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Metallurgy and Ceramics

AEC Research and Development Report

QUANTITATIVE CORRELATION OF IRRADIATION GROWTH WITH PREFERRED ORIENTATION IN URANIUM

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W. R. McDonell

Pile Materials Division

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E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, South Carolina

Printed in USA. Price $1.00
Available from the Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.

255111

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(TID-4500, 15th Ed.)

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Printed for
The United States Atomic Energy Commission
Contract AT(07-2)-1

Approved by
P. H. Permar, Research Manager
Pile Materials Division

ABSTRACT

A review is presented of the methods developed at the Savannah River Laboratory and elsewhere for quantitative correlation of irradiation growth with preferred orientation in uranium, with preferred orientation characterized by X-ray and dilatometric techniques. In an application of the methods, it is shown that a strongly oriented uranium plate, clad in aluminum, was much more stable in dimensions than would be expected from the observed dimensional behavior of either unrestrained single crystals of uranium or aluminum-clad polycrystalline rod. The greater stability of the plate is attributed to a relatively strong cladding and other restraining effects.

CONTENTS

LIST OF TABLES AND FIGURES

QUANTITATIVE CORRELATION OF IRRADIATION GROWTH WITH PREFERRED ORIENTATION IN URANIUM

INTRODUCTION

Uranium that has a preferred crystallographic orientation is, in general, dimensionally unstable during reactor irradiation. This instability, or "irradiation growth", is due to the behavior of the alpha uranium crystal, which, under irradiation, elongates in the direction of the b-axis and shortens in the direction of the a-axis while remaining approximately constant in the direction of the c-axis.[1] Since control of the "irradiation growth" of uranium is of great importance in the operation of nuclear reactors, a quantitative correlation of irradiation growth with the type and degree of preferred orientation would be very useful.

This report considers the correlations between preferred orientation and irradiation growth developed in this Laboratory and elsewhere through early 1958, with preferred orientation characterized by X-ray and dilatometric techniques. The correlations provide a means for systemization of the effects of significant structural and irradiation variables on the dimensional stability of uranium under irradiation, and serve as essential background for the present and future dimensional stability studies of this Laboratory. An application of the derived correlations to the dimensional behavior of a representative uranium specimen is presented, to substantiate the correlations and to illustrate the utility of the analysis in isolating the effects of preferred orientation from that of several other variables, such as specimen geometry, cladding restraint, and irradiation time and temperature, which may also influence the dimensional stability of uranium.

SUMMARY

Several methods are reviewed for correlating preferred orientation and irradiation growth in uranium. By means of these methods values of γ_b, the effective irradiation growth of an incremental volume of metal in the b-axis direction, are derived for two types of irradiated specimens. The values of γ_b that are obtained are compared with a value of 420% elongation/atom % burnup measured for unrestrained single crystals at Argonne National Laboratory.

The diffraction technique of the Savannah River Laboratory utilizes an X-ray orientation parameter, or "growth index", to correlate preferred orientation with irradiation growth. A value of γ_b = 114% elongation/atom % burnup, is derived for a highly textured specimen in the form of hot-press-bonded, aluminum-clad uranium plate.

An analagous diffraction technique that employs an alternative orientation parameter (Φ) was developed by Barss, of Atomic Energy of Canada, Limited. With this technique, a value of γ_b = 610% elongation/atom % burnup was obtained for hot-rolled rod sheathed in aluminum.

The dilatometric technique of the Savannah River Laboratory makes use of an "orientation coefficient" that is obtained by measuring the thermal expansion just following a reversal in temperature. This procedure avoids extraneous deformations due to grain interaction effects. A value of γ_b = 120% elongation/atom % burnup is obtained by the dilatometric technique for the aluminum-clad plate used in SRL X-ray correlation.

