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ORIENTATIONS OF LARGE GRAINS PRODUCED
BY STRAIN-ANNEAL TREATMENT OF HIGH
PURITY URANIUM

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The orientations of 16 large grains, which were produced in a rod of high purity uranium in an attempt to prepare a single crystal by the strain-anneal method, have been determined by the Greninger¹ method, using back-reflection Laue photographs obtained from the individual grains. The orientations are reported here relative to the rod axis and to a longitudinal plane of examination parallel to the rod axis. The large grains were formed by strain-annealing from a structure of fine grains produced by 575°C recrystallization after high reduction at 300°C. The results of this study show a preferred orientation of (010) poles in the direction of the rod axis, and all but 3 of the grains have the (001) poles within a 90° range of rotation around the rod axis.

Specimen History

The rod from which the specimen was obtained was produced by rolling and subsequent swaging of a high purity ingot formed by consolidation melting² of electrolytically-produced uranium crystals.³ Chemical analysis showed the following impurities:

Impurity	Analysis (ppm)
Carbon	41
Aluminum	10
Iron	2
Silicon	<10
Nitrogen	<10
Oxygen	not determined
Others	below limit of detection

The small ingot (approximately 3/4" diameter x 1/2" long) was reduced by rolling at 600°C to 57% reduction in area followed by a 74% reduction in area at 300°C. The final diameter, after machining to remove the porous surface, was 0.135".

The rod was annealed for 2 hours at 575°C and strained in a tensile testing machine to 1.5% elongation. The rod was then heated to a maximum temperature of 635°C by passing it through a temperature gradient, and was held at the final temperature for 24 hours. Figure 1a shows the macro-etched rod after the final anneal. The short lengths at the ends of the rod, which were not strained prior to final annealing, retained a fine-grained structure. The large grains averaged 4 - 7 mm in length x 2 mm in cross-section.

Figures 1b, 1c, and 1d are representative microstructures of longitudinal sections showing the grain size at the rod ends (1b), the junction of the large and small grains (1c), and the elongated shapes of the large grains (1d).

Texture of Fine Grains at End of Rod

The texture in the fine-grained structure at the ends of the annealed rod (Figure 1b) was partially determined by the X-ray reflection spectrometer technique.^{4,5} A section through a fine-grained end was ground and polished perpendicular to the rod axis. The diffracted intensities of the (020), (110), (131), (111), and (021) reflections were determined for positions between the perpendicular to the rod axis and 30° tilts from the perpendicular. The variations of intensities versus degree of tilt are shown in Figure 2.

Because of the small size of the specimen no attempt was made to determine complete pole figures. Consequently, the texture cannot be definitely described. On the basis of the texture that has been reported for uranium rods recrystallized after 300°C rolling,⁵ it is assumed that a fibre texture existed. However, in comparing the curves shown in Figure 2 with those obtained by reflection from specimens of the known (140) texture, there was a significant difference in the position at which the peak (110) intensity was observed. The peak in the (110) curves shown in Figure 2 does not correspond to a (140) texture. However, the (020) intensity curve shown in Figure 2 is similar to that obtained from specimens of known (140) texture. A peak in the (020) intensity occurring at the rod axis was generally observed in rods which consisted primarily of the (140) texture. Based on this comparison, it is surmised that the texture in the fine grains was either a single fibre texture, such that the poles aligned with the rod axis were primarily of some plane approximately 15° from (110) towards (010), or a duplex fibre texture consisting of 2 distinct components, one of which was (010) and the other close to (110).

Orientations of Large Grains

A 1" length was cut from the end of the rod shown in Figure 1 and mounted in lucite for metallographic polishing. The polished face was parallel to the rod axis. This section consisted of a large area of small grains adjacent to a coarse-grained structure. Figure 3 is a reproduction of the microstructure at approximately 9 times the original size. The 16 large grains which were studied with regard to orientation are designated by number.

When the microstructure was examined with the polarized light system of the B & L Metallograph, the color of the light reflected from each of the large grains was noted at a fixed rotational position of the microscope specimen stage. The colors observed are noted in Table I.

