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ANL-5489

Metallurgy and Ceramics

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QUARTERLY REPORT
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METALLURGY DIVISION

ABSTRACT

Advanced Water Reactor Program - Activity 4102

Twenty-seven plates of the ternary EBWR reference alloy, integrally clad with Zircaloy-II, were evaluated for uniformity, core distribution, and bond quality. A comparison between twenty-one plates rolled in plain carbon steel jackets and six plates rolled in Ti-Namel (titanium-killed steel) jackets showed no measurable differences in bond quality or cladding uniformity. Width spread on rolling was 1.5 - 4.0% in Ti-Namel jackets, as compared with 3.0 - 6.5% in plain carbon steel jackets; however, width spread may be compensated for in the design of the billets. Corrosion testing of plates rolled in plain carbon steel jackets in 290°C water showed no detrimental effects from the steel. On the basis of these results, the use of Ti-Namel jackets offers no particular advantages over plain carbon steel jackets.

Roll cladding in narrow plain carbon steel jackets (6" in contrast to the standard 8" width) resulted in width increases of 8.0 to 14.0% during rolling, with appreciable variation within a 6 to 8" length. The use of narrow steel jackets to save material therefore cannot be justified.

Increasing the zirconium content of binary alloy cores from 5 w/o to 7.5 and 10 w/o improved the uniformity of finish rolled clad plates.

Postirradiation corrosion tests in distilled water at 290°C of samples from clad plates of the ternary alloy indicated that irradiations of even low burnups, of the order of 0.021 to 0.074 a/o, destroyed the corrosion resistance of the alloy. Samples similarly heat treated but not irradiated remained unaffected under the same corrosion testing conditions. The samples had a 1/8" diameter hole drilled through the clad to the core.

Development of the casting technology of the ternary EBWR core alloy has continued, with the effort directed toward the elimination of alloying element segregation, shrinkages, and blowhole cavities. The alloying irregularities and the imperfections that are revealed by ultrasonic testing have not been completely removed as yet. Bottom pouring in a three-cavity mold did not result in sound castings. The mold has been changed so that the three cavities are filled by top pouring. Greater inductive coupling of the metal in the melting furnaces improved alloying element distribution.

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✓ An ultrasonic through-transmission technique has been developed for evaluating the soundness of the EBWR core castings nondestructively. By utilizing a frequency of 5 Mc, this technique makes it possible to locate and record areas of excessive porosity, piping, and holes. Of 156 experimental cores tested, 27 were found to be free from defects. Generally, cores cut from the tops of the castings were characterized by considerable porosity; cores from the bottoms of castings were free from large defects.

✓ Most of the facilities required for the fabrication of the EBWR fuel plates have been installed and tested. Some existing equipment was re-located and additional equipment was obtained to handle billet component cleaning, billet assembly, roll cladding, and heat treatment of finished plates. The modifications provide for facilities to manufacture fuel plates on a projected schedule of 10 to 12 plates per 8-hour day.

✓ A method has been developed for measuring the clad thicknesses of EBWR plates by utilizing the attenuation of beta radiation emitted by Pa^{234} . A scintillation detector is used to detect the beta radiation after it has passed through the cladding material. At a scanning speed of 4" per minute, thickness variations of 0.0005" can be detected in 2" wide sections; at a scanning speed of 24" per minute, variations of 0.003" could be detected.

Fast Power Breeder Reactor Program - Activity 4104

Slugs from a prototype EBR-II fuel rod which had been irradiated in CP-5 to an estimated burnup of 0.38 a/o have been examined for dimensional changes. The slugs were uranium-2 w/o zirconium castings and were irradiated at 590° - 615°C; however, due to periodic safety scrams the lower slugs were apparently cycled briefly into the beta-phase region. This thermal history produced grain coarsening, gross bumping, and surface distortion on the lower slugs, whereas the upper slugs retained excellent dimensional stability.

A uranium-21 w/o plutonium-5 w/o molybdenum alloy casting, poured at 1510°-1525°C, yielded a casting density of 17.7 grams per cc, as compared with a theoretical X-ray density of 18.4 grams per cc. The grain size was fine and its machinability was roughly equivalent to the binary uranium-plutonium alloy. X-ray diffraction indicated a BCC structure with $a_0 = 3.42 \text{ \AA}$. The results of thermal expansion tests indicate, however, that the phase is probably metastable; repeated thermal cycling developed an arrest point at about 530°C. Below the transformation temperature an expansion coefficient of $18 \times 10^{-6} \text{ inch/inch/}^\circ\text{C}$ was obtained. The alloy could not be worked by rolling at 400°C.

A series of irradiation tests in the MTR on extruded uranium-plutonium alloy specimens containing 10, 15, and 20 w/o plutonium indicated that the 20 w/o alloy is quite promising as regards dimensional stability behavior under irradiation. The material elongated at a relatively low rate and maintained a smooth surface up to burnups of at least 0.60 a/o.

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General Reactor Development - Activities 4530 and 4540

A series of uranium powder compacts of (a) solid, full density material, (b) full density material with a 1/16" diameter axial core hole, and (c) material with approximately 12.5% porosity, were irradiated in the MTR up to 0.53 atom percent burnup. The full density material, both with and without a core hole, shortened under irradiation; the partial density compacts lengthened and developed elliptical cross sections.

Cast specimens of a uranium-5 w/o zirconium-1.5 w/o niobium alloy which had been quenched from salt at 850°C and aged at 400°C for various times were corrosion tested for 71 hours in degassed, distilled water at 290°C. The as-quenched sample acted in the normal, expected fashion; the aged samples corroded in a completely different fashion than did the vacuum treated and similarly aged metal. The hydrogen concentration increased with the aging time.

Open-ended sections of two Zircaloy-II clad, coextruded rods of a uranium-3.8 w/o epsilonized silicon alloy were tested in 290°C distilled water; after 10 days both core and bond appeared good. Postirradiation (0.71 a/o burnup) corrosion testing of an epsilonized cast specimen of this alloy indicated no harmful effects due to irradiation. The corrosion rate at 290°C was 31.9 mg/cm²/day.

Two cast and heat treated samples of an uranium-3 w/o niobium-0.48 w/o tin alloy were corrosion tested in 260°C water for 70 hours after an irradiation burnup of 0.19 and 0.58 a/o. The samples completely disintegrated in the corrosion test. Preirradiation corrosion rates at 290°C were 28.3 and 31.3 mg/cm²/day.

Two pressed and sintered UO₂ specimens were irradiated in the MTR to 0.16 and 0.38 a/o uranium burnups; both specimens broke into three pieces during irradiation.

U₃O₈-ThO₂ pressed specimens containing from 60 to 90 w/o U₃O₈ cracked when fired in air at 1650°, 1750°, and 1850°C. Additions of 2.5 w/o of Al₂O₃ or MgO appeared to eliminate the cracking problem. Similar additions of CaO, CaF₂, and ZrO₂ proved ineffective.

Thoria specimens containing 10 w/o enriched urania were examined after irradiation in a water loop in CP-5. The specimens broke up into discrete pieces as the exposures continued. It is likely that the cycling operation of the reactor has much to do with the progressive nature of the fragmentation.

A direct hot extrusion process is under investigation to produce partially enriched urania-aluminum fuel plates from powder mixtures of aluminum and urania in concentrations of 25 to 75 w/o urania. Conditions have been established for the satisfactory extrusion of 25 and 50 w/o mixtures.

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Additions of 0.01 w/o Be, 0.074 w/o Mg, 0.20 w/o Cr, 0.19 w/o Fe, 0.15 w/o Ti, and 0.03 w/o B to a high purity aluminum-0.9 w/o nickel base alloy proved ineffectual in reducing the corrosion rates of the alloy in 350°C water. In some cases the alloys disintegrated. The results, however, are not fully conclusive since the alloys picked up high silicon contents during melting, which is known to be detrimental. The tests will be repeated using higher purity aluminum crucibles to reduce the silicon pickup.

2S aluminum plates that had been prepared by Kanegin nickel plating and by nickel electroplating, and two bare plates of a 2S Al-0.46 w/o Ni alloy were tested in the MTR loop for 530 hours in 260°C water and additional periods of 50 hours at 230°C and 150 hours at 175°C; the total integrated flux was 5×10^{20} nvt. Examination showed that much of the Kanegin and electroplated nickel had been corroded and flaked away, exposing the underlying aluminum to attack. Corrosion of the bare aluminum-nickel alloy was not as severe as the corrosion of the 2S plates after failure of the nickel coatings.

As part of a program in the development of control rod and shielding materials, specimens of stainless steel containing boron or mixed rare earth oxides have been prepared by direct hot extrusion of the powder mixtures. Extrusions of nearly theoretical density were obtained using mixtures containing 3 and 10 w/o mixed rare earth oxides or 3 w/o boron in Type 302 stainless steel. Mixtures containing 10 w/o boron were extremely hot-short, and broke up upon leaving the die.

For accountability and for avoidance of overheating due to high local concentrations of fuel in small areas, it is often desirable to measure the amount of enriched fuel material present per unit area of a given plate. A gamma-ray transmission gauge has been developed, using a 0.25" diameter beam of gamma rays from a thulium source, which permits the detection of concentration variations of the order of 0.05 mg/cm².

A total of 2709 Borax-III fuel plates were bond tested by ultrasonics utilizing a through-transmission technique. Of these, 2706 were accepted and three were rejected due to lack of bond and because of cracks in the cladding layer.

Production, Treatment, and Properties of Materials - Activity 5410-01

The difficulties encountered in the preparation of 1-cm diameter single crystals of alpha uranium that are to be used for elastic constants measurements have been traced to the presence of a cross-sectional grain size gradient in the rods prior to coarsening. This condition arises because of differences in work hardening at the center and at the rod surfaces; the center of the rods work hardened to a lesser degree than did the rod surfaces. A number of variations in the step reduction schedules were tried in an attempt to minimize both banding and cross-sectional

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grain size gradients. To date, the minimum degree of grain size nonuniformity was obtained in 3/4" rods by a reduction schedule in which the first three reduction steps were 20 to 22% reductions of area, followed by two reduction steps of 30 and 42% reductions of area. Four rods swaged by this schedule appeared to contain a relatively uniform distribution of primary recrystallized grains, and it is hoped that they will provide suitable material for the production of single crystal of the requisite size.

A steep temperature gradient annealing furnace, which permits the maintenance of a 100°C temperature difference between the tip of a pencil shaped rod at 650°C and a point 1/8" away, has been constructed. The furnace consists of a large silver heat sink enclosed in a bell jar; it will be used to control the nucleation of single grains at the pointed ends in the coarsening process to produce single crystals.

Studies on the recrystallization textures of slightly deformed single crystals have been continued to include a crystal which was compressed approximately 8% in a direction 70° from the [100], 49.5° from the [010], and 47.5° from the [001]. The operative deformation mechanisms were twinning on {172} and {130} planes, and slip on the (010) plane. Of six recrystallized grains that were analyzed, two were found to have nucleated from the original orientation, three from the {172} twins, and one from the {130} twin orientation. The axis and magnitude of rotation necessary to bring the recrystallized orientations into coincidence with the original orientation is in agreement with the results reported for other crystals in previous quarterly reports.

