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TEXTURE AND IRRADIATION GROWTH IN EBR-II DRIVER FUEL

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ABSTRACT: Preferred-orientation studies of centrifugally bonded EBR-II fuel pins showed that the pins have a [110], [001] axial texture, which is responsible for their diametrical growth and shortening in length during irradiation. The growth indices calculated from the texture data correlate well with the observed diametrical irradiation growth rate of the pins and give a single crystal growth rate, g , of 430 percent per percent burnup, which is in the range measured for uranium and uranium alloys.

The texture is shown to be caused by compressive stresses from centrifuging during the $\gamma \rightarrow \alpha$ transformation of the fuel pin at 500°C. Atomistic mechanisms for the texture formation produced by the compressive stress are discussed.

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1. INTRODUCTION

The Experimental Breeder Reactor II (EBR-II) Mark IA driver fuel element is composed of an injection cast 0.366-cm-diameter by 34.3-cm-long uranium-5 wt% fission alloy pin. Fission is an equilibrium concentration of fission product elements left by the pyrometallurgical reprocessing cycle designed for EBR-II. The 5 wt % fission includes 2.44 wt% molybdenum, 1.94 wt% ruthenium, 0.28 wt% rhodium, 0.19 wt% palladium, 0.085 wt% zirconium, and 0.015 wt% niobium. The pin is sodium-bonded to a 0.442-cm-O.D. by 0.023-cm-wall annealed Type 304L stainless steel jacket. The overall length of the element is 46.2 cm. The principal phase in cast pins is metastable body-centered-cubic gamma uranium. To ensure a good thermal bond between the pin and the cladding, the element is heated to about 500°C and impacted 500 times. The heat treatment also results in transformation of retained gamma to orthorhombic alpha uranium (the principal phase present during irradiation).

Fuel elements were fabricated by this technique for several years at Argonne National Laboratory (ANL) and exhibited acceptable dimensional stability during irradiation. More recently, the fuel elements were fabricated by a technique that utilized centrifugal, rather than impact, bonding. Initial irradiation results showed that the centrifuged fuel pins shortened under irradiation. Axial shortening of up to 2.3 cm for pins initially 34.2 cm long, with

accompanying radial growth varying from zero at the pin top to about 8% at the bottom, was measured after ~ 0.6 atom % burnup.¹

Preliminary x-ray diffraction studies² detected the presence of preferred orientation (texture) in these pins with an excess of [110] (shrinkage-type) texture and a deficiency of [021] (growth-type) texture in the axial direction. The studies also showed more of this preferred orientation in the pin bottoms than in the tops. The [110] and [021] orientation are referred to as shrinkage and growth textures because neutron irradiation experiments on single crystals of alpha uranium³ have shown that growth occurs in the "b" crystallographic direction with a corresponding shrinkage in the "a" direction and no change in the "c" direction. Hence, any preferred orientation that is present in polycrystalline uranium will cause anisotropic dimensional changes during irradiation.

Preferred orientation in alpha uranium has been a concern in fuel element manufacture for many years,⁴ but has been normally associated with extruded or rolled fuel and not with cast material. Texture has been reduced or eliminated in mechanically formed uranium by heat treating and quenching from the beta phase. Quenching rates⁵ are important and can, in themselves, produce texture generally associated with internal stresses resulting from the transformation.

A careful comparison between the impact bonded and centrifugally bonded fuel element fabrication processes was made.

Both processes employed injection casting to produce the fuel pins. The pins were then loaded into stainless steel jackets with about 0.5 g each of sodium.

The primary difference in the processes arises from the methods used to produce a thermal bond between the fuel and jacket. In the impact process, the element is heated to $\approx 500^{\circ}\text{C}$ and impacted 500 times. This heating requires about one hour during which the gamma phase in the cast pins transforms to alpha prior to impacting. In the centrifuge process, the element is centrifuged during a one hour heating period to $\approx 500^{\circ}\text{C}$ and then centrifuged at temperature for ≈ 20 minutes. The gamma phase in the cast pins, therefore, transforms to alpha phase while the centrifugal force is being applied. This centrifugal force imposes an axial compressive stress on the fuel pin. Hence it was believed² that the stress during the $\gamma \rightarrow \alpha$ transformation, which varies along the pin from zero at the top to a maximum at the bottom, was responsible for the observed preferred orientation and the associated anisotropic irradiation growth.

Preferred orientation produced by an externally applied stress during a phase transformation has not been previously reported for uranium or uranium alloys. In fact, this phenomenon has apparently been detected in only a few isolated instances. Bronisz and Tate⁶ ($\beta \rightarrow \alpha$ transformation in plutonium) and Shimizu and Horiuchi⁷ (disordered \rightarrow ordered transformation in Pt - Co, Au - Cu, and Pd - Fe alloys) have studied the effects of

applied stress during transformation on texture. Their results indicate that significant textures can be produced at moderate stresses without plastic deformation.

If the centrifugal-bonding process was responsible for the fuel shortening observed during irradiation, then either impact-bonding in place of centrifugal-bonding, or a heat treatment at about 500°C to transform retained gamma to alpha prior to centrifugal bonding could be used to produce nonshortening pins. Because impact bonding does produce an axial stress in the pins, they are held at 500°C for a sufficient time prior to impacting to allow the gamma-to-alpha transformation to occur. Furthermore, centrifugally bonded fuel already containing an undesirable texture could be reclaimed by heat treating. This heat treatment would consist of heating the elements to a temperature above 642°C to transform the textured alpha uranium in the bonded pins back to the gamma phase, and then transforming the new gamma back to alpha with a second heat treatment below the $\gamma + \alpha \rightarrow \alpha$ transformation temperature of 552°C.⁸ Irradiation of fuel elements processed by the above methods gave the expected result, i.e., no shortening of any pins. In fact, the EBR-II is presently operating on reclaimed fuel that received a heat treatment at 660°C for 1 hour followed by a second heat treatment at 500°C for 1 hour without any shortening problems.

The present report describes a method developed and employed to measure texture and to correlate it with the irradiation growth of EBR-II Driver Fuel for various methods of heat treat-

ment and fabrication. The texture measurements together with studies of the microstructure and hardness were used as the basis for discussion of possible mechanisms for the formation of the texture. The texture measurements were also used to determine the basic growth coefficient, "g",³ which gives the percent growth per percent burnup referred to a single crystal of uranium.

Textures were measured by the inverse-pole-figure method⁹ because this method is rapid and lends itself readily to the correlation of texture in a particular physical direction with a particular physical property, in this case irradiation growth. In the inverse-pole-figure method, a set of X-ray diffraction intensities, I_{hkl} , is measured on a diffractometer from a flat surface of the specimen normal to the direction of interest. The diffraction intensities are used to determine the proportion of grains for each orientation of the a, b, and c crystallographic axes in the physical direction in interest.

2.0 EXPERIMENTAL AND ANALYTICAL PROCEDURES

2.1. Specimen Preparation

The x-ray diffraction measurements of texture were performed on specimens prepared by two sampling methods: a) twenty-four transverse sections of a single fuel pin were mounted to form a composite specimen representing the bottom two-thirds of the pin length, and b) axial "texture gradient" measurements were made on single transverse sections taken at various distances along the length of the pin.

