also identical. From these data,

it was concluded that stress at temperature had no embrittling effects on the vessel shell steel. The neutron dosage received by the steel tested is trivial inasmuch as these steel plugs were in the vessel shell well above the operating water level of the reactor.

2. Magnetic Properties

New, unirradiated SA-212B magnet bar standards were prepared, hardened and tempered for the study of the influence of tempering temperatures on magnetic properties below 700°F and above 1150°F (see Progress Report, April 1961, ANL-6355, p. 57). Comparison of 120 cps inductance bridge measurements at three different excitation voltages on fully annealed unirradiated bars, revealed that the short one-hour tempering, and the longer two and three-hour times at tempering temperature, failed to restore the magnetic properties to that of the annealed condition at all temperatures below 1250°F. At the 1350°F temper temperature, the steel started to soften magnetically again but was still harder than fully annealed material.

Preliminary 100 cps inductance bridge measurements comparing irradiated steels with a fully annealed unirradiated steel standard revealed that the irradiated steel is magnetically softer than unirradiated quenched and tempered specimens having the same mechanical hardness. Furthermore, the magnetic properties of the as-irradiated steel were unaffected by annealing for one hour at 700°F, 800°F, 900°F, and 1000°F. Additional irradiated magnet bars are in preparation.

3. Sodium Coolant Decontamination

A study was made utilizing a loop in which sodium was contaminated with radio-cesium. The loop was operated initially with the valves to the adsorption section closed. Under these conditions the activity at various reference points in the outer leg of the loop showed a uniform distribution, with no decrease due to extraneous mechanisms. Following this determination the valves to the test section were opened permitting the sodium to flow through the bed of graphite. The graphite particles were 12 to 14 mesh in size.

After 72 hours of flow through the test section with the loop maintained at approximately 260°C, the activity dropped by a factor of two. An additional run of 72 hours at approximately 426°C further decreased the activity by another factor of two. No buildup of activity was observed to have taken place in the cold trap.

It is believed that the removal mechanism is dependent upon the formation of a cesium carbide by adsorption and diffusion of the cesium into the graphite structure. Present efforts are being directed toward the

separation of the contaminated graphite from the sodium without appreciably altering the chemical structure of the graphite. When this is accomplished, X-ray diffraction techniques will be used to check for an interstitial compound.

D. Reactor Components Development

1. Development of Viewing Systems

a. The Role of Cerium in the Suppression of Radiation Induced Coloration in Glasses - It has been found that the relative concentration of Ce^{3+} to Ce^{4+} ions in glass, as well as the total cerium content, is an important factor in the suppression of radiation induced visible coloration. In a potassium-alumino-borate glass which does not contain cerium, the induced coloration in the visible is due to the overlap of two absorption bands; one centered at about 2.36 ev (525 mμ) and a band in the ultraviolet at 4.0 ev (310 mμ). It is postulated that the visible band at 2.36 ev can be attributed to positive hole centers resulting from the loss of electrons from non-bridging oxygens, and the near ultraviolet band at 4.0 ev can be attributed to electron trap centers. The role of cerium can be explained on the basis of its change in oxidation state when irradiated and its effect on both induced bands. While the Ce^{3+} ions suppress the hole centers by supplying electrons and changing to Ce^{4+}, the Ce^{4+} ions suppress the electron trap centers by attracting electrons and changing to Ce^{3+}.

The change in oxidation state of cerium in the above borate glass can be determined by changes in the ultraviolet absorption spectra. The absorption of Ce^{4+} ions in this glass has been found to be considerably stronger than the absorption due to Ce^{3+}, and is a very wide band ranging from about 400 mμ to below 200 mμ. This absorption is considered to be due to a charge transfer between Ce^{4+} ions and the surrounding oxygens. The intrinsic absorption due to Ce^{3+} consists of a series of 6 bands in the range of 350 to 200 mμ. Based on available literature on the absorption of Ce^{3+} in solutions and crystals these absorption bands are attributed to the 4f to 5d transitions of the 4f electron.

The role of cerium as described in the above postulate is based on a number of supporting observations. In the case of a glass containing mostly Ce^{4+} ions, the absorption due to Ce^{4+} is decreased by irradiation in a manner consistent with a change of some Ce^{4+} to Ce^{3+} ions. This is accompanied by the observation, on the same glass, that the induced band attributed to electron trap centers (4.0 ev) is of a much lower intensity as compared to an irradiated glass containing Ce^{3+}. In the case of a glass containing Ce^{3+} the ultraviolet absorption is increased by irradiation in a manner consistent with a change of some Ce^{3+} to Ce^{4+} ions. The band attributed to the positive hole center (2.36 ev) is of a much lower intensity

as compared to an irradiated glass containing Ce^{4+}. Hence, the necessity of having both Ce^{3+} and Ce^{4+} ions in the glass in the proper relative concentrations in order to suppress both types of centers contributing to the induced visible coloration.