Further microstructural studies have been made of uranium-zirconium alloys containing from 0.09 w/o to 1.07 w/o zirconium to define more accurately the solubility limits of the beta-phase region. The results indicate that the solubility of zirconium in beta uranium is at a maximum of slightly less than 0.41 w/o between 690° and 710°C, and is greater than 0.2 w/o at all temperatures above 670°C and below 750°C. Thermal analysis data of a 1.07 w/o Zr alloy indicated good agreement with the data of Summers-Smith (Journal of the Institute of Metals, 83, 277 1954-55) as regards the temperature horizontals of the phase diagram in this composition range.

The investigation of the uranium-niobium phase diagram has been extended to 10 w/o niobium; the eutectoid occurs at $634 \pm 2^\circ\text{C}$, and its composition is estimated at 8 ± 1 w/o niobium.

Metallographic studies of ternary uranium-zirconium-niobium alloys have determined the curved surface which bounds the lower limit of the single-phase gamma field (ANL-5465), but failed to delineate the lower surface of the two-phase region immediately below the gamma field. An alloy containing nominally 8 w/o zirconium and 2 w/o niobium has been investigated in an attempt to locate this transformation by electrical

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resistivity measurements. The data indicate that below 630°C gamma no longer exists as a stable phase, although the results are not firmly conclusive as yet.

The investigation of the influence of the nature and the metallographic features of second phase dispersions on the high temperature corrosion behavior of aluminum has been extended to aluminum-nickel-iron alloys. The aluminum-iron second phase compound was found to precipitate randomly throughout the grains; aluminum-nickel compound, on the other hand, precipitates largely at grain boundaries. These characteristics seem to be retained in the ternary aluminum-nickel-iron alloys. The photomicrographic observations of the second phase distributions provide an explanation for the role of iron in improving the corrosion resistance of the aluminum-nickel alloys, in conformity with the formerly postulated "hydrogen blister" theory. Nickel and iron apparently both form compounds which are cathodic to the aluminum grains, moving the corrosion cathodes away from the aluminum. However, if the geometry of the dispersion of the cathodic second phase permits comparatively large open areas, the aluminum metal in the center of these areas may successfully compete in the cathodic corrosion reaction. From photomicrographic observations it is apparent that the aluminum-nickel compound alone produces only a somewhat favorable geometry, but when combined with the random deposits of the aluminum-iron compound the distances between the second phase cathodic sites is reduced considerably, thus accounting for the improved corrosion resistance upon adding iron to aluminum-nickel alloys.

Hydrogen analyses of 2S aluminum before and after corrosion for 65 hours in 290°C water indicated a hydrogen increase from 0.4 ppm before corrosion to 1.3 ppm after corrosion. A similar test on a 2S aluminum-0.9 w/o nickel alloy corroded 57 days in 350°C water showed no such hydrogen pickup. Penetration of the metal by hydrogen does not appear to be a significant part of the corrosion process for this alloy.

Effects of Irradiation on Materials - Activity 5430-01

A group of natural uranium specimens was irradiated in the MTR in NaK-filled capsules under conditions such that the central metal of some of the specimens transformed to the beta phase. The central metal temperatures were varied by varying the diameters of the specimens. Some of the specimens were cycled into and out of the reactor for 50 times. Each specimen was subjected to very nearly the same integrated thermal flux; burnups were in the order of 0.0040 total atom percent. A 300°C rolled specimen which was cycled and did not exceed the alpha-beta transformation temperature had a slightly higher growth rate than a similar specimen which did transform in the central region. A cycled beta-quenched specimen whose center exceeded the alpha-beta transformation temperature developed a central void; a cycled beta-quenched specimen which remained in the alpha phase showed no void formation and shortened

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slightly. It is not known why the beta-quenched specimen developed a void while the 300°C rolled specimen of the same size showed no such effect; possibly the higher mechanical properties of the latter specimen enabled it to resist to a greater degree this type of deformation. Upon continuous irradiation, beta-quenched specimens showed no marked differences in over-all growth rate and no voids, regardless of whether the center had reached beta phase temperatures or not. The results would seem to point to cycling above the alpha-to-beta transformation temperature as a factor in void formation; in this connection it should be noted that voids were also obtained by some investigators on thermal cycling alone into the beta phase, under no irradiation.

Neutron flux distributions in irradiation specimens are being studied in a series of modified Wilkins experiments in which thin uranium foils are sandwiched between uranium wafers and then assembled into "solid" rods for activation in CP-5. The ends of the rods are separated by nonabsorbing graphite spacers of various thicknesses. The neutron flux distribution is obtained from measurements of the fission product activities of small punchings taken from the foils at various locations after irradiation. A considerable amount of data has been accumulated on the neutron flux distribution in 1" and 1/2" uranium rods and a statistical analysis of the data is in progress. Preliminary results indicate that in 1" diameter rods the neutron flux is 38% higher at the surface than at the center of a rod at locations away from the spacer. At a location adjacent to the spacer the increase in neutron flux at the surface varies with the thickness of the spacer: with no spacer the increase is 38%, with a spacer 0.300" thick it is 21%, and with very large spacers it is only 15%. The corresponding values for a 1/2" rod are 13%, 8%, and 4%, respectively.

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I. ADVANCED WATER REACTOR PROGRAM - ACTIVITY 4102

1. Fabrication and Evaluation of Uranium Alloy Fuel Plates (C. H. Bean, J. R. Lindgren, R. E. Macherey)

A sufficient number of plates have been prepared to further the development of the U-5 w/o Zr-1.5 w/o Nb alloy cores roll clad with Zircaloy-II. Additional data were obtained from plates scaled to the size of the EBWR fuel elements to establish billet design and dimensional tolerances for finish-rolled plates. Continued corrosion and irradiation testing of representative plates prepared during the past 18 months has contributed to the evaluation of the cladding process.

Binary uranium-zirconium alloys, considered as possible alternates for the EBWR reference ternary alloy, were roll clad with Zircaloy-II using procedures developed for the reference alloy. Increasing the zirconium content from 5 w/o to 7.5 and 10 w/o improved the uniformity of the rolled plate and of the cladding. Two clad plates of an U-5 w/o Zr alloy proved defective when corrosion tested in 290° C water; clad plates with 7.5 and 10 w/o zirconium have not been evaluated as yet.

a. Roll Cladding of Uranium-5 w/o Zirconium-1.5 w/o Niobium Alloy Plates

Twenty-seven plates of the EBWR reference alloy integrally clad with Zircaloy-II were evaluated for uniformity, core distribution, and bond quality. Plates scaled to the size of the EBWR fuel elements were included to establish the effects of steel jacket variables on the dimensional uniformity. Plates 0.250" and 0.280" in nominal thickness were prepared for subassembly testing.

A comparison between 21 plates rolled in plain carbon steel jackets and 6 plates rolled in Ti-Namel (titanium-killed steel) jackets showed no measurable differences in bond quality or cladding uniformity. Corrosion testing of plates rolled in plain carbon steel jackets in 290° C water showed no detrimental effects from the steel. From the viewpoint of corrosion, no advantage can be attributed to the use of Ti-Namel steel jackets. Physical measurements of width spread on rolling showed that billets rolled in Ti-Namel jackets spread only 1.5 to 4.0%, as compared with 3.0 to 6.5% when rolled in plain carbon steel jackets of the same size. Width spread may be compensated for in the design of the billets, making it possible to roll plates to a predetermined size in either Ti-Namel or plain carbon steel jackets.

Cladding billets Nos. 211 to 214, inclusive, were rolled in plain carbon steel jackets with the width reduced to 6" from the standard 8" width. The purpose of this change was to determine whether EBWR plates could be rolled to a uniform width in a narrower jacket to effect a saving in the steel

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used per jacket. The width of these four billets increased from 8.0 to 14.0% during rolling, with appreciable variation within a 6" or 8" length of any given plate. The use of narrow steel jackets to save material cannot be justified due to the lack of plate uniformity.

Although zirconium-tipped Zircaloy-II screwed plugs provided a satisfactory seal for purge-welded and evacuated billets, the rolled-plate ends frequently contained continuous or segmented inclusions around the original screwed plug. A redesigned tapered seal plug was substituted for the screwed plug on billets Nos. 195 - 217. Six of the plate end closures containing these tapered seal pins were examined metallographically and showed very few segmented inclusions at the seal-plug to end-plug interface.

b. Roll Cladding of Binary Uranium-Zirconium Alloy Plates

Four binary uranium-zirconium alloy plates, Nos. 187 - 190 inclusive, were clad with Zircaloy-II. Billets 187 and 188 contained cast U-7.5 w/o Zr alloy cores, and billets 189 and 190 contained cast U-10 w/o Zr alloy cores. The cladding components were prepared and the billets were assembled according to the procedures developed for cladding the EBWR reference alloy. The four billets were roll clad 80% reduction in thickness at 850° C. The rolled plates were uniform in width, length, and thickness, and the plate surfaces were smooth and free from defects. There were little or no irregular end effects or side effects at the core-to-clad interface. Ultrasonic through-transmission tests indicated probable weak or discontinuous bonding in some areas of plate No. 189, particularly between the side cladding and the core. No bond line defects were detected in the other plates. Complete evaluation of cladding uniformity, metallographic examination, and bond strength will be completed after corrosion testing in 290° C water.

c. Effects of Irradiation on Corrosion Resistance of Uranium-5 w/o Zirconium-1.5-w/o Niobium Alloy Plates (S. Greenberg)

Samples from two clad plates, AH-10-2 and AH-12-2, were tested; the indicated burnups were 0.021 a/o and 0.079 a/o, respectively. Exposed core ends and edges were weld clad with Zircaloy-II. The samples were heated 15 minutes at 850° C, water quenched, and aged at 425° C for 45 minutes. After irradiation a 1/8" diameter hole was drilled through the clad to the core of each sample.

Sample AH-10-1 was tested in degassed, distilled water for 58 hours between 240° and 275° C. At the end of this period there was loose oxide at the hole but no swelling. Sample AH-10-2 was similarly tested for 66 hours at 290° C. At the end of this period the undrilled cover plate was almost completely severed from the assembly (attached at one corner). The drilled cover plate was not as completely separated. There was no

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swelling of the cover plate at the drilled hole. All failures were along welds. The core was swollen to at least twice its original thickness and was apparently completely converted to oxide.

A sample duplicate in every respect to AH-10-2 and AH-12-2, except that it was not irradiated, was tested at 290° C for 95 hours; there was no change except difference in color between core and clad. It appears certain that even irradiation to low burnup destroys the corrosion resistance of this alloy.

d. Irradiation Stability of Uranium-5 w/o Zirconium-1.5 w/o Niobium Alloy (J. H. Kittel, S. H. Paine)

A group of uranium-zirconium-niobium specimens having optimum pre-irradiation corrosion properties has been irradiated and examined. Some of these specimens were heavily restrained by tight jackets in the hope that dimensions and surface smoothness might be relatively unaffected by the irradiation. At burnups ranging up to 0.33 a/o, the restraining jackets proved to be very effective in maintaining dimensions and surface smoothness of the specimens. Subsequent corrosion testing of this group should indicate the effect, if any, of surface condition on postirradiation corrosion rates.

