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PRELIMINARY ANALYSIS OF FISSION-INDUCED DIMENSIONAL
CHANGES IN SINGLE CRYSTALS OF URANIUM

by

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ABSTRACT

Irradiations of uranium true single crystals and lineage crystals are described. Analysis of the dimensional and shape changes results in the crystallographic deformation rates: $G_{[100]} = -420 \pm 20$, $G_{[010]} = 420 \pm 20$, and $G_{[001]} = 0 \pm 20$, where G is the logarithmic increment divided by the fission ratio. The lineage crystals elongated in the $[010]$ direction at appreciably higher rates than the single crystals.

A. INTRODUCTION

During the course of a preliminary investigation concerning the irradiation behavior of natural uranium⁽¹⁾ with neutrons, small single crystals of natural uranium became available. Specimens which had been manufactured by Fisher^(2,3) at Argonne National Laboratory were examined after irradiation under this general program. Qualitative results agreed well with similar work done at about the same time at Knolls Atomic Power Laboratory,^(4,5) using an ANL crystal. The present report describes the ANL experiment and the analytical method whereby quantitative growth rates in the three major crystallographic directions were deduced.⁽⁶⁾

B. DESCRIPTION OF IRRADIATION

A total of six crystals were prepared for irradiation. Three of them were made by grain coarsening in the high alpha temperature range;⁽²⁾ three were obtained by beta-to-alpha gradient transformation.⁽³⁾ The crystallographic structure of all specimens was determined by back reflection X-ray photograms, typical examples of which are shown in Figure 1. It was found that the coarsened specimens were true single crystals, whereas those obtained by transformation were lineage crystals with considerable scatter in orientation.

Figure 1. Laue Photographs of Uranium Crystals. a) Single Crystal "B", b) Lineage, or Pseudo Crystal.

21731

13865

Copper radiation; Beam diameter, 1 mm; film distance, 3 cm.

The original shape of the true crystals is shown in Figures 2-a, 3-a and 4-a. They were approximately right circular cylinders, 0.12" in diameter and from 0.07" to 0.20" long. As a fiducial mark on each specimen, a flat was ground on one side and was beveled at one end. The back reflection patterns were obtained on the unbeveled ends, and the [010] pole was found to be within 17° of the cylindrical axis in all specimens.

The pseudo crystals were somewhat larger, ranging from 0.18" to 0.19" in diameter, and from 0.45" to 0.68" in length. The fiducial flats were much deeper than those on the single crystals, cutting down the diameters as much as 15-30% along one side. The [010] pole was found to be approximately $25 \pm 5^\circ$ from the cylindrical axis of these specimens.

The single crystals were photographed to record their shape (Figures 2-4) and were enclosed in three capsules for irradiation in the Materials Testing Reactor. The pseudo crystals were placed in loosely fitting stainless steel sleeves to minimize lateral warping, and were paired with the single crystals in the same capsules. The encapsulating technique, using liquid NaK alloy as a heat transfer agent, has been described elsewhere.⁽⁷⁾

The capsules were irradiated for differing periods, to produce total atom burnups ranging from 0.1% to 0.4%. It is estimated that the temperature during irradiation did not exceed 150°C and probably was much lower.

C. DISCUSSION OF RESULTS

The appearance of the specimens after irradiation is seen in Figures 2 to 5. In general, the crystals became elliptical cylinders with oblique ends, lengthening axially and shortening in the diameter corresponding with the [100] direction. The true crystals suffered various minor irregularities in their deformation, but two of them could be measured fairly successfully. The pseudo crystals deformed so radically that one of them could not be removed from its retaining sleeve, and only one could yield a fair length measurement (Figure 5).

The dimensional and shape data obtained from the pre- and post-irradiation photographs were subjected to a geometrical analysis, details of which appear in Appendix A. The data, together with the results of computations, are tabulated and discussed in Appendix B.

The work confirms the KAPL observations⁽⁵⁾ which noted null growth in the [001] and equal growth rates, of opposite sign, in the [010] and [100] directions. The KAPL workers were able to obtain their results by simple linear analysis because their specimen happened to possess

Figure 2. Pre- and Postirradiation Appearance of Uranium Single Crystal A.

13602

13600

- a) Before irradiation, magnified 10X. End and longitudinal views show the flat and bevel marks used to identify X-ray orientation.

14328

14332

14329

- b) After 0.110 a/o total burnup, magnified 4X. The views are along the $[001]$, $[100]$ and $[010]$ crystallographic directions (left to right) and are reversed by a mirror.