E. Heat Engineering

1. High Void Natural Circulation Study

The fabrication of a new air separation tank was completed, and the tank was installed (see Progress Report, March 1961). Preliminary runs indicated that the maximum void fraction which can be obtained in the 12 in. riser was 47%. At this condition, the water surface above the riser was very violent with splashing that carried 2 to 4 ft above the surface.

Experiments have been started using only one downcomer since the flowmeter of the second downcomer is undergoing repair. A series of experiments is in progress in which orifice plates are successively added to the downcomer to measure the effect of the single-phase flow resistance.

2. Steam-Water Separation Studies

Preliminary data on steam carryunder have been obtained at 600 psi on the large scale loop. The data indicate that the inception of carryunder occurs in the neighborhood of 1 ft/sec and the weight fraction of carryunder (per cent of total steam produced) is in excess of 50% at 2.0 ft/sec (superficial downcomer velocity) in the downcomer. Additional data are to be taken at pressures of 1000 and 1500 psi. These data are in good agreement with the correlation developed from the previously reported atmospheric air-water studies.

3. Void and Velocity Distributions in Two-Phase Flow

The study on the determination of the dynamic properties of the steam void fraction in boiling water channels was terminated. The findings of the study can be summed up as follows: The gamma-ray transmission method of void measurement works satisfactorily, if the signal-to-noise ratio can be kept high or if the recording time can be chosen large enough to compensate for the effect of the low signal-to-noise ratio. The noise is due to the statistics of the gamma-ray source and the scintillation crystal. A procedure was outlined for the choice of the recording time under given conditions.

A comparison of the results obtained with the gamma-ray and velocity methods indicates that neither of the two models developed in these studies satisfactorily relates the velocity variations with the steam

void variations over the entire range of frequencies studied. For low frequencies, both models are useful and have approximately the same accuracy. For instance, if the steam transit time is 0.1 sec, the deviation from the gamma-ray results is 3 db at one cycle per second. For higher frequencies, the second model gives slightly better results than the first. The accuracy of both models increases with decreasing steam transit time.

4. Hydrodynamic Instability

The causes of hydrodynamic instability are being investigated on an instrumented natural circulation loop. Results with a test section, 91.5 cm long with an inside diameter of 1.08 cm, and a 1.6 cm inside diameter riser indicate that flow becomes unstable at low heat fluxes. To compare geometrical effects on flow stability a larger inside diameter test section is being designed.

A new series of performance tests with improved instrumentation is yielding information on conditions at the onset of instability for a 1.6 cm inside diameter test section and riser.

5. Hydrodynamic Computer Program

A computer program that will permit complete hydrodynamic calculations of almost any conceivable core geometry is being prepared for the IBM-704 computer. The equations and calculation procedures have been simplified somewhat to reduce the complexity associated with certain portions of the calculation. All fluid property functions have been fitted satisfactorily with polynomials. With the exception of the two-phase pressure drop correlation which must be reviewed, the equations are ready for programming.

6. Sodium Boiler

A source of sodium vapor is required for a number of high temperature liquid metal studies that have been proposed. To meet this need the design of a "long-lived" sodium boiler capable of operating at 3 atm was recently initiated.

The reference design of the boiler has been completed. The boiler will operate by means of natural circulation, thereby eliminating development of a circulating pump, and will provide one pound of sodium vapor per minute with a designed power input of 30 kw. This vapor flow rate could be increased at least 50%, but with a sacrifice of the heater lifetime.

7. Heat Flux Distribution in Flow

In preparation for numerical studies of the effect of heat flux distribution on Nusselt number in the heat transfer to liquid metals problem (Progress Report for April, ANL-6355, p. 59), programming on the LPG-30 was initiated. The particular solution where conduction in the direction parallel to stream flow is neglected has been programmed and the tape punched. A survey of the capabilities of the machine using the floating point interpretive system indicates that programming the problem with streamwise conduction should involve no new difficulties.