In these comparisons, the γ_b values for the aluminum-clad plate derived by the X-ray and dilatometric techniques of SRL are in good agreement, but the average value of 117% elongation/atom % burnup is considerably less than the value of 420 observed for unrestrained single crystals, as well as the value 610 derived by the AECL technique for aluminum-clad rod. This difference is ascribed largely to the restraint imposed on the uranium plate by a relatively stronger aluminum cladding.

DISCUSSION

BACKGROUND

IRRADIATION GROWTH OF URANIUM SINGLE CRYSTAL

Uranium in the alpha phase has an orthorhombic crystal structure. Single crystals exhibit strong anisotropies in several properties, including mechanical properties and thermal expansion coefficients, that may give rise to dimensional instability under irradiation.[1] Unrestrained single crystals have been observed at Argonne National Laboratory to elongate with an irradiation growth coefficient, G_i*,

* The irradiation growth unit, G_i, which can be applied either to single crystals or polycrystalline specimens, is defined as follows:

$$G_i = \frac{\ln(L/L_o)}{N_f/N_t}$$

in which L/Lo is the ratio of final to initial dimensions of the specimen, and N_f/N_t is the ratio of fissioned atoms to total atoms.[1] At low growths, G_i may be approximated by the relation:

$$G_i = \frac{(L - L_o)/L_o}{N_f/N_t} = \frac{\text{Per cent growth}}{\text{Atom per cent burnup}}$$

equal to 420 ±20 in the (010) direction (b-axis), to shorten by the same amount in the (100) direction (a-axis), and to show no growth in the (001) direction (c-axis), during irradiations in the range of 0.1 to 0.3% burnup of total uranium atoms at less than 150°C.[1] Experiments at Harwell are in general agreement with these results.[2]

The dimensional behavior of uranium is markedly dependent upon the conditions under which irradiation occurs, especially with regard to temperature and external restraint. Irradiation growth is at a maximum in the temperature range 100 to 300°C; at very low temperatures, -178°C, growth is greatly reduced; and at very high temperatures, 500°C, growth is negligibly small.[1,2,3] External restraint due to cladding or rigid containers may inhibit markedly the growth of uranium.[4] The effect of neutron flux level on irradiation growth has not been characterized.

IRRADIATION GROWTH OF POLYCRYSTALLINE URANIUM

A polycrystalline specimen with preferred orientation, or "texture", exhibits more or less the same dimensional instability as a uranium single crystal, modified to some extent by intergranular interactions.[1] Qualitative correlations of irradiation growth with the type and degree of preferred orientation have been well established. Specimens with predominantly b-axis textures lengthen on irradiation, while specimens with a-axis textures shorten, in agreement with single crystal behavior, and combinations or intermediate textures show intermediate behavior.[5,6] Moreover, changes in preferred orientation due to the irradiation per se have been noted, with the result that specimens that grow during irradiation may in some cases do so at an increasing rate, and specimens that contract may do so at a decreasing rate. However, these changes were observed principally in specimens with well-developed duplex textures; dimensionally stable specimens and specimens with predominantly single textures showed only minor texture changes during irradiation.[6]

Only a few quantitative correlations of preferred orientation with irradiation growth in polycrystalline specimens had been attempted at the time the Savannah River work was initiated. In early work, Barss showed a linear relationship between irradiation growth of wrought uranium rods and a complex preferred orientation parameter derived from diffraction data.[7] This correlation will be considered in detail later in this report and will be compared with the results of analogous Savannah River techniques. In recent work, Shupe, Cummings, and Watts demonstrated a quantitative correlation between irradiation growth of wrought rod specimens and the relative diffraction intensities of a few predominant orientations in the metal.[6] Chiswik demonstrated an empirical correlation between irradiation growth of small cylindrical specimens and their thermal expansion coefficients.[8]

The effect of grain size and intergranular interactions on the dimensional behavior of polycrystalline uranium is not well defined. Present evidence suggests that in moderately oriented specimens the effect of grain size is not large,[8,9] though highly oriented polycrystals and psuedo-single crystals have been observed to grow more than twice as much as true single crystals.[5,6,10] Such grain interaction effects may be dependent on their relative orientations.[6] Coarse-grained uranium does show severe surface roughening on irradiation due to anisotropic growth of individual grains or grain clusters.[1,2]