The specimens were ground by standard procedures through 0.25 μm diamond abrasive and electropolished to remove worked metal. The electropolishing was performed in a solution of 200 ml ethyl alcohol, 125 ml H_3PO_4 , and 125 ml ethylene glycol. The electropolishing conditions were: stainless steel cathode, 18 volts, and 0.3 to 0.6 amps for 20 sec. Although this electropolish is a standard one for uranium, it preferentially attacks the region around the grain boundaries in uranium-fissium, so the effect of this attack on the X-ray results was studied by electropolishing one specimen more uniformly¹⁰ in a solution of one part of a mixture consisting of 118 g chromic acid (CrO_3) and 100 ml water to four parts of glacial acetic acid.

2.2. X-ray Diffraction

The specimens were scanned on a conventional North American Phillips diffractometer with a fine focus $\text{CuK}\alpha$ X-ray tube and a rotating specimen holder. An Advanced Metals Research Model 3-202 monochromator with a graphite single crystal was used on the detector side of the diffractometer. A special cup-shaped specimen holder¹¹ was used for the texture gradient measurements.

2.3. Microstructure and Hardness Measurements

Specimen surface preparation for these analyses was the same as for X-ray diffraction. Replica electron microscopy was performed on an RCA Model EMU microscope. The specimens were prepared by replicating with *Formvar** backed by *Parlodion*** and

* Registered tradename of Shawinigan Products Corp., Englewood Cliffs, N. J.

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shadowed with uranium. Scanning electron microscopy was performed on the Cambridge Stereoscan Mark IIa Microscope. Quantitative microanalysis for U, Mo, and Ru was performed on a Materials Analysis Corporation Model 400 electron probe microanalyser. Microhardness was measured on a Tukon Hardness Tester using a 100 g load and a Vickers diamond pyramid indenter.

2.4 Diameter Measurements on Irradiated Fuel Pins

Postirradiation diameters of fuel pins were measured using a Bausch and Lomb DR-25B Optical Gauge. Three diameters were taken at each axial location. Readings were made to 0.00002 cm and are considered accurate to ± 0.0002 cm. Calibrated diameter standards were employed, and the measurements were corrected for temperature. Because accurate preirradiated diameters were not available, the nominal pin diameter of 0.366 cm was used to compute $\Delta D/D_0$ values.

2.5 Measurement and Analysis of Diffraction Peak Intensities

The uranium-fission alloy gave a very complex diffraction pattern because the peaks from the predominant phase, alpha uranium, were broadened, shifted and overlapped due to lattice strain and generally contained contributions from the minor phases in the alloy, namely U_2Ru , U_2Mo , and possibly distorted gamma-phase uranium.⁸ About 70 diffraction peaks were observed. The peaks were predominately from distorted alpha uranium. However, at least 15 were from the minor phases. Only twelve alpha uranium peaks were found sufficiently separated for the preferred orientation measurements: [020], [110], [021], [111],

[112], [130], [131], [113], [133], [114], [223], and [312].

All twelve were employed for the composite specimens; however, only six were intense enough to use for the small transverse sections: [110], [021], [111], [131], and [114]. The [002] was also measured but does not contribute to growth.

The diffraction intensities were determined from planimeter measurements of the areas of the peaks on the strip chart records. Special care was exercised to resolve the [110], [021], [002] triplet before planimentering, since the [021] and [110] are the most important peaks (Figure 1) for determining the preferred orientation. The contraction of the uranium-fissium lattice in the [010] direction displaced the [021] peak towards the [002] peak, making the overlap with the [002] greater than in pure uranium. Furthermore, the [021] was always broader because of the contribution from the [002] and the [110] peaks. The [002] peak was also distorted by contributions from the group of unknown peaks between the 2θ values of 36.5° and 38.5° (Figure 1). Because there are allowed reflections in the 2θ region from U_2Ru , U_2Mo , and gamma uranium, the phase or phases that caused these unknown peaks could not be identified.

The best method found for resolving the [110], [021], [002] triplet was as follows: Start with the low angle half of the [110] peak and assume it is a perfect "half peak." Sketch this in as the high angle half of the [110], and then subtract it from

the composite [021] + [110] to obtain the low angle half of the [021] and so forth until all three peaks have been resolved. The remainder after the [002] is assumed to belong to the group of peaks between 36.5° and 38.5° 2θ. Such a graphical method may lack precision, but it was adequate for this study because of the large specimen-to-specimen variation in peak shape due to variation in grain size, chemical composition, aging condition, etc., of these fuel pins. In determining the I₁₁₀/I₀₂₁ ratio, peak heights were found to be as accurate as peak areas.

A computer program was written to determine the peak areas by curve fitting the peaks with a Gaussian distribution function. The program gave good results for cast and impact bonded fuel pins, and also for specially treated pins (double heat-treated, annealed at 500°C prior to centrifugal bonding, or impact-bonded rather than centrifugally bonded) because these have "relatively" strain free structures. However, the program gave poor results for cast and centrifugally bonded pins, because the peaks were too distorted in shape to fit a Gaussian distribution function.

The preferred orientation measurements were analyzed by the inverse-pole-figure method.¹² In this method, the orientation is described by the quantity, P_{hkl}, where

$$P_{hkl} = I_{hkl}/I_{hkl}^{\circ} \div 1/n \sum_{hkl} (I_{hkl}/I_{hkl}^{\circ})$$

where: n = number of planes, I_{hkl} is the measured diffraction intensity for the plane $[hkl]$, and I_{hkl}° is the calculated intensity for the plane $[hkl]$ in a random orientation. Thus, $P_{hkl} = 1$ for random orientation, $P_{hkl} > 1$ for an excess of a given orientation, and $P_{hkl} < 1$ for a deficiency of a given orientation. Calculated "random" intensities¹³ were used for I_{hkl}° for the composite specimens. Values for I_{hkl}° could not be calculated for single transverse sections because the X-ray beam area was larger than the specimen, so a "random" standard was prepared from pin No. 12367, which was injection-cast, heat-treated at 660°C for 1½ hours, and impact-bonded. The measured standard intensities compared reasonably well with previous measurements¹¹ (Table I) on a cross section of 0.3175-cm-diameter rod machined from an unalloyed uranium plate which was prepared by powder metallurgical methods. One should expect some differences between uranium and uranium-fissium because of the "strained" state of the latter, as well as the small differences in atomic scattering factors from the alloying constituents.

The tendency for the specimen to grow or shorten during irradiation is expressed by the growth index,^{9,14} GI, given by the equation

$$GI = \sum_{hkl} (P_{hkl} - 1) (\cos^2 \beta_{hkl} - \cos^2 \alpha_{hkl})$$

where α_{hkl} and β_{hkl} are respectively the angles which the pole of the plane [hkl] makes with the "a" and "b" crystallographic axes. GI is positive for growth and negative for shortening. More refined forms^{15,16} of the growth index (called G_2 and G_3) are available that take into account the asymmetrical distribution of measured diffraction planes. However, an insufficient number of diffraction planes were measured to justify an exact computation of G_2 or G_3 . The refined growth index, G_2 , is normalized to equal one for a perfect texture (i.e., a single crystal) and hence is convenient for computing the basic irradiation growth coefficient "g".³ For calculations of g in the present report, G_2 is approximated by the expression $G_2 = GI/n$, where n is the number of planes used to calculate GI.