2. Manufacture of EBWR Fuel Plates

a. Melting and Casting of Uranium-5 w/o Zirconium-1.5 w/o Niobium Alloy (R. J. Dunworth, R. E. Macherey)

During the past quarter, the production of EBWR alloy cores of 93.5 w/o U-5 w/o Zr-1.5 w/o Nb was of primary concern. Almost all the foundry time was used in the effort to make sound castings, homogeneous in alloy distribution, and free from shrinkage and blowholes. The mold used for some 17 castings was the one described in ANL-5465 (1-1/4" x 11-1/2" in cross section and 35" deep), which is suitable for seven EBWR cores. Gross defects such as cold shuts, large surface blowholes, and large shrinkage areas on the sides of the castings were eliminated after three or four castings were made. The alloying irregularities and the imperfections that were revealed by ultrasonic testing have not as yet been removed completely. The last two castings showed defects in the top 7" (2 cores); the bottom 18" (5 cores) of each casting were sound. The zirconium and the niobium were segregated from top to bottom and from center to edge, with the alloying element concentrations higher at the top and in the center of the castings. The castings were made in a 20" I.D. quartz tube furnace; the graphite crucible was 14" O.D. x 11" I.D.

Six castings were made in a newly designed, three-cavity graphite mold. The cavities were separated by graphite plates so that each cavity was 1-1/4" x 3-1/2" in cross section and 24" deep, each cavity

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can produce two EBWR cores. The mold was designed for bottom filling, that is, the metal was poured into the central cavity and bottom gated into the two side cavities. Sound castings were not obtained by this procedure and the mold has been changed so that all three cavities are filled by top pouring. The castings made by bottom filling, although unsound due to secondary pipe, were quite homogeneous in alloy distribution. The castings were made in a 12" quartz tube furnace with a 14" I.D. induction coil and an 8" I.D. x 9-1/4" O.D. graphite crucible. The greater inductive coupling of the metal obtained in this furnace is considered to be the primary factor responsible for the homogeneous alloying element distribution in these castings.

b. Ultrasonic Testing of EBWR Core Castings (W. J. McGonnagle, N. Beck)

One hundred fifty-six experimental core castings of the reference EBWR alloy were tested by an ultrasonic through-transmission technique. The test items measured approximately 11" in length, 3-1/4" wide, and 1-1/4" deep. The cores were identified by a stamped number located at one end of the plate. A prefix number was used to identify the casting and a suffix number to locate the individual core in the mold. By utilizing a frequency of 5 Mc it was possible to locate and record areas of excessive porosity, piping, and holes. It was also possible to denote an over-all change in grain structure of one core as compared to another.

In order to establish a criterion for acceptance, several of the tested cores were sectioned and the extent of the flaw determined. A particular section was then selected and used as a standard to which all subsequent tests were compared. In this manner it was possible to evaluate individual cores in terms of a known defect. Of the 156 cores tested, 27 were found to be free from defects. The remainder indicated piping, holes, or porosity equal to or greater than the standard. The core cut from the top of the casting was generally characterized by an area of considerable porosity; the cores cut from the bottom of the casting were free from large defects.

Inasmuch as the testing method constituted a through-transmission technique, and the defect recorded may be located either directly below the surface or in the middle of the core, comparison tests were made of cores which had an "as cast" finish with those that had a machined finish. It was noted that surface characteristics effects were minor compared with a flaw in the middle of the core.

The fabrication process was changed through the program and for that reason it is not possible to denote a progressive tabulation of results. It was noted, however, that castings K32 and K21, which were cut into seven cores, had defects in only the top two cores. This ratio of acceptance was above average for all the other castings that were tested

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c. Installation and Testing of Equipment for Manufacture of Fuel Plates (R. E. Macherey, C. H. Bean)

Most of the facilities required for the manufacture of the EBWR fuel plates have been installed and tested. Some existing equipment was relocated and altered, and additional equipment was obtained to handle billet component cleaning, billet assembly, roll cladding, and heat treatment of finished plates. These modifications were necessary to manufacture fuel plates on a projected schedule of 10 to 12 per 8-hour day.

Equipment for Cleaning Billet Components: Two Vaporblast Model B-2 machines were relocated and converted to automatic operation to uniformly clean hot-rolled Zircaloy-II sheet and cold-finished cladding components. Each machine was equipped with two additional blast nozzles, increased pumping capacity, and a larger compressed air supply line. All three nozzles on each machine are attached to a hydraulically driven reciprocating arm to give a uniform cleaning pattern across the work. The work travels under the nozzles on a variable speed power-driven carriage.

One smaller air-water-grit blasting machine with a stainless steel cabinet was constructed for cleaning the uranium alloy cores prior to assembly. A specially designed nozzle mounted on an air-driven reciprocating arm blasts a uniform 4" wide pattern across the core. Cores are placed on a stainless steel rack and are totally enclosed inside the cabinet during blasting. Controls for all three vapor blasting machines were located in a central control panel.

Facilities were installed for rinsing Zircaloy-II in running tap water to remove loose grit after vapor blasting. An automatic air-water blast machine was built to remove embedded as well as loose grit from the surface of alloy cores. Three air-water nozzles on this machine were also attached to an air-driven reciprocating arm which was mounted on a power-driven carriage over an open tank containing the material to be cleaned.

Welded stainless steel pickling tanks and various clamps, handling tongs, and storage racks were added to the existing metal cleaning equipment.

Equipment for Assembling Cladding Billets: The press welding facility with pressurized water-cooled copper platens on vertical and horizontal rams which was used for clad plate development work was considered adequate for fuel element production with but few alterations. Slightly larger water-cooled copper platens were required for the EBWR billets, and an automatic welding carriage was added to produce more uniform assembly welds.

Substitution of tapered billet end seals for screwed seals has simplified the closure and welding of evacuated billets. Necessary equipment has been designed to handle the evacuation and end closure of a minimum of 12 billets per day by this improved method.

Equipment for Roll Cladding Fuel Plates. In order to have sufficient space for heating, handling, and rolling the EBWR fuel plates without relocating the 16" x 24" United Mill, a 16' x 16' Butler type building was added to the west of the fabrication building, D-16.

The 6', 36-kw Hevi Duty electric resistance element furnace used for clad plate development was relocated on the east side of the 16" x 24" United Mill. Heat-resistant alloy tracks were placed on the furnace hearth and heat resistant alloy roller buggies were purchased to use for charging cladding billets into this furnace for billet preheating.

A new 9', 150-kw Hevi Duty electric resistance element furnace was installed for heating billets and plates during rolling. This furnace was placed on the west side and in tandem with the mill. Doors at each end of the furnace and a heat resistant alloy roller hearth simplify manipulation of billets and plates to and from the mill. Guides were placed between hearth rollers to help locate three billets or plates across the width of the hearth. Recording temperature controllers and temperature limiting controls for both furnaces were located in the building annex.

Furnace entry tables and mill feed tables were needed to convey hot billets and plates between the furnaces and the rolling mill. A short extension table with tracks for the roller buggies is used to transfer preheated billets from the 36-kw furnace to the mill feed table. Power-driven variable speed reversible roller feed tables were located on each side of the mill. These tables were designed to handle billets or plates up to 12" in width and 8' long. Both tables are equipped with air cylinder operated side rollers to position or center the work before it enters the mill. A 42" wide x 116" long power-driven variable speed reversible roller table was built to transfer material from the 150-kw furnace to the mill. Cylinder operated guides on each side of this table align billets for rolling or re-entry to the furnace

The 16" x 24" United Mill was modified to increase the mill capacity and simplify control of screwdown settings for production rolling. The spring-type balancing mechanism for the top roll was replaced with hydraulic lifters, and the bronze roll neck bearings were replaced with 4-row tapered roller bearings in new steel chocks. A new set of 17" diameter, hot flat rolls was installed. New 17" diameter, cold finishing rolls are also available if needed to cold finish Zircaloy-II strip for billet cladding components. The addition of an automatic screwdown control mechanism by which any desired rolling schedule may be preset eliminates operating errors when rolling several billets simultaneously.

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The 50-ton capacity Aetna Standard hydraulic straightening press was modified by placing 10" x 84" water-cooled steel platens on the ram and bed of the press. Hot-rolled plates are reheated in the large Hevi Duty furnace after the last roll pass and then flattened in the press.

Equipment for Heat Treating Rolled Plates: One of the existing Hevi Duty pot type furnaces was completely rebuilt and relocated adjacent to a similar furnace to facilitate isothermal heat treatment of finish rolled plates. A new 500-pound capacity electric hoist was installed to move plates in and out of these furnaces.

d. Measurement of Clad Thickness of EBWR Fuel Plates
(W. J. McGonnagle, N. Beck)

The attenuation of the beta radiation emitted by Pa^{234} (a second daughter of U^{238}) can be used to measure clad thickness of EBWR fuel elements. A scintillation detector is used to detect the beta radiation after it has passed through the cladding material. The scintillating material is an anthracene disk, 1" in diameter. The scintillator is attached to a Dumont 6292 phototube.

Three EBWR type fuel plates have been scanned by this method. Variations in the clad thickness of the three plates were observed, and strips of stainless steel of 2" width were glued to the plates in an attempt to detect their presence as artificial variations in the clad thickness. It was concluded that with a scanning speed of 4" per minute, a variation of 0.0005" in the cladding could be detected in sections of 2" widths or wider; and at a scanning speed of 24" per minute, 0.003" variations could be detected. With the adjustments as described, the change in the output was clearly above statistical variation. With a scanning speed of 24" per minute, a 0.005" variation in widths of 2" is easily seen. For the faster scanning speed it was found desirable to reduce slightly the time constant of the count-rate meter. Choice of scanning speed depends upon sensitivity desired and time available.

e. Fabrication of EBWR Subassemblies (C. C. Stone, R. A. Noland, F. D. McCuaig)

A semiautomatic welding device which will permit inert arc spot welding of Zircaloy-II clad EBWR subassemblies in a sealed, helium-filled chamber is being constructed. This machine is expected to be capable of welding a minimum of two subassemblies per eight-hour day.

II. FAST POWER BREEDER REACTOR PROGRAM - ACTIVITY 4104

1. Examination of Fuel Slugs from EBR-II Prototype Fuel Element (F. L. Brown, S. H. Paine)

The Reactor Engineering Division requested examination of slugs disassembled from prototype fuel rod RE-17 which had been irradiated in a CP-5 thimble to an estimated maximum burnup of 0.38 a/o. The fuel element was made from U-2 w/o Zr castings and had been operated in the nominal range of 590° - 615° C (1100° - 1140° F). However, a periodic safety scram test of the reactor gave a large enough enhancement of flux to cycle the lower slugs in the rod briefly into the beta-phase region. This thermal history produced grain coarsening, gross bumping, and surface distortion on the lower slugs, whereas the upper slugs retained excellent dimensional stability. A comparison of upper and lower slugs is shown in Figure 1.

2. Properties of an Uranium-21 w/o Plutonium-5 w/o Molybdenum Alloy (J. E. Baumrucker)

Several castings of nominal composition U-21 w/o Pu-5 w/o Mo were made. Pouring temperatures in the range 1510° - 1525° C yielded a fluid pour, but because of turbulence, porosity was not entirely eliminated. Casting density obtained was 17.7 grams per cc, somewhat less than the theoretical density of 18.4 grams per cc calculated from X-ray data; casting hardness was in the range 76.5 - 72.5 Rockwell A. Fracture tests indicated a fine grained material, which was corroborated by metallographic examination. Machinability was roughly equivalent to that of the binary uranium-plutonium alloy. X-ray diffraction showed the structure to be BCC with $a_0 = 3.42 \text{ \AA}$. However, results of thermal expansion tests indicated that this phase is probably metastable. On repeated cycling in the apparatus, an arrest point developed in the vicinity of 530° C. After three cycles an irreversible elongation of 0.02" per inch remained; most of this elongation occurred above 530° C. The maximum temperature reached was 713° C. Below the transformation temperature the heating and cooling curves were roughly parallel with an expansion coefficient of $18 \times 10^{-6} \text{ inch/inch/}^\circ\text{C}$.