8. Packed Bed Reactor Studies

a. In-Pile Loop Experiment - Design and selection of components for a steam-cooled in-pile packed bed fuel test is in process. The anticipated location of the effluent steam desuperheater-condenser and condensate tank has been changed from below to above the shielding plug of one of the large holes in the graphite of CP-5. This allows a considerably larger condensate tank and consequently a longer operating time at a given power level. (For the experiment the expected power output of 1.5-2 kw could be sustained for about four hours of continuous operation at 750°C.) A 3.8 cm diameter stainless steel thimble penetrating a shield plug is being considered to allow relatively quick installation and removal of test specimens and instrumentation. The planned design of the condensate tank, tubing, and shielding plug is being modified to accommodate this feature. An instrumentation "stalk" of sheathed thermocouples and pressure taps has been designed for easy assembly into the packed bed assembly and the test facility.

Efforts to design a predictable desuperheater-condenser have not been successful. A number of heat exchanger geometries for which design information is available (including tube banks and concentric tubes) appear to be incompatible with either the low flow rates involved, high temperature differences or space limitations. A laminar flow design intended to permit operation of the bed at 4 kw, 750°C will require a proof test. The question of placing the heat exchanger and tank outside the permanent shielding will be reopened. This would allow the test to be placed in a smaller hole with a higher flux. Higher power density, longer operating time, and more flexible operation could result.

b. Power Supply - An investigation of induction heating of iron particles for a heat transfer study on packed beds is continuing. Induction heating specialists at two outside laboratories have indicated that the problem can be solved only by experiment, since the experimental and theoretical knowledge necessary to predict heat generation in particulate masses does not exist.

Experiments have been conducted to isolate variables to determine a method of producing the desired heating effects. Surface condition of the particles was found to be a major factor affecting the depth of heat generation in the mass and the average power generation within the mass. For electrically-insulated particles, the average power generation was low and heating was uniform over the volume of particles. For electrically contacting surfaces, the average power generation was high, but most heating occurred near the surface of the mass. Further study will be directed toward achieving the desired heating characteristics by varying surface resistivity of iron particles to obtain a heat generation intermediate between the cases for high and low surface resistivity.

F. Separations Processes

1. Fluidization and Fluoride Volatility Separations Processes

a. Direct Fluorination of Uranium Dioxide Fuel - Pilot plant studies of a direct fluorination process for the recovery of uranium dioxide fuel are being continued. Previous runs had suggested the effectiveness of higher gas rates (>0.5 ft/sec) in reducing tendencies of caking of the inert bed, and of oxygen in promoting the overall reaction rate. In the present period these effects were demonstrated in two runs at 500°C in which 4.5 kg batches of hydrogen-fired pellets were completely fluorinated. In these runs the fluorine was diluted with oxygen, (avg. conc. = 40%) and about one-half the time (6 to 8 hr for complete reaction) was required compared to similar operations with nitrogen diluent. Good fluidization and heat transfer in the inert calcium fluoride bed was shown by nearly uniform temperatures in the reaction zone. Gas recycle by means of a diaphragm pump was successfully demonstrated in these runs. This technique has the advantage of fluorine economy.

b. Processing Stainless Steel-Clad Uranium Dioxide Fuel Elements - A two-zone fluid-bed reactor is being used to study chlorination and fluorination reactions for the decladding and recovery of uranium from stainless steel-clad uranium dioxide fuel elements. Potential materials of construction for the fluid-bed decladding step are being evaluated in detailed corrosion studies initiated this past month. Nickel, Inconel and Hastelloy-C coupons were exposed to once-through chlorine at 600°C in a one-inch horizontal tube furnace for periods up to 200 hours. Weight loss data showed substantially lower corrosion rates (less than five mils/month) for these relatively long-term tests compared to the short-term results reported previously (about 20 mils/month in 20-hr tests). Preliminary inspections of Inconel and Hastelloy for intergranular corrosion were inconclusive. However, nickel corroded severely by this mechanism and must be considered unsuitable for the decladding step.

c. Evaluation Tests of Decladding of Zircaloy-Clad Uranium Dioxide Fuel - Scouting runs were made in a two-inch reactor to evaluate alternative schemes for removing Zircaloy cladding from uranium dioxide fuel. Because of the great simplicity that would result if no separate reagents were required for decladding, two tests were made to determine the effectiveness of direct fluorination as a means of removing cladding and of fluorinating the fuel material through holes punched in the simulated fuel elements. In a run made in a fluid bed at temperatures up to 500°C a 1 mil/hr attack on the alloy was observed and about 10 per cent of the uranium was fluorinated in 4.5 hours. In another run without an inert bed penetration rates of up to 6.5 mils/hr were obtained, but little uranium was fluorinated out of the tubes. Large temperature excursions were encountered in this run. In general, the direct fluorination approach does not appear attractive at this point. Decladding with hydrogen chloride in a two-stage operation is currently favored.