3. RESULTS AND DISCUSSION

3.1 X-ray Diffraction Studies of Fuel Pins

3.1.1. Texture versus Irradiation Growth

Table II summarizes the end-section texture data for seven cast and centrifugally bonded pins, two cast and impact bonded pins, and two specially heat-treated pins. The texture results are in excellent agreement with the EBR-II irradiation test, which showed that only centrifugally bonded pins shortened during irradiation. Centrifugally bonded pins No. 40008, No. 01776, No. 14171, and No. 35424 had the small-grained end under high compressive stress during bonding and have an I_{110}/I_{021} ratio of 0.96 to 1.01. Centrifugally bonded pins No. 44588, No. 02112,

and No. 41397 had the large-grained end under high compressive stress during bonding and had an I_{110}/I_{021} ratio of 1.2 to 1.3. The large gamma grained end is always at the bottom of the cast pin because the casting is cooled from the top, but the casting bottom was not always located in the bottom of the jacketed element. The impact bonded and specially heat-treated pins had a ratio of 0.77 to 0.80. A ratio of 0.79 represents perfectly random orientation.

Composite specimens were also made from a group of transverse slices taken over 2/3 the length of each of two fuel pins. Intensities from twelve diffraction planes [hkl] were measured. In these measurements, a composite specimen from impact-bonded pin No. 049H23 (batch 0494) had an I_{110}/I_{021} ratio of 0.81 and a GI values of +0.15 ($G_2 = +0.01$). The GI value appeared to be slightly biased in the positive direction, although some of these pins lengthened slightly during irradiation. The composite specimen from centrifugally bonded pin No. 01776 (batch 115) had an I_{110}/I_{021} ratio of 1.21 and a GI value of -0.43 ($G_2 = -0.0358$). Fuel pins from this batch shortened up to 1.27 cm during irradiation.

The detailed texture measurements along the length of centrifugally bonded pin No. 41397 were used to calculate GI as a function of length. Intensities from six diffraction planes, (hkl), were measured. These data are compared with measurements of diametrical irradiation growth rate versus length for a companion pin that was irradiated (Figure 2). The variation of diametrical

irradiation growth with growth index is shown to be an approximately linear correlation (Figure 3).

3.1.2. Texture versus Stress Distribution

To verify the postulate² that the texture was induced by the stress, detailed texture measurements were made along the length of centrifugally bonded pin No. 41397. The axial texture distribution, plotted as the I_{110}/I_{021} ratio, was compared with the axial compressive stress distribution during centrifuging in Figure 4. The texture varied approximately linearly with the stress and went to zero (i.e., I_{110}/I_{021} approached 0.79) when the stress was reduced to about 0.3 kg/mm^2 . Hence, the threshold for inducing texture by externally applied stress during the $\gamma \rightarrow \alpha$ transformation in this alloy is on the order of 0.3 kg/mm^2 .

3.1.3. The Basic Irradiation Growth Coefficient, "g"³

For small dimensional changes, g is equal to the percent growth per atom percent burnup of a single crystal. Since the growth index, G_2 , is equal to 1 for a single crystal, an approximate value for g can be extrapolated from polycrystalline texture data by dividing the percent growth per atom percent burnup by G_2 .

The G_2 value for the composite specimen of pin No. 01776 discussed above was $G_2 = -0.0358$. Companion pins from this group shortened 1.27 cm from an original length of 34.3 cm after

irradiation to 0.35 atom percent burnup. Only 2/3 of the pin length underwent shortening; therefore:

$$g = \frac{1.27}{34.3} \cdot \frac{100}{2/3} \cdot \frac{1}{0.35} \cdot \frac{1}{-0.0358} = 443$$

A value of g may also be obtained from the diametrical growth rate versus Growth Index data (Figure 3). The slope of the line gives a diametrical growth rate of 3.5 percent per atom percent burnup per 0.1 GI. To get the corresponding shortening in length, assume conservation of volume, in which case the length shortening rate equals twice the diametrical growth rate, i.e., 7 percent. Now $G_2 = GI/n = 0.1/6 = 0.0166$, and $g = 7/0.0166 = 422$. Hence, the g obtained from the length shortening data is quite consistent with that obtained from the diametrical growth data.

The average growth rates obtained from the composite and profile can be compared with Buckley's¹⁷ results. Buckley found g for pure polycrystalline uranium to vary considerably as a function of fission rate and irradiation temperature. For the EBR-II fission rate of approximately 7×10^{13} fission/(cm³-sec), and a temperature of 470°C corresponding to the average irradiation temperature of the bottom two-thirds of the EBR-II fuel pins, Buckley's g value for a pure uranium single crystal is 500 (percent growth per percent burnup). Hence, the uranium-fissium g value of ≈ 430 is reasonable considering that one of the main constituents of this alloy, molybdenum, has been found by McDonnell, et al.¹⁸ and by Baron and Cadalbert¹⁹ to lower the

growth rate compared to pure uranium. Other alloying constituents (e.g., Fe) increase the g value compared to pure uranium. The alloying constituent will affect the type and amount of texture formed under stress as well as the resistance to radiation damage. Hence, with an alloy system as complex as uranium-fissium, it would be difficult to predict the effect of each alloying constituent (Mo, Ru, Pd, Zr, Nb) and impurity (Fe, Si, Ni, etc.) on the growth rate as a function of temperature and fission rate.

3.1.4. Growth versus Burnup and Irradiation Temperature

For a given texture the diametrical growth rate, g , of the EBR-II pins diminished with burnup as shown in Figure 5. In contrast, Kittel and Paine²⁰ have shown that the growth rate of uranium rods rolled at 300°C was undiminished for burnups up to 0.6 atom percent and fission rates in the range 0.4 to 3.2×10^{13} fission/(cm³ -sec) and temperatures of 60 to 160°C. These as-rolled rods had very large [010], [041] axial texture. When preferred orientation in the rods was reduced by beta-treating at 735°C for 30 min and then quenching in water, their growth rate diminished considerably with burnup.²⁰ These beta-treated rods had very small textures like the EBR-II fuel pins. McDonnell also found that growth rate decreased with burnup for variously heat-treated uranium alloys with small textures.¹⁸

One possible mechanism for the reduced growth rate with burnup for heat-treated uranium with small textures is the larger

amount of intergranular interactions that occur among the approximately randomly oriented grains as they strain against one another and become increasingly hardened and distorted. The higher irradiation temperature of the EBR-II fuel pins (430-550°C) may also cause reduction of growth rate with increased burnup by affecting the equilibrium state between dynamic vacancy cluster generation and annihilation by the irradiation growth mechanism proposed by Buckley.¹⁷

Regarding effects of irradiation temperature, Walter, et al.²¹ have shown that for a given texture, the diametrical growth rate is decreased by a factor of about four when the irradiation temperature is increased from 440 to 540°C.