Figure 3. Pre- and Postirradiation Appearance of Uranium Single Crystal B.

13599

13601

- a) Before irradiation, magnified 10X. Specimen slightly conical, and beveled to mark $[001]$ orientation.

14887

14890

- b) After an estimated 0.27 a/o total burnup, magnified 2X. Views are along the $[001]$ and $[100]$ crystallographic directions (left to right) and are reversed by a mirror.

Figure 4. Pre- and Postirradiation Appearance of Uranium Single Crystal C.

13597

13598

- a) Before irradiation; magnified 10X. Flat and beveled surfaces were used to identify X-ray orientation.

14896

14894

- b) After an estimated 0.37 a/o total burnup, magnified 2X. The views are along the $[001]$ and $[100]$ crystallographic directions (left to right) and are reversed by a mirror.

Figure 5. Postirradiation Appearance of Lineage Crystals. a) Pseudo A, 0.110 a/o Burnup, b) Pseudo B, 0.27 a/o Burnup, c) Pseudo C, 0.37 a/o Burnup. Note retaining sleeve in photograph b) which could not be removed.

a)

14333

4-X

b)

14891

2-X

c)

14892

2-X

symmetry between its geometry and the crystallographic directions. It is statistically rare for this to occur in the cylindrical single crystals produced at ANL.

The magnitudes of the irradiation growth rates which result from the present analysis have been reported elsewhere.⁽⁶⁾ They are:

$$\begin{aligned} G_{[100]} &= -420 \pm 20 \\ G_{[010]} &= 420 \pm 20 \\ G_{[001]} &= 0 \pm 20, \end{aligned}$$

where G is the logarithmic increment in the indicated direction divided by the fission ratio producing it. Further studies are in progress to determine the effect of the irradiation variables upon the magnitudes of these rate coefficients.

The KAPL workers reported⁽⁵⁾ that the $[010]$ elongations in single crystals and pseudo crystals appear to have the approximate relationship 1:1.5. The ANL work cannot confirm these numbers quantitatively, inasmuch as the growth rates of the pseudo crystals could not be computed. However, from Table II, Appendix B, it may be seen that the ratio of final length as normalized to initial length of Pseudo A is some 15% greater than for Crystal A, which had the same burnup. It appears from this that the ratio between their $[010]$ growth rates should be appreciably greater than 1.15, inasmuch as the skew angle of the pseudo crystal greatly exceeds that of the single crystal. The work, therefore, does lend qualitative support to the KAPL observations.

D. CONCLUSIONS

Although the preliminary character of this experiment is quite apparent, it confirmed contemporary findings at KAPL concerning the basic dimensional behavior of uranium under neutron irradiation and provided the first quantitative data concerning the magnitude of the effect. The analytical method developed for determining the orthogonal deformation ratios does not require initial alignment of the crystal geometry with the crystallographic directions. Under optimum conditions the accuracy possible in the analysis is considerably better than the present accuracy of burnup and temperature determinations in irradiation experiments.

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APPENDIX A

Method of Geometric Shape Analysis

A rigorous three-dimensional geometric analysis of the shape changes in anisotropic crystals is possible if accurate measurements of dimensions and skew angles can be made. Such an analysis can resolve the distortion into an orthogonal system of three principal growth rate vectors and specify the angular relationship of these to the initial and final crystal geometry. Unless one of the vectors is aligned with an axis of symmetry in the specimen, it is impossible to isolate any two of the vectors in the same plane of symmetry. In general, therefore, the analysis must first specify the symmetrical planes in which the growth rate vectors lie, and then determine the angular relationship of the vector to the symmetrical axes of these planes.

When accuracy of measurement is not very good, as in the case of the crystals under consideration, some simplifying approximations are justified. It may be assumed with fair accuracy that, if the [010] is near the cylindrical axis of the specimen, a plane can be chosen by inspection, which may be considered to include two of the growth rate vectors. The natural choice, of course, would be the plane with which the [010] and the [001] directions are nearly parallel, as it is easily photographed and measured, and its skew angle is small. The [100] [010] plane can be photographed only when the crystal is standing on edge, and the skew angle must be deduced from the minor axis of the elliptical end surface.