The material was not workable by rolling at 400° C. Rolling with unheated rolls at preheat temperature of 675° C permitted approximately 9% reduction before breakup. Further attempts at fabrication including the use of heated rolls and extrusion are in progress.

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Figure 1. Slugs from Irradiated Rod RE-17.

1-a. Upper and middle slugs showing undistorted surfaces.

Macro 17592

6-X

1-b. Lower slugs showing bumps and surface distortions resulting from cycling into the beta phase.

Macro 17586

6-X

3. Effects of Irradiation on Extruded Uranium-Plutonium Alloys
(J. H. Kittel, S. H. Paine, R. J. Fousek, F. Pausche)

An earlier Quarterly Report (ANL-5439) contains a description of the effects of irradiation on a group of cast specimens containing 5, 10, and 15 w/o plutonium in uranium. These specimens were all badly damaged by the exposure they received and this appeared to be chiefly the result of the large grain size of each specimen. In an effort to produce finer grained material, use has been made of the extrusion process. A total of 14 specimens was made by L. R. Kelman by this means, using compositions of 10, 15, and 20 w/o plutonium in uranium. They were irradiated in the MTR in NaK-filled capsules and have been examined for changes resulting from irradiation. A summarized description of the specimens and the changes observed are given in Table I. Photographs of typical irradiated specimens are shown in Figures 2 through 9.

It can be seen from the data in the table and from Figure 8 that the U-20 w/o Pu alloy in the as-extruded condition is quite promising in its behavior under irradiation. The material elongated at a relatively slow rate and also maintained a smooth surface up to burnups of at least 0.60 a/o, (5100 MWD/T).

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TABLE I

Effects of Irradiation on Uranium-Plutonium Alloys

Nominal w/o Pu ^(a)	Condition	Co ⁶⁰ Indicated Burnup		Calculated Central Temp. (°C)	Length Change (%)	Growth Rate (microinches/inch/ ppm burnup)	Remarks
		a/o	MWD/T				
0	As extruded ^(b)	0.045	380	85	-4.1	-91	Part of specimen could not be removed from irradiation capsule.
0	As extruded	0.048	410	90	-5.1	-110	
10	As extruded	0.70	6000	780	-	-	
10	As extruded	0.35	3000	410	-	-	Could not be removed from irradiation capsule.
10	Heat treated ^(c)	0.45	3800	520	-	-	Ignited and burned before measurements could be taken.
10	Heat treated	0.25	2100	300	1.5	6	Very rough surface.
15	As extruded	0.32	2700	380	47	150	Very rough surface.
15	As extruded	0.80	6800	890	97	85	
15	Heat treated	0.70	6000	780	39	47	
15	Heat treated	0.14	1200	190	49	285	Very rough surface.
20	As extruded	0.60	5100	680	5.2	9	Rough surface.
20	As extruded	0.40	3400	460	5.7	14	
20	Heat treated	0.52	4400	590	5.2	10	
20	Heat treated	0.25	2100	300	10	40	Rough surface.

(a) Balance natural uranium.

(b) Extrusion temperature, 450° to 525°C.

(c) All heat treatments consisted of heating to 645°C, cooling to and holding at 500°C for one hour, and cooling to room temperature.

Figure 2. Appearance of a Typical Uranium-Plutonium Alloy Specimen Before Irradiation.

Macro 17225

2-X

Figure 3. Unalloyed, As-Extruded Uranium Specimen After 0.045 a/o Burnup (380 MWD/T) at a Calculated Central Temperature of 85°C.

Macro 17663

2-X

Figure 4. As-Extruded U-10 w/o Pu Alloy Specimen After 0.70 a/o Burnup (6000 MWD/T) at a Calculated Central Temperature of 780°C.

Part of this specimen could not be removed from its irradiation capsule.

Macro 17665

2-X

Figure 5. Heat-Treated U-10 w/o Pu Alloy Specimen After 0.25 a/o Burnup (2100 MWD/T) at a Calculated Central Temperature of 300°C.

Macro 18740

2-X

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Figure 6. As-Extruded U-15 w/o Pu Alloy Specimen After 0.80 a/o Burnup (6800 MWD/T) at a Calculated Central Temperature of 890°C.

Macro 17666

2-X

Figure 7. Heat-Treated U-15 w/o Pu Alloy Specimen After 0.70 a/o Burnup (6000 MWD/T) at a Calculated Central Temperature of 780°C.

Macro 18742

2-X

Figure 8. As-Extruded U-20 w/o Pu Alloy Specimen After 0.60 a/o Burnup (5100 MWD/T) at a Calculated Central Temperature of 680°C.

Macro 18743

2-X

Figure 9. Heat-Treated U-20 w/o Pu Alloy Specimen After 0.25 a/o Burnup (2100 MWD/T) at a Calculated Central Temperature of 300°C.

Macro 17668

2-X

III. GENERAL REACTOR DEVELOPMENT - ACTIVITIES 4530 AND 4540

1. Development of Fuel Element Materials

a. Uranium Powder Compacts: Irradiation Effects (J. H. Kittel, S. H. Paine, R. J. Fousek, F. Pausche)

As stated in the last Quarterly Report (ANL-5465), irradiations are being made on three types of hot-pressed uranium powder compacts, prepared by Sylvania Electric Products, Inc., and 3/8" in diameter and 1" long. The following types of compacts are being investigated: (1) solid, full density material; (2) full density material with a 1/16" diameter axial core hole; (3) partial density material with approximately 12.5% porosity. The initial group of specimens described in ANL-5465 was irradiated to total atom burnups ranging up to 0.28% (2400 MWD/T).

More recently, a second group was irradiated at higher temperatures in the MTR to burnups ranging up to 0.53 a/o (4500 MWD/T). The observed results of this irradiation are summarized in Table II. Photographs of typical specimens of both groups are shown in Figures 10 through 12.

TABLE II

Effects of Irradiation on Hot Pressed Uranium Powder Compacts

Specimen Type	Co ⁶⁰ Indicated Burnup(a/o)	Irradiation Temp. Range	Growth Rate (Microin/in/ ppm Burnup)	Remarks
Solid	0.49	$\alpha + \beta$	-	Could not be removed from irradiation capsule.
Solid	0.52	$\alpha + \beta$	-	Same
Solid	0.45	α	- 7	Uneven diameter change.
Cored	0.53	$\alpha + \beta$	- 50	Badly distorted.
Cored	0.16	α	- 25	Defect at one end.
Cored	0.46	$\alpha + \beta$	- 26	Warped.
13.0% porosity	0.51	$\alpha + \beta$	39	Oval-shaped cross section.
12.5% porosity	0.33	α	53	Same

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Figure 10. Effects of Irradiation on Full Density Compacts.

The specimen with 0.52 a/o burnup could not be entirely removed from its irradiation capsule, part of which is shown in the photograph.

	Macro 17622	2-X	Macro 17601	2-X	Macro 18699	2-X	Macro 18694	2-X
Burnup, a/o	0		0.28		0.45		0.52	
MWD/T	0		2400		3800		4400	
Calculated Central Temperature, °C	-		660		610		740	

Figure 11. Effects of Irradiation on Full Density Compacts with a 1/16" Diameter Axial Core Hole.

The specimen with 0.53 a/o burnup ignited and burned to oxide shortly after it was photographed.

Macro 17622

2-X

Macro 17603

2-X

Macro 18710

2-X

Macro 18700

2-X

Burnup, a/o

0

0.28

0.46

0.53

MWD/T

0

2400

3900

4500

Calculated Central
Temperature, °C

-

660

670

770

Figure 12. Effects of Irradiation on Compacts with 12.5% Porosity.

	Macro 17622	2-X	Macro 17607	2-X	Macro 18716	2-X	Macro 18715	2-X
Burnup, a/o	0		0.24		0.33		0.51	
MWD/T	0		2000		2800		4300	
Calculated Central Temperature, °C	-		580		410		730	

The full density material, both with and without a core hole, shortened under irradiation. These compacts were formed by end pressing. The partial density compacts, on the other hand, which were formed by side pressing, lengthened under irradiation and developed elliptical cross sections. The results indicate that a slight preferred orientation is present in the texture of the compacted specimens.

b. Corrosion Resistance of Uranium-5 w/o Zirconium-1.5 w/o Niobium Alloy (S. Greenberg)

Work designed to correlate corrosion damage and hydrogen absorption in the U-5 w/o Zr-1.5 w/o Nb alloy has continued. As previously reported, a relatively high hydrogen content, e.g., 35 ppm as found in a sample quenched from salt at 850°C, is not detrimental to corrosion resistance in the as-quenched condition; this is, perhaps, because of the uniform distribution of the hydrogen.

A group of samples which had been quenched from salt were aged for various times at 400°C and tested for 71 hours in degassed, distilled water at 290°C. The results are shown in Table III. The as-quenched sample acted in the normal expected fashion. The aged samples corroded in a completely different fashion than did vacuum-heat treated and similarly aged metal. It is thought that the aging process causes the hydrogen to concentrate, perhaps at grain boundaries. This concentration may act as a nucleus for further rapid hydrogen absorption. It is significant that this is the first instance in which very small pieces of metal close to cracked surfaces have been analyzed. Very high hydrogen contents were found, as had been previously postulated. Unfortunately, the remnants of the corrosion test were not large enough to permit both hydrogen analysis and metallography. The series of heat treatments will be repeated and the samples examined before corrosion testing.

TABLE III

Effect of Aging on Corrosion Resistance and Hydrogen Absorption
of U-5 w/o Zr-1.5 w/o Nb Alloy No. L-170B

Cast and quenched from salt at 850°C

Sample	Aging Time at 400°C (hours)	Corrosion Rate (mcd)	Hydrogen Content of Alloy After Test (ppm)	Appearance of Sample After Test
1	0	13.5	60	Dull, pitted.
2	1	337*	832	Sample cracked into an irregularly shaped piece and slurry. The piece was in good condition.
3	2	450*	1220	Same as above.
4	3	616*	1340	Same as above.
5	4	-	-	Completely disintegrated.

*Based on remaining piece.

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c. Uranium-3.8 w/o Silicon Alloy

Irradiation Effects (J. H. Kittel, S. H. Paine,
R. J. Fousek, F. Pausche)

Two specimens of a U-3.8 w/o Si alloy cast by Nuclear Metals, Inc., and scheduled for postirradiation corrosion testing were irradiated to uranium atom burnups of 0.12 and 0.33%. The specimens changed in length 2.1 and -1.4%, respectively, and some surface cracks developed in the specimen having the higher burnup. The specimens were homogenized at 800°C for 2-3/4 days by the supplier prior to irradiation. The appearance of the specimens before and after irradiation is shown in Figures 13 through 15. The effects of irradiation on the two specimens were generally the same as noted on an earlier group (described in ANL-5465) which was irradiated at higher temperatures and to higher burnups.

Corrosion (S. Greenberg)

Open-ended sections of two Zircaloy-II clad coextruded rods of a U-3.8 w/o Si alloy, fabricated by Nuclear Metals, Inc., were tested at 290° C in degassed, distilled water. The alloy had been heated at 800° C for seven days prior to extrusion. After about 10 days of testing, both core and bond appeared good. The corrosion rates were comparable with as-cast material. Additional samples will receive further epsilonizing treatment to determine if the corrosion rate can be lowered.