QUANTITATIVE CORRELATION OF IRRADIATION GROWTH WITH PREFERRED ORIENTATION

DIFFRACTION METHOD OF SAVANNAH RIVER LABORATORY

In previous publications, the "texture coefficient", defined in equation (1), and the diffraction "growth index", defined in equation (5c), have been designated (TC) and (GI), respectively. For convenience in mathematical manipulations, the symbols T and Δ have been substituted in this report.

Theory of SRL Diffraction Method

The X-ray method of the Savannah River Laboratory for measurement of preferred orientation is derived from the "rho technique", as originated by Harris[11] and modified by Mueller[12] and Morris[13]. In this technique, the relative amount of metal in a given orientation* (hkl) with respect to a reference direction, d, in the specimen, is approximated by a "texture coefficient", $T_{d(hkl)}$, which is defined in equation (1).

$$T_{d(hkl)} = \frac{I_{(hkl)}/I_{o(hkl)}}{\frac{1}{n}\sum I_{(hkl)}/I_{o(hkl)}}$$

In this equation, $I_{(hkl)}$ and $I_{o(hkl)}$ are the diffracted intensities of the planes (hkl) in the textured specimen and in a randomly oriented specimen, respectively, as measured on a cross section perpendicular to the reference direction in the specimen; n is the number of planes surveyed to characterize the specimen.

* For characterization of its irradiation growth in a given direction the orientation of an increment of metal may be described by the designation of the plane (hkl), which lies perpendicular to the given direction.

To correlate the type and degree of preferred orientation with the dimensional behavior of the specimen under irradiation, it is assumed that the irradiation growth, γ_d, of an incremental volume of metal with orientation (hkl) with respect to the reference direction, d, is given by equation (2).*

$$\gamma_d = \gamma_a \cos^2\theta_{a(hkl)} + \gamma_b \cos^2\theta_{b(hkl)} + \gamma_c \cos^2\theta_{c(hkl)} \qquad (2)$$

In this equation γ_a, γ_b, and γ_c are effective irradiation growth coefficients in the a-, b-, and c-axis directions of the alpha uranium crystal, and θ_a, θ_b, and θ_c are the direction angles which the a-, b-, and c-axis make in orientation (hkl) with the reference direction, d.

Summing over the contributions of all possible orientations (hkl) yields the irradiation growth coefficient, G_i, of the specimen in the reference direction, d, as indicated in equation (3).

$$G_i = \sum \left[\left(\frac{\Delta V}{V}\right)_{(hkl)} \gamma_a \cos^2\theta_{a(hkl)} + \left(\frac{\Delta V}{V}\right)_{(hkl)} \gamma_b \cos^2\theta_{b(hkl)} + \left(\frac{\Delta V}{V}\right)_{(hkl)} \gamma_c \cos^2\theta_{c(hkl)} \right] \qquad (3)$$

Assuming that the relative volume of metal in orientation (hkl) is representative of that in more or less closely lying orientations, the relative volume, $\left(\frac{\Delta V}{V}\right)_{(hkl)}$, in any given orientation (hkl) is given by the measured texture coefficient, $T_{d(hkl)}$, as shown in equation (4).

$$\left(\frac{\Delta V}{V}\right)_{(hkl)} = \frac{T_{d(hkl)}}{n} \qquad (4)$$

Substituting this relation and the empirically observed relations $\gamma_c = 0$, $\gamma_b = -\gamma_a$ in equation (3) yields equation (5a), or alternatively, equation (5b).

$$G_i = \frac{\gamma_b}{n} \sum^{(hkl)} T_{d(hkl)} (\cos^2\theta_b - \cos^2\theta_a)_{(hkl)} \qquad (5a)$$