In general, any longitudinal axial section of a right circular cylindrical uranium crystal will be a rectangle before irradiation and an oblique parallelogram after. Figure 6 illustrates this effect. Any point, $P(d, h)$, on the surface of the rectangle, $OABC$, will be translated to $P'(d', Lh + d' \tan \beta)$ after irradiation. Here d , l , d' and l' represent the lateral diameter and vertical length of side before and after deformation; L is the ratio of l' to l ; h is the distance AP ; Lh is the distance $A'P'$; and β is the skew angle. In this construction it is required to find the maximum and minimum of the ratio

$$R = \frac{OP'}{OP} = \sqrt{\frac{d'^2 + (Lh + d' \tan \beta)^2}{d^2 + h^2}} \quad (1)$$

The condition $\frac{dR}{dh} = 0$ yields the solution:

$$R_m = \sqrt{L^2 - \frac{N^2}{2(M \pm \sqrt{M^2 + N^2})}} \quad ; \quad (2)$$

$$\left. \begin{aligned} M &= D^2 \sec^2 \beta - L^2 \\ N &= 2DL \tan \beta \\ L &= l'/l, D = d'/d \end{aligned} \right\} \quad (2a)$$

Figure 6

Diagram of a Longitudinal Axial Section through a Right Cylindrical Uranium Single Crystal before and after Irradiation.

The two values of R_m obtained from (2) may then be used to compute the magnitudes of the maximum and minimum growth vectors lying in the plane being considered, if the per cent total atom burnup, B , is known:

$$G_i = \frac{100 \ln R_m}{B} \quad . \quad (3)$$

Now if it be further assumed that the crystal suffered no volume change during irradiation, the magnitude of a third orthogonal growth vector may be immediately deduced from the relationship

$$G_1 + G_2 + G_3 = 0 \quad . \quad (4)$$

The directions of the maximum and minimum growth vectors before and after deformation of the parallelogram are designated as angles ϕ and ϕ' in Figure 6. They are readily found from the relationships

$$\tan \phi = \frac{h}{d} = \frac{-M \pm \sqrt{M^2 + N^2}}{N} \quad (5)$$

$$\tan \phi' = \frac{L}{D} \tan \phi + \tan \beta \quad . \quad (6)$$

This approximate method of analysis may be readily extended to include nonrectangular initial geometry and a real volume change. The latter condition is obtained easily from the general relationship

$$G_1 + G_2 + G_3 = \frac{100 \ln \rho_1 / \rho_2}{B} \quad , \quad (7)$$

of which equation (4) is a particular solution. The nonrectangular case requires introduction of an initial skew angle or angles and results in a considerably more complicated expression than equation (2). In any rigorous development of the analysis an irregular geometry would result in equations which have no exact solution and would require numerical approximation to evaluate.

APPENDIX B

Computation of Dimensional Change Rates

The critical length and diameter data for use in computing growth rates were carefully measured on good photographic prints of negatives taken before and after irradiation. Length measurements were made with the assistance of a traveling microscope. Angle measurements, where possible, were made by triangulation. The photographs were initially intended to provide only a qualitative record of the shape changes, or at best only a rough quantitative estimate of the growth rates. The data obtained from them, therefore, are not as adequate for an accurate analysis as could be desired. The following difficulties were noted:

(a) Preirradiation photographs were aligned with the fiducial marks instead of with approximate crystallographic directions. The angular discrepancies involved are listed in Table I.

Table I

RELATION OF CRYSTALLOGRAPHIC POLES
TO SPECIMEN GEOMETRY

Crystal No.	Approximate Angle Between [010] and Cylindrical Axis (Degrees)	Approximate Angle Between [001] and Fiducial Bevel (Degrees)
A	17	37
B	14	22
C	13	8
Pseudo A, B, C	$\sim 25(^*)$	-

*Scatter of approximately 10° about the mean position.

(b) The preirradiation end photographs show the [100] dimension to be 5% less than the [001]. In other words, the cross sections were elliptical rather than circular. The misalignment of photographs noted above made direct measurements of the desired diameters impossible except on the end views.

(c) End surfaces of Crystal A were not perpendicular to specimen axes before irradiation. This did not create any difficulty in the analysis of the [001] [010] plane, but some uncertainty of correction resulted in the analysis of the [100] [010] due to lack of the proper view.

(d) Although the presence of fiducial marks provided an immediate identification of relative growth rates after irradiation, they also made it difficult to obtain clean-cut dimensions and angular data.

(e) The postirradiation magnifications were not optimum. Better data could have been obtained from larger pictures. However, the shift in magnification was satisfactorily adjusted by normalization, based on the assumption that volume of the specimens did not change significantly during irradiation.

(f) Slight changes in magnification from one print to another or due to difference in focal depth in the same print needed to be recognized and brought to account.