A 10% enriched sample (BB-3) of the alloy of this composition, was cast at Argonne and epsilonized at 825° C for two hours, and was corrosion tested bare after an indicated uranium burnup of 0.71 a/o on irradiation. Results of the corrosion tests at 260° C and 290° C are listed in Table IV. Although corrosion behavior was only fair, it was approximately the same as shown by the same alloy (apparently not too well made) before irradiation. It appears that irradiation is not harmful to the corrosion resistance of this alloy.

Figure 13. Appearance of a Typical U-3.8 w/o Si Alloy Specimen Before Irradiation.

Macro 18738

2-X

Figure 14. U-3.8 w/o Si Alloy Specimen After 0.12% Burnup of Uranium Atoms.

Calculated surface temperature was 150°C.

Macro 18647

2-X

Figure 15. U-3.8 w/o Si Alloy Specimen After 0.33% Burnup of Uranium Atoms.

Calculated surface temperature was 310°C.

Macro 18645

2-X

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TABLE IV

Corrosion of Irradiated Uranium-3.8 w/o Silicon Alloy (BB-3)

Test Temperature (°C)	Time (hours)	Corrosion Rate (mcd*)	Appearance After Test
260	24	0.13**	No apparent change except for small silver-colored spot near one end.
290	87	31.9***	Loose particles in water and on sample. Relatively large piece cracked off one end. Other end roughened and has hole in cylindrical surface.

*Preirradiation corrosion rate at 290°C was 15.7 mcd after 88 hours.

**Weight change small enough to have been balance error.

***Based on major piece.

d. Uranium-3 w/o Niobium-0.48 w/o Tin Alloy (S. Greenberg)

Two 10% enriched samples of the above alloy that were cast at Argonne, Nos. BB-5 and BB-9, were tested in degassed, distilled water at 260°C for 70 hours. The samples were water quenched after 4 minutes at 800°C and were aged at 350°C for 6 hours. Indicated irradiation burn-ups were 0.58 a/o and 0.19 a/o, respectively. Preirradiation corrosion rates at 290°C were 28.3 and 31.3 mg/cm²/day, respectively, for 88-hour tests. After irradiation the samples were roughened and warped. Growth rates on irradiation were 15 and 38 microinches/inch/ppm burnup, respectively. The samples were completely disintegrated in the post-irradiation corrosion test.

e. Urania Fuel Bodies (J. H. Kittel, S. H. Paine)

Two pressed and sintered 3/16" diameter specimens of UO₂ were irradiated to 0.048 a/o uranium burnup in a NaK-filled capsule in the MTR. It was found on examining the specimens after irradiation that they had fractured into a number of small pieces. The possibility of damage because of mechanical shock could not be ruled out, however, because the specimens were free to move about in their capsules.

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Accordingly, two identical specimens (shown in Figure 16) were irradiated after packaging in steel wool to eliminate the possibility of mechanical damage. Irradiation conditions were as follows:

	<u>AB-102</u>	<u>AB-103</u>
Uranium atom burnup, %	0.16	0.38
Calculated central temperature, °C	190	315
Calculated surface temperature, °C	95	140
Diameter change, %	0.0	0.08

Both specimens broke into three pieces during irradiation, as shown in Figures 17 and 18.

f. Urania-Thoria Fuel Bodies

Fabrication (J. H. Handwerk, R. A. Bach)

Additional U_3O_8 - ThO_2 mixtures containing from 60 to 90 w/o U_3O_8 were fabricated by isostatic pressing under a pressure of 50,000 psi. These bodies were fired in air to temperatures of 1650°, 1750°, and 1850°C for various time cycles in an effort to eliminate the cracking which had been encountered in bodies of these compositions. All of the bodies disintegrated in firing. As an aid to stabilize high U_3O_8 bodies, several oxides (MgO , CaO , CaF_2 , Al_2O_3 and ZrO_2) were added in amounts of 2.5 w/o to U_3O_8 . These compositions were fired to 1750°C in air. The body containing Al_2O_3 appeared to be sound, and the body containing MgO appeared to be good, but showed a few minor cracks which could be due to faulty pressing; the other bodies were cracked rather severely.

Irradiation (F. L. Brown, S. H. Paine)

A number of examinations have been made of thoria bodies containing 10 w/o enriched urania after irradiation in a water loop in the CP-5 reactor. Such bodies are apparently highly susceptible to fragmentation under the thermal stresses imposed by the irradiations and break up into discrete pieces as the exposures continue. Specimens and their fragments seem to be unaffected by the water present during irradiation. It is very likely that the cyclic operation of the reactor has much to do with the progressive nature of the fragmentation.

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Figure 16. Appearance of UO_2 Specimens AB-102 and AB-103
Before Irradiation.

Macro 16126

2-X

Figure 17. UO_2 Specimen AB-102 After 0.16% Burnup of the Uranium Atoms
at a Calculated Central Temperature of 190°C .

Macro 18635

2-X

Figure 18. UO_2 Specimen AB-103 After 0.38% Burnup of the Uranium Atoms
at a Calculated Central Temperature of 320°C .

Macro 18636

2-X

g. Urania-Aluminum Fuel Plates (D. E. Walker, R. A. Noland,
J. H. Handwerk)

A direct hot-extrusion process is under investigation to produce partially enriched urania-aluminum fuel plates from powder mixtures of atomized aluminum and urania. Compositions ranging from 25 to 75 w/o urania in multiples of 25 w/o have been investigated. Conditions which will permit satisfactory extrusion of 25 and 50 w/o mixtures have been developed. Roll cladding of the 50 w/o material has been successfully accomplished.

h. High Temperature Corrosion-Resistant Aluminum Alloys
(W. E. Ruther, C. A. Youngdahl)

An attempt was made to improve the aqueous corrosion resistance of 2S Al-1% Ni alloys by further alloying. It had been shown (Quarterly Report, March 31, 1955, ANL-5439) that the weight gain curves of 0.5, 1, 1.5, and 2% nickel in 2S aluminum when corroded in 315°C distilled water had the same slope (corrosion rate). This suggested that the cathodic corrosion reaction (liberation of hydrogen) was not the rate-controlling step in the corrosion, since changing the amount of cathodic area by a factor of 4 did not influence the corrosion rate. However, this argument is not clear-cut, since it is possible that the rate is controlled in the anodic part of the reaction, and the ability of the anodes to control might still be a function of the hydrogen liberated.

Attention was focused on the anodic reaction (oxidation of aluminum). In the field of gaseous oxidation it is well established that the oxidation rate of many metals is sensitive to relatively small additions of certain other alloying metals. The alloying metals are usually of different ionic charge but of equivalent ionic size to the base metal. It is considered that these metal ions substitute for the base metal ion in the ionic structure of the oxide, modifying its diffusion characteristics.

Tables of ionic size and ionic charge were used to select the alloying additions for aluminum. About 0.1 atomic per cent was the intended amount to be added to a base alloy of a high purity Al-0.9 w/o Ni. The results are shown in Table V. Silicon analyses (pickup from the crucible) are also shown since previous work has indicated the need for low silicon content. The figures shown in the column on corrosion behavior were obtained by testing samples for three days and two weeks. The difference in metal loss/decimeter² divided by the time (14 days minus 3 days = 11 days) for these two samples was the corrosion rate for this 11-day period. The 3-day time period was chosen because a large part of the initial rapid corrosion is over by this time.

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TABLE V

Effect of Alloying Additions on the Corrosion of an Aluminum-0.9 w/o Nickel Alloy in Distilled 350°C Water

Additive	Analyzed Concentration of Additive and Silicon (w/o)	Corrosion Behavior (Rate in mdd for period 3-14 days)
None	0.009 Si	2.4 (Surface blistering at 14 days)
Be	0.01 Be-0.09 Si	Disintegrated
Mg	0.074 Mg-0.03 Si	Disintegrated
Cr	0.20 Cr-0.09 Si	Disintegrated
Fe	0.19 Fe-0.05 Si	2.3
Ti	0.15 Ti-0.07 Si	1.6
B	0.03 B-0.01 Si	Disintegrated
2S Al-0.9 w/o Ni Alloy		2.3

It is unfortunate that the silicon pickup was high, as it precludes a clear-cut interpretation of the results. High purity aluminum crucibles have been ordered to reduce the silicon pickup. In the case of boron, however, where the silicon pickup was low, it would appear that boron additions are detrimental.

Tests are now in progress to evaluate titanium as an additive. Mixed additions of iron and titanium were ineffectual in reducing the corrosion rate of the aluminum-nickel alloy.

i. Examination of Nickel-Treated Aluminum Plates from Corrosion Test in the MTR Loop (F. L. Brown, S. H. Paine, J. F. Haran)

Six nickel-treated aluminum corrosion test plates from Reactor Engineering Division Test No. MTR-W5 were examined after irradiation. Two each of 2S aluminum plates had been prepared by Kanegin nickel plating and by nickel electroplating, and two plates were specially prepared alloys of 2S Al-0.46 w/o Ni. They were tested in 260°C (500°F) water for 530 hours and had additional periods of 50 hours at 230°C (450°F) and 150 hours at 175°C (350°F). Total integrated flux was 5×10^{20} nvt. The examination showed that much of the Kanegin and electroplated nickel had

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been corroded and flaked away, exposing the underlying aluminum to corrosive attack. Corrosion of the nickel alloy, however, did not appear to be as severe as the corrosion of the aluminum plates exposed after failure of the nickel coatings, as shown in Figures 19, 20 and 21.

Figure 19. Typical Area of General Corrosive Attack on Aluminum Sheet, Nickel Plated by the Kanegin Process.

Macro 17363

7-X

2. Development of Structural, Shielding, and Control Rod Materials

a. Preparation of Rare Earth Ceramic Bodies (J. H. Handwerk)

To prepare sufficient densified $\text{Sm}_2\text{O}_3\text{-Gd}_2\text{O}_3$ powder (Lindsay Code #920) for irradiation tests, 20 pounds of the mixed oxides were isostatically pressed into cylinders under a pressure of 50,000 psi. The cylinders were fired in an air atmosphere to a temperature of 1200°C . The apparent density of the compacted specimens was 6.5 g/cc. The fired material was then crushed to pass a 50-mesh screen, and the bulk density of the calcined powder was 4.8 g/cc. The powder was sent to J. H. Monaweck (Reactor Engineering Division) for irradiation tests.

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Figure 20. Area of General Corrosive Attack on Aluminum Plate,
Nickel Plated Electrolytically.

Macro 17368

7-X

Figure 21. Typical Corrosive Attack on Aluminum-0.46 w/o Nickel Alloy Plate.

Macro 17373

7-X

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- b. Direct Extrusion of Stainless Steel, Boron, and Rare Earth Oxides Powder Mixtures (F. D. McCuaig, R. A. Noland, J. H. Handwerk)

AISI Type 302 stainless steel specimens containing boron or mixed rare earth oxides have been produced by direct hot extrusion of powder mixtures. High purity metallic boron powder was used as the boron source in the stainless steel-boron materials. Lindsay #920,¹ a mixture containing approximately 50 w/o samarium oxide, 25 w/o gadolinium oxide, plus other rare earth oxides, was used in the stainless steel-rare earth series.