3.1.5 Effect of Specimen Electropolishing Solution on Texture Measurements

The cold-work layer caused by grinding was routinely removed from the surface of X-ray diffraction specimens by electropolishing. The microstructural appearance of the uranium-fissium alloy after electropolishing, shown by scanning electron microscopy in Figure 6, suggested that two major phases may have been present, even though density, hardness, and x-ray diffraction measurements indicated that the alloy was greater than 95% alpha uranium. The different microstructural appearance of the alpha around the grain boundaries from that in the matrix may be due to a difference in chemical reaction between the electropolish and the grain boundary alpha. The grain boundary alpha had a somewhat different chemical composition than the matrix alpha

because of the minor phases present in the grain boundary region.

The question arose as to whether both regions containing alpha uranium were textured. When electropolished with phosphoric acid, the alpha uranium around the grain boundaries was in relief, and hence contributed less to the diffraction intensities because of preferential absorption of X-rays diffracted from the regions in relief. This fact was used to determine the relative contribution of the two regions of alpha uranium to the measured diffraction intensities, by grinding and electropolishing the same specimen (the bottom end of injection-cast and centrifugally bonded pin No. 02112) twice using a different electropolish each time. (See Section 2.1. for detailed procedures.) The X-ray scan from the surface more uniformly polished with chromic-acetic acid was more characteristic of a random texture as shown by the alpha uranium triplet (Figure 7), e.g., compare with Figure 1a in which the intensity ratios are nearly those for random texture. The experiment was repeated several times on the same specimen using freshly polished surfaces and gave the same results. Hence, this limited study suggested that the form of alpha uranium surrounding the grain boundary had a more nearly random texture.

Metallography, microprobe, and hardness observations indicated that in the grain boundary region the form and amount of alloying constituents were different than in the bulk of the grains; these differences may have prevented the formation of texture. There is also evidence from work on other uranium alloys²² that the

amounts and kinds of alloying constituents affect the susceptibility of a material to stress-induced texture.

Assuming that the grain boundary alpha phase indeed has a more nearly random texture than the matrix, it follows that less texture will be induced in pins that have the small-grained end under high compressive stress during centrifuging, since there is more grain boundary phase in the small-grained end of the pin. This is consistent with our observations for pins Nos. 40008, 14171, 01776, and 35424 (Table II).

3.1.6. Line Broadening and Lattice Contraction

Compared to pure uranium, the alpha-phase diffraction lines of the uranium-fissium fuel pins were broadened and the orthorhombic alpha lattice was contracted approximately 0.05 \AA in the [010] direction (Figure 1). In general, the diffraction lines of the centrifugally bonded pins were much broader than those for the impact-bonded or the specially heat-treated pins.

Regarding lattice contractions, the [OKO]-type reflections of both centrifuge- and impact-bonded pins were shifted to high 2θ values; for example, [020] was shifted from 30.46° to 30.75° and the [021] was shifted from 35.55° to 35.75° . Delaplace²³ observed similar [010] lattice contractions (0.03\AA) for uranium-1.15 wt% molybdenum cooled at rates greater than $400^\circ\text{C}/\text{sec}$. The lattice parameter returned to its normal value after this alloy was annealed for 50 hours at 500°C .

3.2. Microstructure, Microprobe, and Hardness Studies

The microstructures of the impact-bonded and centrifugally bonded fuel pins are shown in Figure 8. Both types of fuel pins had small gamma grains at the top and larger gamma grains at the bottom end of the pin compared to the as-cast condition, because the pins were cooled from the top after injection casting. All the pins showed the original gamma grain boundaries, even after transformation to the alpha phase during bonding. In both types of pins, the alpha phase had a mottled appearance in the regions around the grain boundaries, however, the alpha grains were not metallographically distinguishable. These regions were also slightly softer than the bulk grain regions. Because of the grain size difference, there was generally more of the grain boundary alpha phase on the small-grained end of the pins. Microprobe studies showed that there was more ruthenium than molybdenum around the grain boundaries (aside from the U_2Ru particles) and more molybdenum than ruthenium in the matrix.

The scanning electron micrographs (Figure 6) show the equiaxed dark gray grains of the matrix and an acicular, light gray structure around the grain boundaries. The white phase that delineates the original gamma grain boundaries is believed to be U_2Ru . The randomly dispersed, star-shaped precipitates were found by microprobe analysis to be rich in zirconium. These micrographs also clearly show that the grain boundary regions are preferentially removed during electropolishing.

The replica electron micrograph (Figure 9a) gives the best view of the acicular structure in the grain boundary region of the impact-bonded pins. There is also some evidence of a banded structure (Figure 9b) in the equiaxed grains of the large-grained end of both impact bonded and centrifugally bonded pins. These bands may be evidence of a martensitic transformation or a coarser form of the acicular structure around the grain boundaries. Zegler and Nevitt⁸ observed a martensitic alpha structure, but only in a U-3 wt% fissium alloy, water-quenched from 720°C. This alloy transforms from the gamma phase at 720°C, through the beta phase, to the alpha phase, whereas the beta phase does not occur during cooling of the U-5 wt% fissium alloy.

Harding and Waldron²⁴ have also observed decomposition structures of gamma-phase uranium alloys (10 at% Ti, Nb, Zn, Mo), with differing metallographic appearance, that exhibited a similar x-ray lattice contraction to the uranium-fissium alloy. The structures were designated "distorted alpha" since there was a contraction in the b-direction of the normal orthorhombic alpha cell. The difference in appearance was attributed to transformation both by a shear and by a nucleation and growth mechanism. The type of transformation produced depended upon the composition, the cooling rate from the gamma phase, and the effect of the solute element on the decomposition rate of the metastable gamma phase.

The Vickers diamond pyramid hardness (DPH) values found for an as-cast pin, a centrifugally bonded pin, and an impact bonded pin are summarized in Table III. In the bonded pins, the hardnesses of both the grain boundary region and the matrix were measured. The hardness of the matrix was about the same for both centrifugally bonded and impact bonded pins; however, the region around the grain boundary was softer in both impact bonded and centrifugally bonded pins and was softest for the impact bonded pins. Since the microstructure and hardness of the grain boundary regions were different from the matrix, it was not unreasonable that the texture was also different.

Even though there is a variation of hardness between the matrix (635-640) and grain boundary regions (556-584) of the bonded pins, all the values correspond to the alpha phase of uranium; the value of an injection-cast pin before bonding corresponds to the hardness of gamma uranium (232).

3.3 Mechanism for Stress-Induced Texture

Although the atomistic details of the $\gamma \rightarrow \alpha$ transformation in uranium-fissium alloys is not well established, a discussion of how stress might influence the transformation is in order. The mechanism proposed by Riggs and Neumann²² to explain texture induced in cylinders of uranium end-quenched from the beta phase provides a reasonable mechanism for the present case;

namely, that the texture formation is a stress-relieving mechanism in which grains are oriented during nucleation and growth of the alpha phase such that the "weaker" bonds of the [010] crystallographic directions are parallel to the directions of the tensile stress.

3.3.1. Similarity of Texture Profiles

The centrifugally bonded pins have a "texture profile" similar to the end-quenched uranium cylinders, i.e., an [001], [110] axial texture and a deficiency of [021] at the high-stress end of the pin. As the distance from the end increases, the [110] decreases, and the [001] increases with little change in the [021] (Figure 10).