Table I

COLOR GROUPS OF LARGE GRAINS AS
OBSERVED IN FIXED SPECIMEN

Grain No.	Color
1 2 3 4	Pink
5 6 7	Light Blue
8	Dark Blue
9	Dark Green
10 11 12	Light Green
13 14 15	Orange
16	Light Green

For X-ray studies the polished surface was electrolytically etched in citric-nitric electrolyte,⁶ so that the grain boundaries could be observed with the unaided eye, and the mounted specimen was placed on a stage for alignment with an X-ray beam. The specimen was aligned so that the X-ray beam was perpendicular to the etched section and within the boundaries of a selected grain. Back-reflection Laue photograms were obtained from 16 of the large grains using unfiltered radiation from a copper target tube operating at 35 KV and 20 milliamps. The X-rays diverged from a 0.25 mm pinhole, so that at a 3 cm specimen-to-film distance the beam covered approximately a 1 mm diameter area of the specimen. Exposure times were 2 hours for each photogram.

Typical Laue patterns are shown in Figures 4a, 4b, and 4c. The photograms are oriented with respect to vertical and horizontal directions of the paper to correspond to the stereographic projection of Figure 5 (discussed below). As shown in Figure 4, most of the grains were oriented so that some (h0l) reflection was close to the center of the photogram, and the hyperbolas formed by these reflections were generally close to vertical.

In solving the patterns it was noted that the hyperbolas formed by reflections from planes of the $\langle 110 \rangle$ zones were most prominent. Reflections from (001), (116), (225), (112), and (111) planes of the above zones were most intense. Reflections from the [010] zone were of relatively feeble intensity after 2 hour exposures. At low angles to (001), the (102), (203), and (101) reflections were prominent, whereas at angles approaching (100), the (102) and (201) reflections were recognizable. The (100) reflection was generally weak, although present.

As shown in Figure 4d, twin traces were observed in most of the large grains in this specimen, as is commonly observed in uranium grains even though the structures are not intentionally strained. Although the twin bands are relatively thin and widely dispersed, Laue spots produced by diffraction from the twin orientations were observed in a number of the photograms. An exceptionally intense pattern of the twin orientation is shown in Figure 4b. When plotting the zones of the twin and parent orientations on the same projections it was found that the twin orientation corresponded to $K_1 = (130)$.

The stereographic projections of all the orientations with respect to the rod axis and to the transverse direction in the longitudinal plane of examination are shown in Figures 5 and 6. In Figure 5, the plane of projection corresponds to the plane of examination with the specimen in a fixed position. In Figure 6, the plane of projection corresponds to a transverse plane perpendicular to the rod axis. Each pole position has the designations of the corresponding grain in Figure 3.

The existence of a texture in the coarse-grained structure is definitely established, as is shown in both projections. The alignment of the (010) poles with the rod axis shows clearly in Figure 6. Of the 16 orientations, only 3 of the [010] directions formed an angle with the rod axis greater than 12° . The largest deviation from this alignment, the orientation of grain #5, is 25° .

A deviation from a true fibre texture is noted in the positions of the (001) and (100) poles. As is more easily seen in Figure 5, a majority of the (001) poles are positioned in the upper hemisphere of the projection. Thirteen of the 16 (001) poles are grouped within a 90° range of rotation around the rod axis.

Discussion of Results

A comparison of the preferred orientation of the large grains with the texture of the adjacent fine grains shows a significant difference. Whereas the [010] alignment with the rod axis is common to both textures, the preferred orientation which caused the peak in the (110) diffracted intensities (Figure 2) is absent in the large-grained matrix. Assuming that the texture in the fine grains is similar to that which existed in the penultimate structure (the fine-grained structure which existed prior to strain-anneal treatment), it may be concluded that the strain-anneal treatment produced a single (010) fibre texture from either a single fibre texture of a different type or from a duplex fibre texture. It is interesting to note that large grains and single crystals, produced by grain coarsening in uranium rods⁷ during annealing of non-strained fine grained structures, are of orientations similar to the large-grained texture reported above. The [010] directions in the coarse grains have seldom been found at angles greater than 15° from the rod axis. Since, because of similar fabrication and annealing history, the structures prior to coarsening are presumably of a texture close to that described for the penultimate structure in this strain-anneal specimen, there appears to be some factor which causes selective growth of grains of the observed orientation. This factor may have also been responsible for the large-grained texture reported in this study. Although the 1.5% strain was necessary for large grains to form during subsequent annealing, it may have had no effect on the orientations of the large grains.

From observations of the shapes of the large grains, and from a correlation of relative grain size and orientation, there is some basis for postulating that the single fibre texture of the large grains may be the result of a preferred direction of grain growth in a fibrous matrix. The fact that most of the large grains were of shapes elongated in the direction of the rod axis indicates that the grain growth was most rapid in this direction. Secondly, the relative grain sizes, as observed at the longitudinal section examined, indicates a grain size decrease with deviation from [010] alignment with the rod axis. Grains #5, 15, and 16 (Figure 3), which showed the largest deviations from the average orientation, also were of smaller than average apparent grain size.