Alloys containing 3 and 10 w/o of either boron or mixed rare earth oxides in Type 302 stainless steel were attempted. Extrusions of approximately full theoretical density were obtained with all compositions except those containing 10 w/o boron; the latter were extremely hot short and broke up upon leaving the die during extrusion.

Sections of the 3 w/o boron, and 3 and 10 w/o mixed rare earth oxide extrusions were examined metallographically: the boron or oxide additions were observed to be fairly uniformly distributed throughout the stainless steel matrix in all cases. A few exploratory tests of these as-extruded sections in distilled water at 350°C for periods of up to 120 hours indicated that they have promising resistance to corrosion.

3. Nondestructive Testing

- a. Gamma-Ray Transmission Gauge for Measuring Segregation and Core Thickness in Enriched Fuel Plates (W. J. McGonnagle, N. Beck)

For accountability purposes and for avoidance of overheating due to high concentration of fuel in small areas, it is desirable to measure the amount of enriched fuel material present per unit area. By scanning the plates with a 0.25" diameter beam of gamma rays from Tm¹⁷⁰, concentration variations in the order of 0.05 mg/cm² have been detected.

The thulium source was encased in its lead carrying container. A collimating hole of 0.25" in diameter in lead produced a beam of approximately 5 r/hr at about 6" from the source. The phototube was a Dumont 6292 operated from 960 - 1050 volts. The scintillation material was NaI (thallium activated) in cylindrical form, 3/4" in diameter and 3/4" in length; it had been sealed in aluminum and glass by Harshaw Chemical Co. and was coupled to the phototube by glass and high viscosity silicone.

¹Lindsay Light and Chemical Co., West Chicago, Illinois.

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A direct measurement of the phototube collector current was made using an RCA ultrasensitive vacuum tube microammeter. It was used on a 1-microamp range for which its load on the collector current was 500,000 ohms. The microammeter was modified by expanding the zero set control so that most of the DC component could be balanced out.

b. Ultrasonic Bond Testing of Borax-III Fuel Plates
(W. J. McGonnagle, N. Beck)

A total of 2709 fuel plates were bond tested by ultrasonics utilizing a through-transmission technique. Of these, 2706 were accepted, and three were rejected due to lack of bond and because of cracks in the cladding layer.

The first 51 plates received were carefully examined in their entirety in order to determine the consistency of the manufacturing process. The remainder of the plates were tested for lack of bond on all edges, as well as along a 3/16" wide strip through the middle of the plate. During the test program several plates were selected at random and subjected to the complete test to substantiate the over-all integrity of the bond.

All of the items tested were identified by an etched number located at one end of the plate. Commencing with plate No. 998, a suffix number, i.e., 1, 2, 3, or 4, was added on all subsequent plates.

It was observed that plates 836 through 846 indicated very weak bonding; however, there was no evidence of complete lack of bond.

The surface characteristics of the first 970 plates were smooth and relatively free from inclusions and scratches. It was observed, however, that the prevalence of surface defects increased as the program progressed.

Approximately 15% of the plates were characterized by undulations which were apparently introduced in the manufacturing process. This defect, however, was not of a magnitude which necessitated straightening the plate before the test could be applied.

Fifty per cent of the plates between the numbers 61 and 1197 had a decreased bond strength at the plate end which was marked with the number. From Nos. 1200 to 3200, 15% of the plates indicated a decrease in bond strength at the end opposite the number. From plates Nos. 4000 to 7600 the characteristics appeared to be random.

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On August 16, 112 plates bearing a suffix number of 5 were examined. Of these, 53 elements were found to have areas in the core indicating a gross change in the bond strength and/or core density. Tensile tests were conducted on one of the specimens and it was found that the core-to-clad bond strength was adequate. Low-intensity radiography, however, revealed areas of nonhomogeneity in the core material. By means of beta-adsorption techniques it was possible to determine the change in the uranium content of the core material. In one plate tested, a deviation in the core density equivalent to 2% was observed.

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IV. PRODUCTION, TREATMENT, AND PROPERTIES OF MATERIALS - ACTIVITY 5410-01

1. Preparation of Single Crystals for Elastic Constants Studies (E. S. Fisher, L. T. Lloyd)

The difficulties encountered in attempting to prepare 1-cm diameter single crystals of alpha uranium have been described in previous Quarterly Reports. Evidence that these difficulties arise from a cross-sectional gradient in grain size prior to grain coarsening was described in ANL-5465. The relatively large primary recrystallized grain size at the center of the cross sections of the specimens was attributed to differences in degree of work hardening caused by the rolling process prior to recrystallization; the center of the rod is work hardened to a lesser degree than is the rod surface. This tendency to produce work hardening differences apparently increases with increase in the diameter of the rod and with decrease in the amounts of reduction of area per step in the multiple-step fabrication schedule. By using large reductions of area in all steps, the gradual gradient in grain size was essentially eliminated; however, due to resulting decrease in the total number of reduction steps, the abrupt changes in grain size, or banding, was considerably increased, so that coarsening resulted in long, narrow grains.

Experiments were then made to simultaneously minimize the banding and the gradual grain size gradient by retaining the original 5-step schedule, but varying the reductions in the first three steps only. In the original schedule the rods were reduced 10 to 15% in area in the first step, and 15 to 20% in the second step. Schedules were tried in which a 20% first step was followed by various percentages of reduction in the second step. The 20% first step appears to significantly decrease the differences in primary recrystallized grain size between the rod axis and the periphery, and also decreases the tendency for grain coarsening to start at the periphery. A 20% reduction in the second step did not result in excessive banding. The minimum degree of grain size nonuniformity was produced from 3/4" diameter rods by a schedule in which the first three reduction steps were 20 to 22%, then followed by 30 and 42% reduction steps. Four rods swaged by this schedule all appeared to contain a relatively uniform distribution of primary recrystallized grains.

The temperature gradient annealing method described in the last Quarterly Report was explored further. A large silver heat sink was heated by a furnace in a bell jar. The tip of a pencil-shaped specimen was inserted into a hole in the silver while the opposite end of the specimen was in a water-cooled copper coil. Temperature control and heat transfer conditions were developed so that a temperature difference of 100° C between the specimen point and a thermocouple located 1/8" above this point could be

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maintained when the specimen point was at 650° C. Four specimens prepared by the older reduction schedule were then annealed by this method to produce coarse grains at the specimen tip and were then inserted into furnaces for full length anneals at 650° C. Although this method was effective in forming the first coarse grains at the specimen tips, because of the matrix grain size gradient (small grains near the rod surface and large grains at the rod axis) other coarse grains formed during the second anneal which prevented the first formed coarse grains from growing to sizes suitable to be isolated for single crystals.

2. Recrystallization Textures of Deformed Alpha Uranium Single Crystals (L. T. Lloyd, M. D. Odie)

A recrystallization texture has been determined for crystal V which was deformed approximately 8% at room temperature by compression in a direction 70° from the [100], 49.5° from the [010], and 47.5° from the [001]. The operative deformation mechanisms in this crystal were twinning on {172} and {130} planes, and slip on the (010) plane. Partial recrystallization of the specimen was brought about by an anneal of one hour at 650° C. Seven different Laue patterns were obtained from the crystal in this condition; one of these served to determine the orientation of the unrecrystallized parent.

The stereographic solutions of the Laue patterns were analyzed similarly to those reported previously (ANL-5382, -5423, -5465). The recrystallized grains were assumed to have nucleated from the three deformed orientations indicated above; two grains were found to nucleate from the original orientation, three grains were found to nucleate from {172} twins, and one grain was found to nucleate from {130} twin orientation.

Figure 22 illustrates the grouping of the principal poles of the recrystallized grains with respect to the original orientation of crystal V in (001) standard projection. The "centers of gravity" may be brought into coincidence with their respective principal poles by a rotation of 34.5° about an axis which is approximately equal to the <322> direction. The magnitude of rotation is in fair agreement with those reported previously; the axis of rotation is displaced several degrees from the other axes which have been observed.

3. The High Uranium End of the Uranium-Zirconium Phase Diagram (S. T. Ziegler, L. T. Lloyd)

From microstructural observations it was tentatively concluded in ANL-5439 that the solubility of zirconium in beta uranium is between 0.2 and 0.4 w/o Zr at 750°, 700°, and 675° C. Further microstructural studies have been made of the same alloys (0.09, 0.21, 0.41, 0.65, and 1.07 w/o Zr) to more clearly define the beta-phase region. Individual

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Figure 22. Crystal V Recrystallized Grain Principal Poles Grouped with Respect to Those of the Original Orientation.

The "centers of gravity" (X) may be rotated into the original material principal poles by a rotation of 34.5° about an axis of identity approximately $\langle 322 \rangle$.

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samples from each alloy were homogenized for 2 hours at 1000° C, water quenched, cold worked, (10% reduction in thickness) and annealed 2 weeks at 670°, 690°, 710°, and 725° C, followed by water quenching. The microstructures of the alloys after the last quench are discussed below.

Alloys containing 0.09 and 0.21 w/o Zr, which were quenched from the above temperatures, were single phase (alpha uranium transformed from beta) and contained a small number of angular impurity inclusions, as illustrated in the microstructures of the U-0.21 w/o Zr alloy quenched from 670° C (Figure 23-a); this agrees with the results reported in ANL-5439 in that the solubility of zirconium in beta uranium is greater than 0.2 w/o Zr. The higher zirconium alloys (0.41, 0.65, and 1.07 w/o Zr) contained two phases, as shown in Figures 23-b and 23-f. The second phase in these alloys quenched from 670°, 690°, and 710° C appears as fine spheroids which, for a constant temperature of treatment prior to final quenching, increased progressively in amount with increasing zirconium composition. Comparatively, the amounts of second phase in alloys quenched from 670° C were greater than in alloys quenched from 690° and 710° C, indicating a greater solubility of zirconium in beta uranium at 690° and 710° C than at 670° C.

The second phase in the series of samples quenched from 725° C appeared in two forms. The 0.41 and 0.65 w/o Zr alloys showed resolvable spheroids similar to those observed in the 0.41, 0.65, and 1.07 w/o Zr alloys which were quenched from 670°, 690°, and 710° C; in the 1.07 w/o Zr alloy, however, the second phase appeared as large grains of acicular product similar to that observed and reported for alloys containing 0.41, 0.65, and 1.07 w/o Zr which were quenched from 750° C (see Figure 24). Both of these types of second phase materials are believed to be related to the gamma phase present at the temperature of heat treatment; in the case of the small spheroidal type the acicular structure apparently cannot be resolved by optical means.

Some justification for the assumption that these two forms of the second phase pertain to the same high temperature phase can be obtained by a consideration of the relative amounts of the phases which should be present in the various alloys from phase diagram considerations. A microscopic quantitative determination of the volume per cent gamma present in equilibrium with beta at 750° C in the 0.41, 0.65, and 1.07 w/o Zr alloys yielded values of 19, 45, and 63 v/o, respectively. A similar determination of the volume per cent gamma present at 725° C in the 1.07 w/o Zr alloy indicated a value of 23 v/o. Calculations for the 0.41 and 0.65 w/o alloys, based on the phase diagram shown in Figure 24, yielded values of only 1.5 and 7.5 v/o gamma present at 725° C, assuming the densities of beta and gamma to be equal. At these low concentrations the gamma phase is apparently present in too small a particle size to permit the microscopic detection of the acicular nature of its transformation product

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Figure 23. Microstructures of U-Zr Alloys, Homogenized, Cold Worked, Heat Treated Two Weeks at Temperatures Shown, and Water Quenched. All specimens were electroetched in phosphoric acid.