* This equation, analogous to that governing the thermal expansion behavior of anisotropic materials, is a specification of the orientation dependence of an anisotropic homogeneous deformation.[14]

$$G_i = \frac{\gamma_b}{n}\left[\sum^{(hkl)} (T_d\cos^2\theta_b)_{(hkl)} - \sum^{(hkl)} (T_d\cos^2\theta_a)_{(hkl)}\right] \quad (5b)$$

The summation terms may be designated the diffraction "growth index", Δ, yielding equation (5c).

$$G_i = \frac{\gamma_b}{n}\,\Delta \text{ where } \Delta = \sum^{(hkl)} (T_d\cos^2\theta_b)_{(hkl)} - \sum^{(hkl)} (T_d\cos^2\theta_a)_{(hkl)} \quad (5c)$$

Growth Index Based on Fourteen Measured Planes

Utilization of equation (5c) for the correlation of preferred orientation and irradiation growth presents certain difficulties due to the unsymmetrical distribution of the measured orientations with respect to their a- and b-axis contributions.[15] In routine practice, the intensities of fourteen planes are measured; their distribution is shown in Figure 1.[15] It must be assumed that these planes represent an adequate sampling of all possible orientations in the specimen to obtain a value for the volume of metal in a given orientation according to equation (1). However, the unsymmetrical distribution of measured planes yields a value for $\sum\cos^2\theta_{b(hkl)}$ greater than $\sum\cos^2\theta_{a(hkl)}$, as shown in Table I, with the result that the predicted

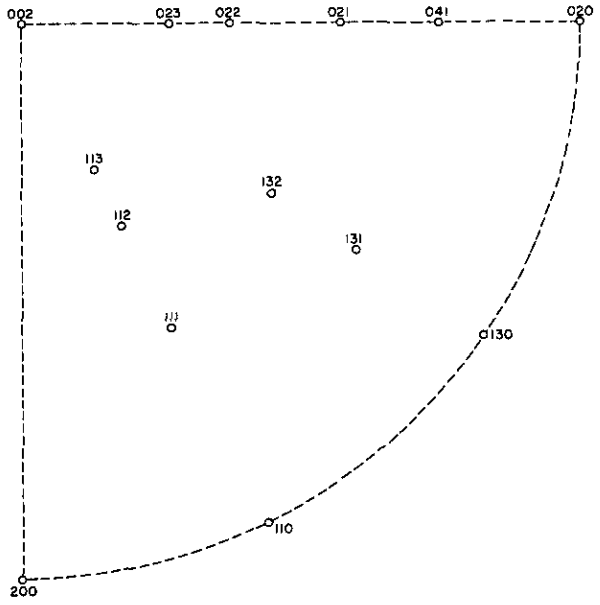


FIGURE 1 – STEREOGRAPHIC PROJECTION OF PRINCIPAL PLANES IN ALPHA URANIUM CRYSTAL (From Reference 15)
The intensities of the fourteen planes indicated are measured in the SRL procedure to characterize preferred orientation. Note the predominance of planes with large b-axis (020) contributions.

TABLE I

Weight Factors for Measured Diffraction Planes (SRL Method)
(From Reference 15)

Plane (hkl)	$\cos^2\theta_a$	Plane (hkl)	$\cos^2\theta_b$
200	1.00	020	1.00
110	0.81	041	0.92
111	0.64	021	0.74
112	0.39	130	0.68
130	0.32	131	0.62
131	0.29	132	0.48
113	0.24	022	0.42
132	0.23	023	0.24
020	0.00	110	0.19
041	0.00	111	0.15
021	0.00	112	0.09
002	0.00	113	0.06
022	0.00	200	0.00
023	0.00	002	0.00

$$\sum \cos^2\theta_{a(hkl)} = 3.92 \qquad \sum \cos^2\theta_{b(hkl)} = 5.59$$

growth of a randomly oriented specimen is not zero.[15] In one means to overcome this difficulty, selected planes may be chosen for the summation of a- and b-axis components respectively, so as to yield a sample for which $\sum \cos^2\theta_a = \sum \cos^2\theta_b$. The planes thus chosen for summation of each component are indicated in Table II.[15]