In Table II are summarized the pertinent data obtained from the photographs; only single crystals A and C and pseudo crystal A were measured. The data from Crystal A permit two analyses of different planes, but only one is possible in the case of Crystal C, because one skew angle could not be measured. Only the length change of the pseudo crystal is recorded.

Table II
SUMMARY OF DIMENSIONAL MEASUREMENTS SCALED FROM PHOTOGRAPHS
AND NORMALIZED TO ZERO VOLUME CHANGE

Crystal	Negatives Measured, No. and Magnification		Normalization Factor	Dimensional Measurements ^(a) (cm)			Skew Angle (tan β)				
				[100]	[001]	[010]	[001] - [010]	[100] - [010]			
<u>Crystal A</u>											
Unirradiated	13600	10X	2.4578	2.86 ⁽³⁾	3.02 ⁽³⁾	4.93 ⁽³⁾	0.092 ⁽⁴⁾	0.277 ⁽⁴⁾			
	13602	10X									
Irradiated	14328, -29, -31 and -32	4X		0.734 ⁽⁵⁾	1.235 ⁽⁸⁾	3.164 ⁽⁵⁾					
	Normalized			1.804	3.035	7.776					
<u>Crystal C</u>											
Unirradiated	13597 and -98	10X		3.218 ⁽³⁾	3.37 ⁽³⁾	1.798 ⁽³⁾			1.111 ⁽⁴⁾		
Irradiated	14894 and -96	2X		0.1658 ⁽⁵⁾	0.6572 ⁽⁵⁾	1.508 ⁽⁵⁾					
	Normalized	4.9146		0.8148	3.230	7.411					
<u>Pseudo A</u>											
Unirradiated	Direct Measurement								1.176		
Irradiated	14333	4X				8.56					
	Normalized	0.25				2.14					

(a) Number of measurements averaged is indicated in parentheses.

Accuracy of the growth coefficient computations also depends directly upon accuracy of the burnup determinations. Table III lists the three values assigned to the crystals. Burnups of crystals A were determined by radiochemical analyses, having a probable uncertainty of approximately $\pm 5\%$. Burnup of crystals B, which is of no significance to the analysis, was obtained from a cobalt foil associated with the irradiation

capsule. Unfortunately, the foil associated with crystals C was lost, and the burnup number assigned is an extrapolation from other capsules in the MTR stringer. The uncertainty in this value is probably greater than $\pm 10\%$.

Table III

BURNUP VALUES ASSIGNED TO IRRADIATED CRYSTALS

Crystal No.		Percent Total Atom Burnup	Type of Analysis
Single	Pseudo		
A	A	0.110	Cs-137
B	B	0.27	Co Monitor
C	C	0.37	Estimate

Growth coefficients resulting from treatment of single crystal data are summarized in Table IV. The results for Crystal A have been subjected to an error analysis in which burnup determination is considered to be exact. It will be seen that the two growth rate evaluations for the [010] agree within 1%, and that their probable error ranges are of like magnitude. Considerable more scatter is shown by the error ranges of the [100] and [001] growth coefficients, as might be expected from a study of the ratio given by equation (2) in Appendix A. If the one result recorded for Crystal B is normalized to $G_{[010]} = 420$, a burnup of 0.346 a/o is needed. This differs from the estimate of 0.37 a/o, shown in Table III, by 6.5% and could reasonably be adopted as the correct burnup because of the uncertainty of the tabulated value. In this case the other coefficients obtained from Crystal B would be $G_{[100]} = -410$, and $G_{[001]} = -10$. These values have been taken into account in assigning to $G_{[001]}$ essentially a null effect in the weighted average. If this is done, $G_{[100]}$ becomes equal in magnitude to $G_{[010]}$ and of opposite sign. It is very likely that the realistic range of error for these values is of the same magnitude as the probable error in specimen burnup.

Table IV

SUMMARY OF GROWTH RATE CALCULATIONS
AND ASSIGNED AVERAGE VALUES

Plane Analyzed	Growth Coefficient, G_i		
	[100]	[010]	[001]
<u>Crystal A</u>			
[001] [010] ^(a)	-442 ± 30	417 ± 1	25 ± 28
[100] [010] ^(a)	-426 ± 13	420 ± 4	$[6 \pm 16]$
<u>Crystal B</u>			
[001] [010]	$[-384]$	393	-9
Tentative assigned Weighted average(b)	-420 ± 20	420 ± 20	0 ± 20

(a) Indicated error ranges are based on analysis of the dimensional data only, assuming the burnup to be exactly known.

(b) Error assigned for the [010] direction is based on an uncertainty of 5% in the burnup analysis of Crystal A.