d. Separation of Uranium from Zirconium-Clad and High Zirconium Alloy Fuels - Further laboratory work has been done to make the Direct Fluorination Volatility Process applicable to the processing of enriched uranium-zirconium alloy fuel. The proposed method includes decladding and removal of the zirconium from the fuel by reaction with hydrogen chloride at or above 350°C. Zirconium tetrachloride is volatilized from the reaction vessel. The uranium remains behind in the form of nonvolatile uranium chlorides. The reaction of hydrogen chloride with zirconium and the subsequent fluorination of the uranium to uranium hexafluoride can be carried out in a fluidized bed of alundum (refractory aluminum oxide). Results indicate that best removal of uranium from bed materials occurred when alundum was used. Poorer results were obtained when zirconium tetrachloride was included in the bed materials. When alundum was used as a bed material for the hydrogen chloride decladding step, and the uranium was then fluorinated in this same bed, 0.006 per cent (>99 per cent uranium removal) uranium remained in the alundum residue. The reaction with fluorine was carried out in a one-inch fluidized-bed reactor at 350°C. Similar results were obtained in static-bed fluorination experiments.

Nickel has been suggested as a container material for the fluorination step. The kinetics of the nickel-fluorine reaction will be determined over the temperature range 100° to 800°C. The knowledge of the rates of reaction will be helpful in obtaining a more complete understanding of the mechanism.

e. Plutonium Fluoride Studies - Laboratory-scale fluorinations of solid solutions of uranium and plutonium dioxide, mixed with solids being considered for use as inert material in a fluidized-bed fluorinator, are being carried out. Upon reacting fluorine with a solid solution of mixed oxides of plutonium and uranium good removal of plutonium was achieved

at 250° and 450°C in the absence of an inert solid. From a starting mixture containing 3.76 per cent plutonium, 98.0 and 99.9 per cent of the plutonium had been volatilized at 250° and 450°C, respectively after ten hours of fluorination. Using calcium fluoride as inert solid, inadequate removal of plutonium resulted when the reactions with fluorine were made at 250°, 450°, and 550°C. A single result suggests that alundum may be useful as an inert solid. After a ten-hour fluorination at 350°C of a mixture of uranium and plutonium oxides and alundum, in which the initial plutonium concentration in the inert bed was about one per cent, the residual plutonium concentration in the inert bed was 0.03 per cent.

A program has been initiated to study the filtration of plutonium hexafluoride and plutonium-bearing particulate matter as it may appear in the glove box atmospheres of our plutonium handling facilities. This information should also be of use in designing plant installations.

2. General Chemistry and Chemical Engineering

a. Steam Hydrolysis of UF_6 to UO_2F_2 - The three-inch diameter fluid-bed reactor being used for the steam hydrolysis of uranium hexafluoride to uranyl fluoride has been modified to permit the entrainment and separate collection of fines formed during the reaction. This was done to circumvent the buildup of fines in the bed, which has been the major problem preventing continuous operation. A satisfactory $4\frac{1}{2}$-hour run was made under conditions used previously, 100 g/min uranium hexafluoride feed, 0.75 ft/sec superficial velocity (225 per cent excess steam), 18-inch bed height and reactor bed temperature of 200°C. The fines (-200 mesh) collected overhead constituted less than one per cent of the total material fed to the column (hexafluoride feed and "seed" particle recycle). Only a trace of fines was found in the bed at the end of the run.

b. Calcination Studies in Small Diameter Columns - Studies are continuing on a dual purpose scheme for the direct conversion of plutonium nitrate to plutonium dioxide. A bed temperature of about 350°C and a feed solution consisting of 1 M aluminum nitrate are currently being employed in these studies in a $2\frac{1}{4}$-inch diameter stainless steel column. Several successful runs up to five hours in duration have been made during the past period. Feed rates up to 18.5 ml/min were employed.

c. Preparation of Uranium and Plutonium Compounds - The possibility of synthesizing refractory plutonium and uranium compounds in liquid metal media is being explored. A plutonium carbide was formed by reaction of plutonium dissolved in liquid magnesium with fine graphite powder. The material was isolated and was found by X-ray diffraction analyses to be Pu_2C_3. No evidence for the presence of PuC could be detected.

3. Chemical-Metallurgical Process Studies

a. Liquid Metal Solvent Studies - The solubility of iron in liquid cadmium may be represented by the empirical equation for the temperature range from 420° to 647°C:

$$\log (\text{atom per cent}) = 1.236 - 3360\ T^{-1}$$

The solid phase in equilibrium with the saturated solutions over this temperature range is pure iron. These data are in good agreement with the preliminary results previously reported.