3.3.2. Effect of State of Stress

For the end quenched cylinder of uranium the sources of stress are the thermal stresses and the stresses caused by the volume change during the $\beta \rightarrow \alpha$ transformation. The stress at the transformation interface increases with cooling rate and hence, is maximum at the quenched end. Riggs and Neumann²² assume the transformation interface to be a plane perpendicular to the cylinder axis so the stress occurs on a cross section. Under these conditions, the alpha phase transformation front is subjected to radial tension, while the adjoining beta phase is in radial compression. Assuming that the $\beta \rightarrow \alpha$ transforma-

tion occurs at 600°C or lower, the beta phase could exert a stress of 6000 psi or greater on the alpha phase before the alpha-phase yielded.

For the centrifugally bonded pins, the principal source of stress is the stress from the centrifuge. The thermal stresses should be small and relieved quickly, because the $\gamma \rightarrow \alpha$ transformation occurs at high temperatures ($\sim 500^\circ\text{C}$) where the thermal energy of the structure is high. The stress from volume changes during the $\gamma \rightarrow \alpha$ transformation is small because the gamma phase has a much lower yield strength than the alpha phase. The radial direction of the centrifugally bonded pin is also in tension during $\gamma \rightarrow \alpha$ transformation, so alignment of the weaker bonds of the [010] direction parallel to the direction of tensile stress would also provide the greatest stress relief during transformation. The maximum stress for the centrifugally bonded pins is estimated to be about 2 kg/mm². In agreement with the higher stress level, there is more texture²² developed in the end-quenched uranium alloys than in the centrifugally bonded EBR-II fuel pins.

3.3.3. Other Mechanisms That Induce Texture

For the end-quenched uranium alloys, Riggs and Neumann²² considered whether the texture might be induced by plastic deformation of the alpha phase by the beta phase, but concluded that slip would require too much elongation to produce the large reorientation required to explain the observed texture. Twinning

would give a large reorientation without a large elongation, but the volume fraction of twinned material was too small.

No twinning was observed for the centrifugally bonded pins. There is, however, a possibility that the banded structure found in the equiaxed grains (Figure 9b) was produced by the centrifuge stress causing shear to occur more easily on one set of planes as proposed by Walter, et al. ²¹

4. CONCLUSIONS

Preferred orientation studies of EBR-II U-5 wt% fissium fuel pins showed that injection-cast and centrifugally bonded pins have a [110], [001] axial texture that is responsible for their shortening in length and diametrical growth during irradiation. Injection cast and impact bonded pins have a nearly random texture and are dimensionally stable during irradiation.

The growth indices calculated from the texture data correlate well with observed diametrical irradiation growth rate and extrapolate to a single crystal growth rate, g , of 430 percent per percent burnup, which is in the range measured for uranium and uranium alloys.

The observed growth also correlates well with the diffraction intensity ratio I_{110}/I_{021} . Hence, this ratio can be measured, nondestructively, on fuel pins and serves as a rapid method of texture control.

Texture measurements at small increments along the length of a centrifugally bonded fuel pin showed that the texture varied approximately linearly with the compressive stress from the centrifuge. Hence, the presence of this stress during the $\gamma \rightarrow \alpha$ transformation (500°C) of the fuel pin induced the texture. The texture was maximum at the bottom end of the pin (where the stress was 2 kg/mm²) and decreased to a negligible value about 23 cm from the bottom (where the stress was less than 0.3 kg/mm²).

Two possible atomic mechanisms for the formation of the texture are suggested: a) the centrifugal stresses cause the grains to orient during nucleation and growth of the alpha phase, such that the "weaker bonds of the [010] direction are parallel to the direction of tensile stress, or b) the centrifugal stresses may cause the grains to slip on only one or two sets of planes during the $\gamma \rightarrow \alpha$ transformation and thereby generate the texture.

The microstructure of the pins bonded by both the impact and centrifuge methods had the appearance of a two-phase structure, even though micro hardness and X-ray measurements showed both structures to be alpha uranium. The X-ray measurements also showed that the diffraction lines were broadened; the lattice contracted 0.05 Å in the [010] direction, and the alpha phase around the grain boundaries was apparently less textured than in the matrix. Previous studies of distorted alpha phase formed from other metastable gamma phase uranium alloys found

similar line broadening and lattice contractions. The two phase metallographic appearance may have been caused by the gamma-to-alpha transformation both by shear and by nucleation-plus-growth mechanisms.

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TABLE I
 COMPARISON OF RANDOM DIFFRACTION INTENSITIES
 FROM URANIUM AND U-5 wt% FISSIUM ALLOY

<u>hk1</u>	<u>I_{hk1}^0</u>	<u>I_{hk1}^0</u>
[110]	0.79	0.70
[021]	1.00	1.00
[002]	0.59	0.50
[111]	0.74	0.69
[112]	0.89	0.76
[131]	0.88	0.70
[133]	0.53	0.37
[114]	0.35	0.24

NOTE: GI = +0.0002 for the I_{hk1}^0 of Pin No. 12367 assuming the pure uranium I_{hk1}^0 to be the random standard. The bottom end of Pin No. 12367 had an average value of GI = -0.02 and the top end an average value of GI = +0.04.

TABLE II
 TEXTURE MEASUREMENTS OF BOTTOM END SECTIONS
 OF EBR-II FUEL PINS

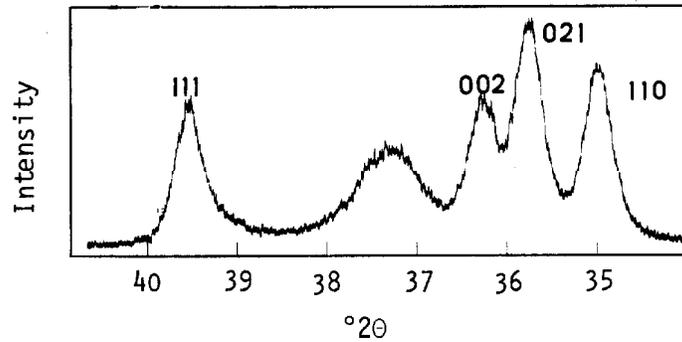
<u>Casting Batch No.</u>	<u>Pin No.</u>	<u>Fabrication History</u>	<u>$I_{[110]}/I_{[021]}$</u>
0494	049H23	Injection cast, impact bonded	0.80
580	38413	Injection cast, impact bonded	0.80
615	40971	Injection cast + 500°C for 1 hr + centrifugally bonded	0.80
295	12367	Injection cast, centrifugally bonded + 660°C for 1.5 hr + impact bonded	0.77
580	35424	Injection cast, centrifugally bonded	1.01
115	01776	Injection cast, centrifugally bonded	0.98
295	14171	Injection cast, centrifugally bonded	0.98
580	40008	Injection cast, centrifugally bonded	0.96
580	44588	Injection cast, centrifugally bonded	1.22
1017	02112	Injection cast, centrifugally bonded	1.31
580	41397	Injection cast, centrifugally bonded	1.32