23-a. U-0.21 w/o Zr (670°C)

23-b. U-0.41 w/o Zr (670°C)

Micro 17798 Bright Field 500-X

Micro 17800 Bright Field 500-X

23-c. U-0.41 w/o Zr (690°C)

23-d. U-0.65 w/o Zr (690°C)

Micro 17534 Bright Field 500-X

Micro 17535 Bright Field 500-X

23-e. U-0.41 w/o Zr (710°C)

23-f. U-0.65 w/o Zr (710°C)

Micro 17477 Bright Field 500-X

Micro 17478 Bright Field 500-X

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Figure 24. Tentative Diagram of the High Uranium End of the Uranium-Zirconium System.

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As a check to determine if phase equilibrium had been obtained in 0.41, 0.65, and 1.07 w/o Zr alloys which had been heat treated for 2 weeks at 775°, 750°, and 700° C, the samples were again cold worked and reheat treated for an additional week at their respective temperatures. The microstructures of the series of alloys quenched from 775° and 750° C showed no change; however, the series of alloys quenched from 700° C showed a slight coalescence of the second phase, as is illustrated in Figures 25-a and -b. Apparently 2 weeks at temperatures of 775° and 750° C is sufficient to obtain phase equilibrium; however, an additional week was necessary to bring about more complete equilibrium at 700° C.

The above observations and the results reported in ANL-5439 for the uranium rich end of the uranium-zirconium phase diagram are summarized in Figure 24. The solubility of zirconium in beta uranium is at a maximum (slightly less than 0.41 w/o Zr) between 690° and 710° C and is greater than 0.2 w/o Zr at all temperatures above 670° C and below 750° C. In Figure 24 the transformation temperature horizontals are those reported by Summers-Smith² from dilatometric data. Thermal analyses of fair accuracy have been made of the U-1.07 w/o Zr alloy; the results agree closely with the data of Summers-Smith.

4. Uranium Corner of the Uranium-Zirconium-Niobium Phase Diagram (A. E. Dwight, L. T. Lloyd)

The investigation of the uranium-niobium equilibrium phase diagram has been extended to 10 w/o Nb. The eutectoid transformation occurs at $634 \pm 2^\circ \text{C}$, and its composition is estimated at 8 ± 1 w/o Nb; characteristic lamellar structures are produced by this reaction.

Metallographic studies of ternary alloys have determined the curved surface which bounds the lower limit of the single-phase gamma field (ANL-5465), but failed to delineate the lower surface of the two-phase region immediately below the gamma field. An alloy containing nominally 8 w/o Zr and 2 w/o Nb has been investigated in an attempt to locate this transformation by electrical resistivity measurements. Resistivity values were calculated from the measured potential drop and current flow at room temperature of cylindrical specimens which had been homogenized at 800° C, transformed for 24 hours by interrupted quench to temperatures between 600° and 700° C, followed by water quenching; Figure 26 shows the curve of resistivity versus interrupted quenching temperature. The horizontal line at 676° C was previously determined metallographically to be the lower limit of the single-phase gamma for the alloy of this composition.

²D. Summers-Smith, "The Constitution of Uranium-Zirconium Alloys," Journal of the Institute of Metals, 83, 277-282 (1954-1955).

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Figure 25. Microstructure of U-0.41 w/o Zr Alloy, Cold Worked and Heat Treated as Shown at 700°C. Specimens electroetched in phosphoric acid.

25-a. 2 weeks at 700°C, W.Q.

25-b. Same as 25-a, cold worked and re-annealed 1 week at 700°C, W.Q.

Micro 16982

Bright Field

500-X

Micro 17782

Bright Field

500-X

Figure 26. Resistivity Versus Transformation Temperature Curve of a U-8 w/o Zr-2 w/o Nb Alloy, Transformed for 24 Hours by Interrupted Quench from 800°C at the Temperature Indicated, Followed by a Water Quench. The resistivity was measured at room temperature.

Macro 19024

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The microstructures at room temperature which result from the interrupted quench transformation between 676° and 600°C appear as two phases: one, the martensitically transformed gamma which was stable at the transformation temperature; and two, alpha platelets which become finer as the transformation temperature is lowered. The resistivity curve is interpreted as indicating that the martensitic product has a higher resistivity than that of the alpha. The fall in resistivity between 676° and 630°C indicates an increasing amount of alpha; the nearly vertical line below 630°C indicates that gamma no longer exists as a stable phase at temperature. Attempts to confirm these conclusions by X-ray diffraction studies are inconclusive because of the strong alpha patterns which are obtained.

5. Corrosion Mechanism of Aluminum Alloys at Elevated Temperatures (W. E. Ruther, C. A. Youngdahl)

a. Metallurgical Factors

A chill cast and rolled alloy of Al-1.1 w/o Fe-0.5 w/o Ni-0.3 w/o Si was found to exhibit different corrosion behavior on the opposite sides of the same $1/8"$ thick sample when tested in 350°C distilled water. After removing the oxide, the corroded sample was examined metallographically using a modified Keller's etch. The side having the finer dispersion of the second phase corroded in a smooth and uniform fashion with an adherent oxide coating; the coarse dispersion side showed serious roughness and lack of uniformity upon corrosion (see Figure 27).

The investigation of the influence of second phase dispersion on corrosion behavior was extended to a series of aluminum-nickel-iron alloys. Corrosion tests had indicated that an Al-0.5 w/o Ni alloy required a minimum of 0.3 w/o Fe to be resistant to 350°C water after rolling. Samples of the various melts were examined in the as-chill cast condition. Specimens were chosen from the same relative spot on the casting to reduce possible variations due to differences in the rapidity of the chill. As shown in Figure 28, the aluminum-iron compound precipitates at random locations throughout the grains, with small concentrations at grain boundaries. Very little of the aluminum-nickel compound shows within the grains; it is largely at grain boundaries. These characteristics seem to be at least partially retained in the ternary alloys, as shown in Figure 29. The alloy containing 0.5 w/o Ni-0.3 w/o Fe was corrosion resistant while the 0.5 w/o Ni-0.2 w/o Fe alloy was just on the border line. The increase in quantity and the slight improvement in the dispersion of the second phase as shown in the photograph are assumed to be responsible for this different corrosion behavior.

The photomicrographs also afford an explanation for the role of iron in the aluminum-nickel alloy. The "hydrogen blister" theory of the corrosion disintegration of aluminum assumes that hydrogen penetration and subsequent blistering occur wherever aluminum becomes a corrosion cathode. The nickel and iron both form compounds which are

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Figure 27. Effect of Second Phase Dispersion on the Corrosion of Al-1.1 w/o Fe-0.5 w/o Ni-0.3 w/o Si Alloy in Distilled 350°C Water.

Cross-section photomicrographs of opposite sides of a 0.125" thick rolled sample.

27-a. Poor Corrosion Behavior.

Micro 17612

250-X

27-b. Better Corrosion Behavior.

Micro 17610

250-X

Figure 28. Distribution of the Second Phase in Aluminum-Iron and Aluminum-Nickel Alloys, As Cast.

28-a. 0.2% Iron in Aluminum.

Micro 17856

250-X

28-b. 0.5% Nickel in Aluminum.

Micro 18679

250-X

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Figure 29. Distribution of the Second Phase in Ternary Aluminum-Nickel-Iron Alloys, As Cast

29-a. 0.5% Ni-0.2% Fe

Micro 18680

250-X

Borderline Corrosion Resistance.
(350°C)

29-b. 0.5% Ni-0.3% Fe

Micro 18681

250-X

Good Corrosion Resistance.
(350°C)

cathodic to the aluminum grains, moving the corrosion cathodes away from the aluminum. The corrosion hydrogen formed on these compounds does not penetrate the aluminum metal but is presumably combined to form molecular hydrogen which diffuses away into the solution. However, if the geometry of the dispersion of this cathodic second phase permits comparatively large open areas, the aluminum metal in the centers of these areas may successfully compete in the cathodic corrosion reaction and form destructive cathodes. As is shown in Figure 28, aluminum-nickel compound alone produces only a somewhat favorable geometry, but combined with the random deposits of the aluminum-iron compound (Figure 29) the distances between the second phase cathodic sites are reduced considerably.

The dispersion factors may also be an important consideration for the undesirability of silicon as an alloying element. Silicon goes into solid solution in the aluminum at low concentrations and may serve to make the aluminum grain potentially a better cathode, thus requiring smaller distances between the cathodic Al-Fe and/or Al-Ni compound particles for successful protection. In excess of the solid solubility, it may be expected that silicon-aluminum compound would play the role of a cathodic second phase; electrical measurements, however, indicate that this compound is not well suited for this role.

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b. Hydrogen Content in 2S Aluminum by the Vacuum Fusion Method

Previous hydrogen analyses, reported in the March, 1955, Quarterly Report (ANL-5439), were found to be erroneous. The use of a Vycor thimble for containing the molten aluminum was a poor choice of material. Subsequent experiments have shown that the reaction between the Vycor and the aluminum yields hydrogen in sufficient quantities to overshadow the original hydrogen content of the aluminum. A thimble of alundum was substituted for the Vycor. Tests in which the same sample is run twice in a row indicate very little hydrogen evolved from the alundum-aluminum reaction.

However, the analysis still presents difficulties. The "leak rate" with the thimble hot but with no sample is on the order of one micron per hour in a 215-cc volume. When the sample is placed in the hot thimble, fairly rapid evolution of hydrogen takes place for the first hour. At the end of this time hydrogen accumulates at a rate about three times the normal "leak rate." Tests have been run for 17 hours and the accumulation is still going on. In order to provide a basis for comparison, the one-hour values are reported although the significance of the continuous accumulation of hydrogen is unknown.

A piece of 2S aluminum plate, 1/8" thick, was cut into two samples. One was placed on test in an autoclave at 290° C for 65 hours in distilled water. The other was retained for analysis. The corroded sample was just beginning to show small blisters. The oxide and blisters were ground off and specimens 3/8" wide were cut for hydrogen analyses. The results are shown in Table VI. The increase in hydrogen content of the corroded metal is in line with the theory previously discussed.

TABLE VI

Hydrogen Analyses of 2S Aluminum Sheet

Conditions	Hydrogen (ppm)
No corrosion	0.4
	0.4
	0.2
65 hours at 290° C., distilled water	1.3
	1.1
	1.3
	2.2

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A similar analysis was made for a sample of M388 alloy (the designation of the Aluminum Company of America for the 0.9 w/o Ni in 2S aluminum alloy) tested in distilled water for 57 days at 350° C. Comparison of the corroded metal and the parent stock is made in Table VII.

TABLE VII

Hydrogen Analyses of M388 Alloy

Conditions	Hydrogen (ppm)
No corrosion	0.4 0.5 (average)
57 days at 350° C, distilled water	0.4 0.6 0.9

Penetration of the metal by hydrogen does not appear to be a significant part of the corrosion process for this alloy, in corroboration of the theory explaining the function of the nickel.

c. Electrochemical Measurements at 200° C

Open-circuit solution potentials and cathodic polarization curves were measured for the second phase present in aluminum after alloying with elements shown in Table VIII. Since the phase diagrams are eutectics for these systems in the high aluminum region, the second phase would normally have the eutectic compositions. All except the copper alloy were rolled into 1/8" thick sheets before testing. The copper alloy was too brittle to roll and was used in the as cast condition.