The existence of a non-uniform penultimate structure may account for the above observations. The microstructures which result from recrystallization after heavy reduction at 300°C are generally fibrous in appearance. The structures appear to consist of bands made up of similarly oriented grains when examined under polarized light. This is shown to some degree in Figure 1b. The bands differ in colors observed and are approximately parallel to the rod axis. It has also been observed that particles of impurity phases also tend to be aligned in fibrous groups, elongated in the rolling direction. Because of either orientation differences between bands, or because of inclusion barriers, a grain may have difficulty in growing in transverse directions. Indications that such barriers exist have been found in other uranium structures, a pronounced example of which is shown in Figure 7. A uranium rod, containing approximately 200 ppm of tantalum, was critically strained and annealed. As shown in Figure 7, the large grains which grew during annealing appeared to be restricted to local bands.

Assuming that the [010] direction is a preferred direction for grain growth, those grains or recrystallization nuclei, in which this direction was close to parallel to the fibres or bands, would have been most favorably oriented for rapid growth. Rapid growth of these orientations would have prevented subsequent growth of less favorably oriented grains.

Summary

The orientations of 16 large grains produced by strain-anneal treatment of a fine-grained uranium rod were determined from back-reflection Laue photograms. The (010) poles of the large grains were significantly aligned close to the rod axis, whereas the texture in the fine grains, as indicated by analysis of partially completed pole figures, consisted of either a duplex fibre texture, (the (010) pole and another pole close to (110) being parallel to the rod axis) or a single fibre texture with a pole lying between the (110) and (120) poles being parallel to the rod axis but exhibiting considerable scatter.

A mechanism is proposed which would account for the observed results, assuming the [010] direction to be a preferred direction for grain growth.

Acknowledgments

All the Laue photograms were taken by Mrs. Alice Smigelskas Fischer. The spectrometer work, by which the fine-grained texture was partially determined, was carried out by Dr. M. Mueller.

References

1. A. Greninger, Determination of Orientations in Metallic Crystals, Trans. AIME, 117, p. 61 (1935).
2. B. Blumenthal: ANL-5019: Melting of High Purity Electrolytic Uranium.
3. R. A. Noland, C. Marzano: ANL-5102: Electrolytic Refining of Uranium.
4. L. G. Schulz, A Direct Method of Determining Preferred Orientation in a Flat Reflection Sample Using a Geiger Counter X-ray Spectrometer, J. Appl. Phys., 20, 1030 (1949).
5. W. P. Chernock & P. A. Beck: ANL-4839: Quantitative Determination of Rolling and Recrystallization Textures in 600°C and 300°C Rolled Uranium Rods.
6. J. Baumrucker: ANL-4398: Metallographic Technique for Uranium.
7. E. Fisher: ANL-5021: Preparation of Uranium Single Crystals.

Figure 1 - Grain Size Distribution in Rod

Figure 1a

Macro 4696

1X

Figure 1b

Micro 7378

B

50X

Figure 1c

Micro 7377

C

50X

Figure 1d

Rod Axis

Micro 7383

D

50X

- Figure 1a - Grain size distribution along full length of rod after final anneal.
 1b - Microstructure at non-strained ends.
 1c - Microstructure at junction of fine and large grains.
 1d - Microstructure at large grained section.

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Figure 3 - Reproduction of Microstructure, Showing Size and the Reference Number of Large Grains

ANL Negative No. 106-983

Figure 4 - Laue Photographs Showing Typical Patterns

Figure 4a

Micro 14084

A

Figure 4b

Micro 14083

B

Figure 4c

Micro 14082

C

- Figure 4a - Pattern from grain #4 - reflections from planes close to (100).
 4b - Pattern from grain #9 - reflections from planes between (100) and (001).
 4c - Pattern from grain #6 - reflections from planes close to (001).

Rod Axis is Horizontal

FIG. 5: Orientation of 16 Grains with Reference to Rod Axis and Transverse Direction in Plane of Examination

FIG. 6: Orientation of 16 Grains with Reference to Rod Axis

Projection as Translated from Fig. 5 - by following Rotation Sequence:

- (1) 90° Clockwise about Vertical Axis
- (2) 90° Counter-Clockwise about Center

Figure 7 - Grain Growth in a Banded Structure

Micro 10050

50X

Large grains of elongated shapes in a fibrous fine-grained structure, after strain-anneal treatment. The shapes of the large grains appear to be associated with the bands within the matrix.