TABLE III

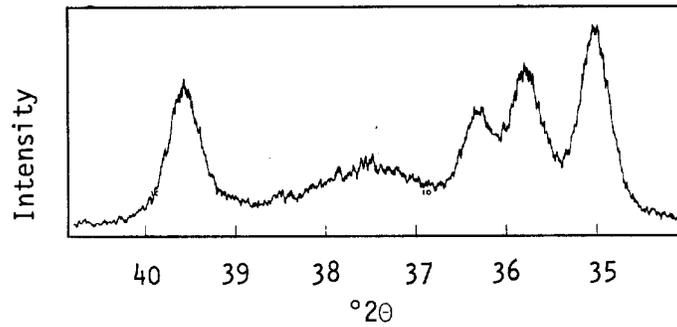
DPH VALUES FOR URANIUM-FISSIUM FUEL PINS

<u>Mode of Fabrication</u>	<u>DPH Value</u>
Injection Cast	232
Injection Cast and Centrifugally Bonded	
Matrix	640
Grain-boundary regions	584
Injection Cast and Impact Bonded	
Matrix	635
Grain-boundary regions	556

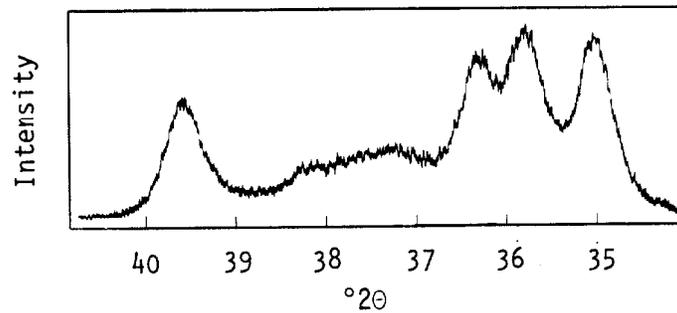
* Tukon hardness tester, 100-g load, Vickers diamond pyramid indenter. Each value is the average of measurements in 5 areas.



a. Impact Bonded Pin 049H23



b. Centrifugally Bonded Pin 02112 (large-grained end)



c. Centrifugally Bonded Pin 01776 (small-grained end)

FIGURE 1. Typical Diffraction Profiles for Bottom End of Centrifugally Bonded and Impact Bonded EBR-II Fuel Pins

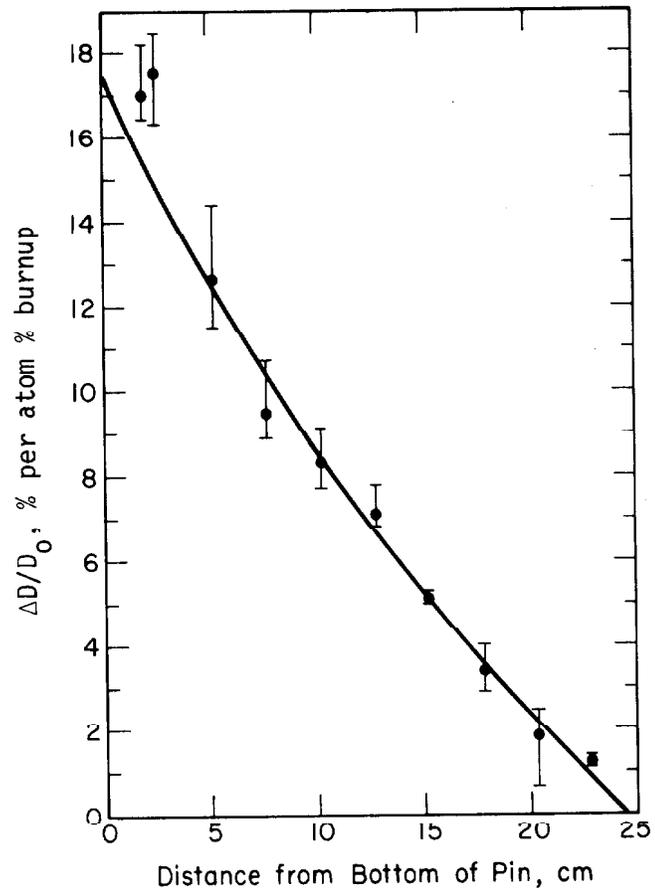


FIGURE 2. Diametrical Irradiation Growth Rate of Centrifugally Bonded Fuel Pin (0.342 % avg. burnup), Corrected for Isotropic Irradiation Swelling. Pin from Batch 580.

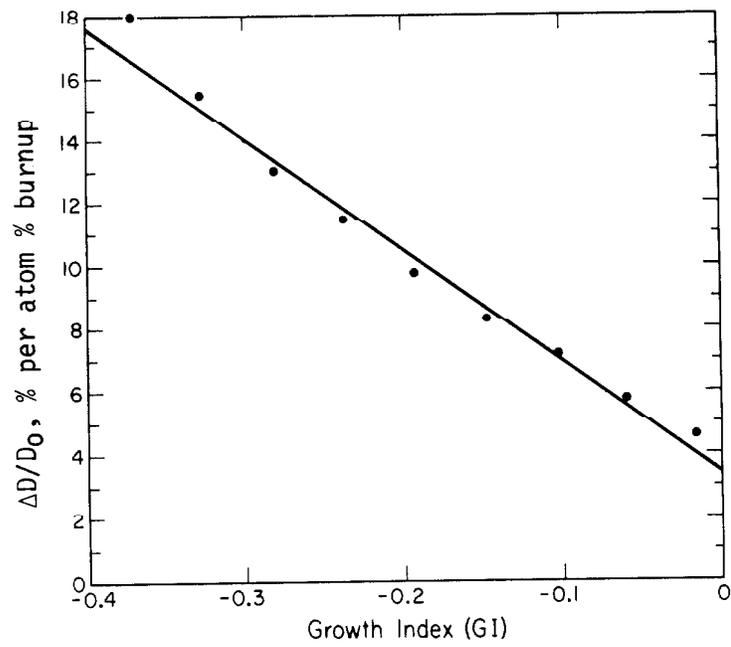


FIGURE 3. Diametrical Growth Rate vs Growth Index for Centrifugally Bonded Fuel Pin (0.342 % atom percent avg. burnup). Pin from Batch 580.

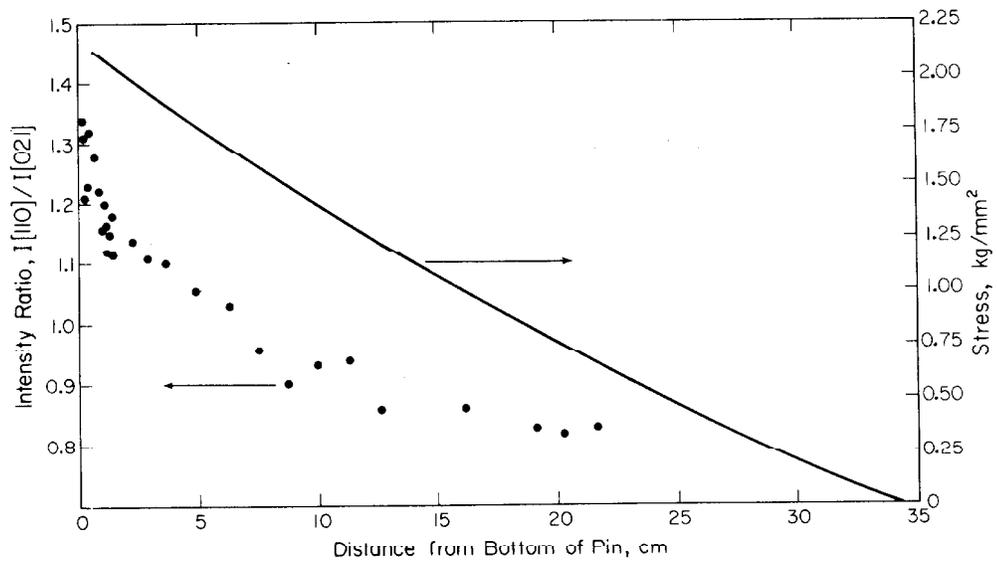


FIGURE 4. Variation of Texture with Compressive Stress Along the Axis of the Pin Line. Pin from Batch 580.