As a first stage in the experiment, the alloys were corrosion tested for one week at 200° C in distilled water. It was assumed that further changes in the oxide film would be relatively slow. One alloy at a time was then suspended in a special autoclave permitting electrical access to the specimen. The open circuit solution potential was recorded after a short time at temperature. A cathodic polarization curve (E vs. I) was then run using an interrupter technique and a recording peak voltmeter developed by this group. A constant rate of current increase was used for all runs. The values obtained are presented in Table VIII.

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TABLE VIII

Potential Measurements in 200° C Distilled Water

Eutectic Composition	Open Circuit Solution Potential, E* (volts)	Potential Change at 15 $\mu\text{a}/\text{cm}^2$, ΔE cathodic (volts)
Al-5.7 w/o Ni	-0.44	-0.12
Al-33 w/o Cu	-0.62	-0.21
Al-1.7 w/o Fe	-0.77	-0.24
Al-1 w/o Co	-0.66	-0.26
Al-11.7 w/o Si	-0.88	-0.34
2S Aluminum	-0.60	-1.2

*Reference electrode is corroded stainless steel.

The table is arranged in order of increasing ΔE at 15 $\mu\text{a}/\text{cm}^2$. This is a measure of the ability of the alloy to pass cathodic current easily. The lower the ΔE , the better the cathode. The open circuit potential is not as reliable since it is measured with respect to stainless steel. Roughly, however, only the nickel alloy caused a significant shift in solution potential in the cathodic direction, such as might be expected from the addition of an actively cathodic phase without alteration of the base metal phase.

Previous corrosion tests with other alloys of these elements indicate that roughly the same order as that in the table describes the relative ability to prevent blister attack on aluminum; with nickel the best, and silicon the worst of these additive elements. This is another piece of corroborative evidence for the "hydrogen blister" theory of aluminum disintegration.

6. Mechanism of Aqueous Corrosion of Zirconium (R. D. Misch, F. H. Gunzel)

The possibility of using chemically deposited silver as a contact surface for measuring the electronic conductivity of the oxide film on zirconium has been investigated. This method has now been discarded since erratic results were obtained. Conduction was observed at room temperature in about half the cases (perhaps attributable to the presence of non-insulating inclusions). The conduction, upon heating the remaining silver spots, was not reproducible.

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The "constriction resistance" of the standard probe was measured while resting on a sheet of smooth platinum. From this value the area of contact made between the probe and a flat sample was found to be 1 to $2 \times 10^{-6} \text{ cm}^2$. A knowledge of this area should be useful in interpreting the results of electrical conductivity measurements made on surface oxides.

Samples having oxide films produced by corrosion in distilled water at 350°C were held in vacuum at 450°C and the oxide conductivity was measured. In all cases the conductivity increased with time until the readings were off scale for the instruments used. The results apparently indicate that changes occur in the oxide at this temperature. Further work will be done in an effort to establish the nature of this change, since it may be closely related to the kind of conductivity exhibited by the oxide during corrosion.

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V. EFFECTS OF IRRADIATION ON MATERIALS - ACTIVITY 5430-01

1. Irradiation Cycling of Natural Uranium in the Alpha and Alpha Plus Beta Temperature Ranges (J. H. Kittel, S. H. Paine)

A group of natural uranium specimens was irradiated in the MTR in NaK-filled capsules under conditions such that the central metal of some of the specimens transformed to the beta phase. The specimens were short cylinders notched for fracturing after irradiation, as shown in Figure 30. Some of the specimens were cycled into and out of the reactor for 50 times, each cycle consisting of 15 minutes under irradiation and 5 minutes of cooling in the canal. Transfer times are estimated to be on the order of a few seconds. The remaining specimens were irradiated continuously for 12-1/2 hours, the same total length of irradiation received by the cycled specimens. All specimens were irradiated consecutively in the same reactor location so that they would receive approximately the same flux density. Cobalt monitors attached to each capsule showed that each had been subjected to very nearly the same integrated thermal flux.

It was intended that some of the specimens be irradiated in the high alpha temperature range and that others be irradiated under conditions where the center was above the alpha phase. The thermal flux, enrichment, and means of heat removal were treated as constants in this experiment, and the desired temperature distributions were obtained by varying the specimen diameters. The proper diameters were determined by making a preliminary irradiation of fine grained material, fracturing the specimens after irradiation, and observing the appearance of the fractured surfaces. Fine grained uranium does not coarsen rapidly in the alpha phase, but after transformation to the beta phase shows pronounced grain growth. It was found that a specimen 0.56" in diameter remained in the alpha phase, as shown in Figure 31, while the center of a specimen 0.60" in diameter had passed through the alpha-beta transformation, as shown in Figure 32. It is not known definitely whether the extreme center also transformed to the gamma phase, but extrapolation of calculated curves³ indicates that this is unlikely.

After the preliminary survey, a number of specimens having appropriate diameters were irradiated and examined. Table IX lists their specifications, the conditions under which they were irradiated, and also gives a summary of the changes observed. It can be noted in the 300°C rolled material that the specimen which did not exceed the alpha-beta transformation temperature had a slightly higher growth rate than the specimen which did transform in the central region.

³V. O. Ericksen, "Temperature Distribution and Heat Stresses Due to Thermal Neutrons in a Cylindrical Uranium Rod," TT-533, (1954).

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Figure 30. Appearance of a Typical Specimen Before Irradiation.
The specimen has been notched for postirradiation fracturing.

Macro 18620

2-X

Figure 31. Fractured Cross Section of 0.56" Diameter Specimen.
Entire specimen remained in the alpha phase during irradiation.

Macro 17434

4-X

Figure 32. Fractured Cross Section of 0.60" Diameter Specimen.
Center metal transformed to the beta phase during irradiation.

Macro 17432

4-X

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TABLE IX

Effects of Alpha and Alpha Plus Beta Irradiation

Specimen No.	Fabrication and Heat Treatment	Diameter (inch)	Type of Irradiation	Irradiation Temperature Range	Burnup (a/o)	Length Change (%)	Growth Rate, microinches/inch/cycle	Diameter Change (%)	Remarks
AC-22	300°C rolled	0.52	50 cycles	alpha	0.0040	5.0	1000	-2.7	Center showed pronounced grain growth.
AC-26	300°C rolled	0.60	50 cycles	alpha + beta	0.0040	3.8	760	-1.7	
AC-61	300°C rolled, beta quenched	0.52	50 cycles	alpha	0.0040	-0.3	-64	-0.1	
AC-65	300°C rolled, beta quenched	0.60	50 cycles	alpha + beta	0.0040	-	-	-	Developed a central void and could not be removed from irradiation capsule.
AC-62	300°C rolled, beta quenched	0.52	continuous	alpha	0.0035	0.0	-	-0.1	
AC-64	300°C rolled, beta quenched	0.56	continuous	alpha	0.0028	0.3	-	0.0	
AC-66	300°C rolled, beta quenched	0.60	continuous	alpha + beta	0.0038	0.9	-	0.0	Extruded central material at one end.

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In the cycled beta-quenched material this comparison could not be made because the specimen which was cycled partially into the beta phase temperature range could not be removed from its capsule for measurement. An end view of this specimen is shown in Figure 33 and a section through the center is shown in Figure 34. The internal void which formed in this specimen is similar to those produced by Hayward⁴ who thermally cycled uranium specimens 50 times from 20° to 680°C. His specimens, however, decreased in diameter, whereas the present specimen increased in diameter. It is not known why this specimen developed a void while the 300°C rolled specimen of the same size showed no such effect. Possibly, the higher mechanical properties of the latter specimen enabled it to resist to a greater degree this type of deformation. The beta-quenched cycled specimen which remained in the alpha phase temperature range shortened slightly.

Continuous irradiation of beta quenched material in both the alpha and the alpha plus beta temperature regions showed no marked differences in over-all growth rate. However, specimen AC-66, the center of which exceeded the alpha to beta transformation temperature during irradiation, extruded material from the center of one end, as shown in Figure 35. A stereoscopic view of this end is shown in Figure 36, and it can be seen that a depression was formed immediately around the extruded material. A fracture of this specimen showed no internal void formation.

Based on these preliminary irradiations, it appears that solid, unalloyed uranium could give rise to difficult operational problems if used as reactor fuel where it was partially above the alpha-beta transformation temperature. This would be particularly true if cycling conditions existed.

⁴B. R. Hayward, "Dimensional Changes Resulting From Alpha-Beta Thermal Cycling of Uranium," Proceedings, Metallurgy Information Meeting, April 19 - 21, 1954, TID-5185, Part 1, p. 183.

Figure 33. End of Specimen AC-65 After 50 Irradiation Cycles.
The specimen is still in its irradiation capsule.

Macro 18626

2-X

Figure 34. Section Through Above Specimen, Showing Central Void
Which Developed During Irradiation Cycling.

Macro 18625

2-X

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Figure 35. Specimen AC-66 After Continuous Irradiation in the Alpha-Beta Temperature Range.

Macro 18628

2-X

Figure 36. Stereoscopic End View of Above Specimen.

Macro 18849

9-X

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2. Neutron Flux Distribution and Wilkins Effect in Irradiation Specimens (F. R. Taraba, S. H. Paine)

The distribution of cumulative damage effects and of temperatures within specimens of fuel alloys under irradiation is dependent upon the internal distribution of neutrons during irradiation. This is specially true of materials containing added fissionable atoms in which it is desired to achieve rapid burnups at elevated temperatures. In the interest of understanding the neutron flux distribution in irradiation specimens, a series of modified Wilkins experiments are in progress, from which it is intended to obtain information permitting good computation of the flux pattern under a wide range of experimental conditions.

In the experimental procedure, thin uranium foils are sandwiched between uranium wafers which are then assembled into a "solid" rod for activation in the CP-5 reactor. A schematic drawing of such an assembly is shown in Figure 37. The distribution of fission product activity is obtained by means of very small samples (500 micrograms) punched from the foils after irradiation. A gamma-ray scintillation spectrometer is used to count the activity. It is assumed that the amount of fission products produced in any region of the rod, and hence the radioactivity of the material in this region, is proportional to the thermal neutron flux at this position. The contribution of fast neutrons to the production of fission products is quite small and may be neglected.

The "Wilkins Effect" may be defined as the increase in the neutron flux at the ends of rods separated by a nonabsorbing spacer (e.g., graphite, aluminum, etc.). It may be evaluated as the ratio of the average flux at the ends (next to the spacer) to the average flux at a location remote from the ends. A considerable amount of data has been accumulated on the neutron flux distribution in 1" and in 1/2" diameter uranium rods and a statistical analysis of the data is in progress. A few preliminary results are presented in Table X.

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Figure 37. Schematic Drawing of Assembly for Neutron Flux Distribution Measurements.

TABLE X

Flux Distribution in 1" and in 1/2" Cylindrical Rods
of Normal Uranium

Diameter of Rod (inch)	Graphite Spacer (inch)	Wilkins Effect(a)	Ratio of Surface to Axial Neutron Flux	
			Removed from Spacer	Adjacent to Spacer
1.000	0	1.00	1.38	1.38
	0.150	1.12	1.38	1.22
	0.300	1.14	1.38	1.21
	very large	1.33	1.38	1.15
0.500	0	1.00	1.13	1.13
	0.050	1.03	1.13	1.08
	0.100	1.05	1.13	1.08
	very large	1.12	1.13	1.04

(a) Ratio of average flux at A and B (see Figure 37).

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