The gamma phase in the titanium-cadmium system has been found to have a body-centered tetragonal symmetry of the C-11 type. The composition of the phase corresponds to Ti_2Cd. The composition and structure of the delta phase, TiCd, has been reported previously (see January 1961, Progress Report).

Studies are being made to establish the epsilon-to-delta transformation temperature of U_2Zn_{17}. Powder compacts of compositions richer in uranium than the U_2Zn_{17} composition are annealed at temperatures in the range of 450° to 650°C. The products of the annealing treatment are subjected to X-ray diffraction examination for the identification of the phases formed. Preliminary results indicate that the epsilon phase transforms to the delta phase between 600° and 650°C.

The distribution coefficient of uranium between the partially miscible liquids lead and zinc has been measured as a function of the relative amounts of the two liquid phases. The distribution coefficient (weight per cent uranium in zinc/weight per cent uranium in lead) at 700°C was approximately 60 for mixtures of lead and zinc between 30 and 60 weight per cent lead.

b. Calorimetry - Preliminary combustions of cadmium and hafnium in fluorine have been completed. The study of the combustion of boron nitride in fluorine is being continued to determine whether the precision of the calorimetry is affected by the length of time required for temperature equilibration of the calorimeter after combustion.

Noncalorimetric combustions of uranium in fluorine are being carried out in an effort to obtain complete reaction.

Experimental work on the combustion of zirconium hydride in oxygen is being directed toward the development of a combustion technique that will allow the sample to be protected from exposure to oxygen prior to ignition.

The furnace of the high-temperature enthalpy calorimeter has been reassembled with a new shell and the system appears to be leak tight.

G. Advanced Reactor Concepts

1. Fast Reactor Test Facility (FARET)

A survey has been completed to explore the requirements for an experimental facility to test advanced fast reactor cores. A conceptual layout for this facility is being made to define the design problems.

For the first or reference core design, uranium monocarbide clad with Type 316 stainless steel is considered feasible for the specified coolant (sodium) outlet temperature of 760°C. Heat transfer calculations for a honeycomb core geometry showed a lower pressure drop and a higher heat flux than for cylindrical elements with the same specific power. Since the fabrication techniques for pin-type elements are well established, efforts are directed toward a design using this type.

An evaluation is being made of the 50-Mw heat exchange system for the facility. A U-tube type is considered for the sodium-to-sodium heat exchanger principally because of its feasibility for operation with a minimum of stress problems at high temperature.

2. Direct Conversion Survey for Mobile Systems

The purpose of this study is to find new applications of nuclear fission energy to so-called "advanced" or direct energy conversion schemes. Particular emphasis is placed on the fuel cell type of system, with consideration being given to both in situ and remote nuclear regeneration of fuel cell reactants.

Certain parameters for typical conversion devices and fuels were analyzed to rate the systems for performance as far as minimum weight requirements for different duration missions. The application chosen is that of the conventional 100 HP gasoline auto engine. For comparison, the final output of each power plant was converted to mechanical force; therefore, the electrical systems (solar cells, batteries, etc.) have an electric motor added. The nuclear reactor uses an indirect cycle liquid metal cooled reactor with a heat exchanger and gas turbine.

For application to passenger vehicles, a mission time of 10 hours was considered. The following table gives the performance of some systems relative to a gasoline engine. The comparison is made on the basis of the energy storage capability or watt-hr/kg for each system.

Table VIII. Performance of Various Power Plants Relative to the Gasoline Engine for Ten Hour Missions

Power Plant	Relative Performance
Gasoline engine	1.0
Unshielded reactor	1.1
H_2-O_2 fuel cell + liq. H_2 and air	0.51
H_2-O_2 fuel cell + liq. H_2 and liq. O_2	0.43
Ag-Zn battery	0.13
Solar cells	0.09
H_2-O_2 fuel cell + H_2 and O_2 in press. tanks	0.07
Shielded reactor	0.01

The conventional internal combustion automobile engine is about a factor of two above the closest competitor, the high pressure hydrogen-oxygen fuel cell. There are, however, disadvantages associated with this fuel cell in that liquid hydrogen is not amenable to routine handling as are more familiar fuels. The fuel cell can be built in a variety of forms, and it is not unreasonable to expect that systems using easy to handle liquid and perhaps even solid electrodes will be developed. In principle, a cell of this type would be no more difficult to handle than the conventional dry cell. Electrode reactants could be replaced directly or regenerated in situ by reaction of two safer, more readily handled primary fuels. It would appear that the prospects for the use of fuel cells as practical converters for use in surface transportation are good.

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