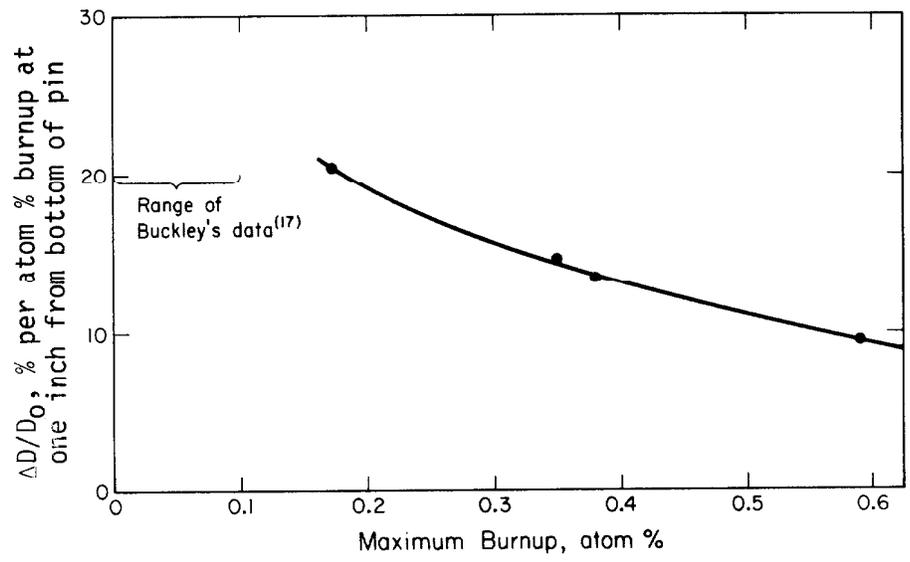
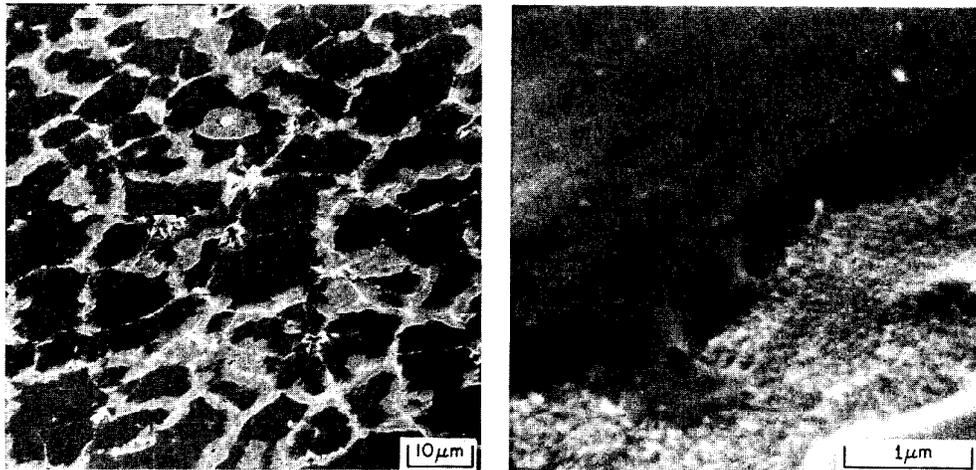
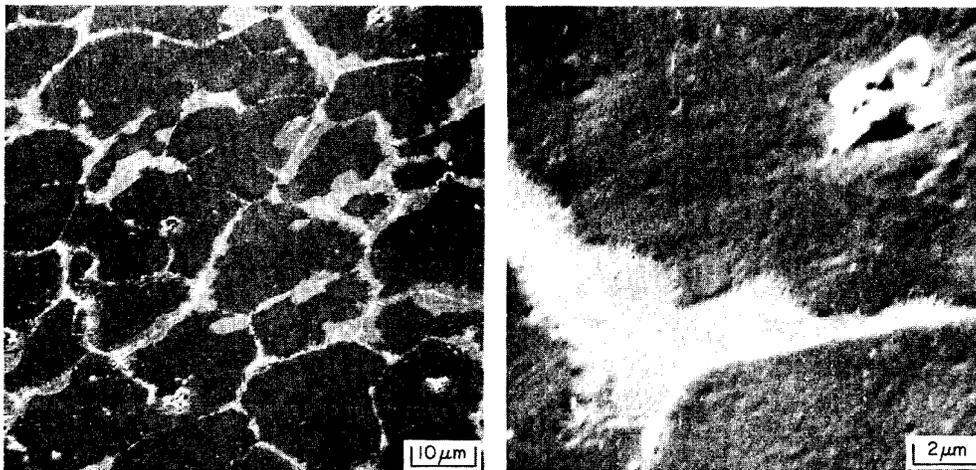


FIGURE 5. Diametrical Growth Rate, g, vs Burn-up

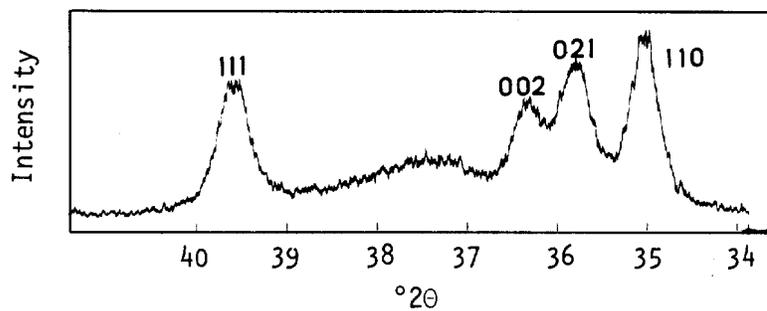


a. Impact Bonded

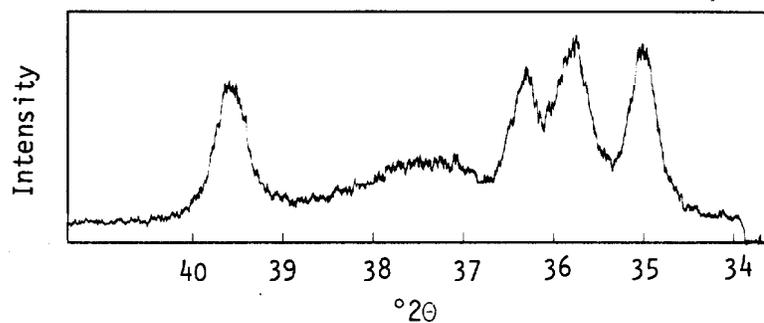


b. Centrifugally Bonded

FIGURE 6. Scanning Electron Micrographs of Impact and Centrifugally Bonded Fuel Pins



a. Standard Phosphoric Acid Electropolish
(Grain Boundary Alpha Phase Preferentially
Removed)

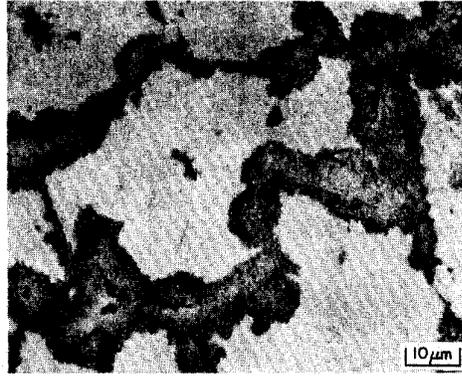
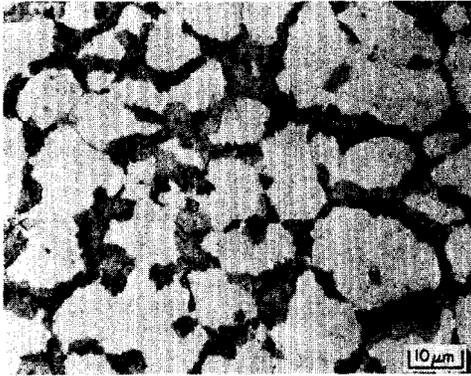


b. Chromic-Acetic Acid Electropolish
(A More Uniform Electropolish)

FIGURE 7. Effect of Electropolish on Diffraction
Intensities of EBR-II Fuel Pins

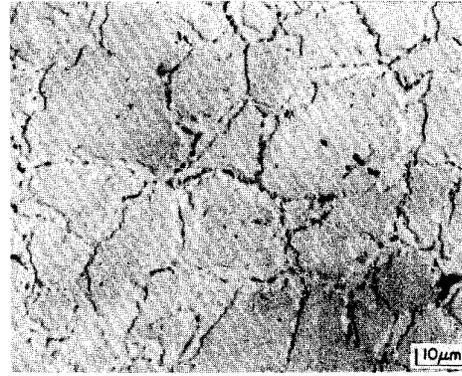
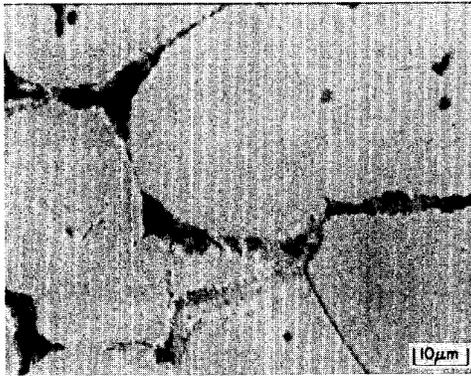
IMPACT BONDED

CENTRIFUGALLY BONDED



a. Top End of Fuel Pin 38413
(bottom end of the casting)

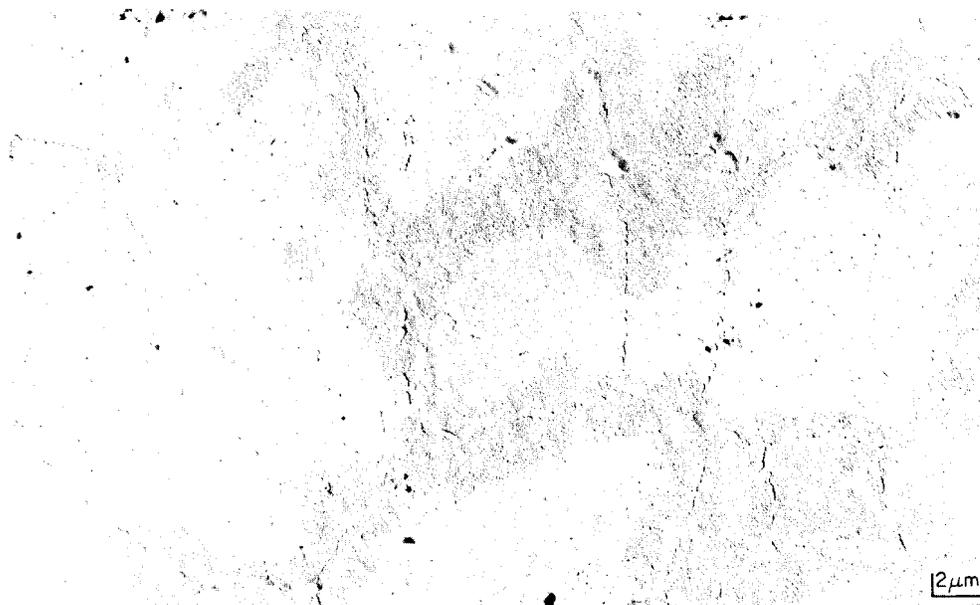
c. Top End of Fuel Pin 44588
and Casting



b. Bottom End of Fuel Pin 38413
(top end of the casting)

d. Bottom End of Fuel Pin 44588
and Casting

FIGURE 8. Optical Micrographs of Impact and Centrifugally Bonded Fuel Pins

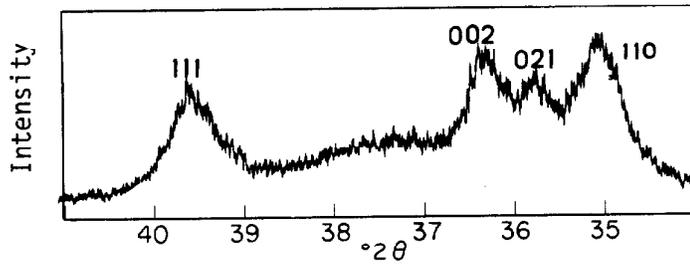


a. Impact Bonded Pin 38413, Small Grained End of Casting (3500X)

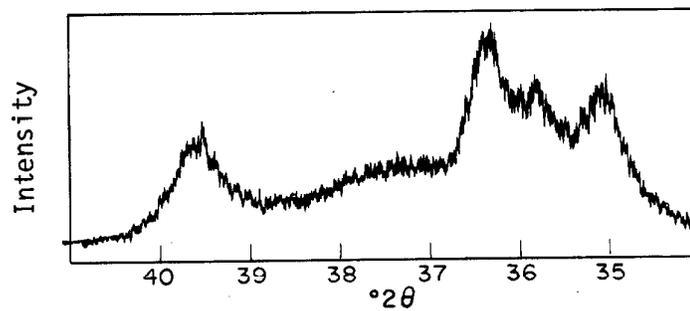


b. Centrifugally Bonded Pin 44588, Large Grained End of Casting (1600X)

FIGURE 9. Replica Micrographs of Grain boundary and Matrix Structure of Impact Bonded and Centrifugally Bonded Pins



a. Bottom End of Pin 41397



b. 1/2 in. from Bottom End of Pin 41397

FIGURE 10. Variation of Texture along the Length of Centrifugally Bonded Pins

Note the increase in $I[002]$ and the decrease in $I[110]/I[021]$ from a to b. This type of variation has been observed in end-quenched uranium cylinders.⁵