TABLE II

Selection of Diffraction Planes to Yield Balanced Weight Factors (SRL Method)
(From Reference 15)

Plane (hkl)	$\cos^2\theta_a$	Plane (hkl)	$\cos^2\theta_b$
200	1.00	020	1.00
110	0.81	041	0.92
111	0.64	021	0.74
112	0.39	130	0.68
130	0.32	023	0.24
131	0.29	110	0.19
113	0.24	112	0.09
132	0.23	113	0.06

$$\sum \cos^2\theta_a = \sum \cos^2\theta_b = 3.92$$

Growth Index Based on Fifty-five Synthetic Planes

An alternative procedure can be used to minimize the formal effects of the lack of symmetry of the originally measured planes. In this procedure, the intensity distribution of the planes is mapped on an inverse pole chart, to allow interpolation of the intensities of a large group of planes that are arbitrarily selected to yield a symmetrical distribution. The intensities of the measured planes are used to construct the map. In this procedure, the inaccuracies that derive from the lack of symmetry of the necessarily small sampling are avoided, but are replaced by inaccuracies that derive from the interpolations necessary to ascribe intensity values to nonmeasured planes.

Application of the two procedures is discussed in the following section.

Correlation with Observed Irradiation Growth

The validity of the formalism for the prediction of irradiation growth was tested by comparison of the orientation parameters, as determined by the above procedures, with the observed growth of several types of uranium specimens. The most precise data available were obtained from experimental uranium plate specimens, irradiated in the MTR;[16] this data will be used to illustrate the correlations derived above. The plate, taken from strip formed by rolling in the high alpha phase, had a moderately strong cubic-type preferred orientation, with (010) planes perpendicular to the length (rolling) direction, (001) planes perpendicular to the width direction, and (100) planes perpendicular to the thickness direction.[16] Irradiation specimens, 18 inches long, 3 inches wide, and 0.180 inch thick were clad in 0.022-inch-thick aluminum, with a bond layer of electroplated nickel. On irradiation to 600 MWD/T (0.077 atom % burnup) at 50 to 150°C in the MTR, the plate specimens changed dimensions as follows: length +4.6%, width -0.8%, and thickness -3.7%.[16]

The preferred orientation of the plate was characterized by measurements of "texture coefficient" on sections cut perpendicular to the length, width, and thickness directions (Table III).[15] This data was used to calculate an orientation parameter for the corresponding directions in the plate by the fourteen-plane method. The values obtained, along with the measured dimensional changes, are shown in Table IV.[15] A plot of irradiation growth versus the orientation parameter is nearly linear, as indicated in Figure 2. A value for γ_b = 114% elongation/atom % burnup, considerably less than the single crystal value of 420, is obtained from the slope of the curve.

TABLE III

Characterization of Preferred Orientation in As-Rolled Uranium Plate
(From Reference 15)

Plane (hkl)	Texture Coefficients		
	Longitudinal	Transverse	Normal
020	0.06	5.25	0.22
110	0.71	1.20	1.52
021	0.32	0.26	0.26
002	5.14	0.01	0.36
110	0.46	0.22	1.20
022	1.07	0.00	0.64
112	1.60	0.05	0.77
130	0.00	2.53	0.61
131	0.40	1.07	0.71
023	1.41	0.01	0.44
200	0.08	0.46	6.14
041	0.06	2.78	0.27
113	2.84	0.00	0.45
132	0.40	0.15	0.41

TABLE IV

Preferred Orientation and Dimensional Instability of As-Rolled Uranium Plate

Direction	Orientation Index(Δ)		Irradiation Growth Coefficient*, G_i
	14-Plane Method	55-Plane Method	
Length	+7.21	+7.38	60
Width	-1.00	-1.45	-11
Thickness	-7.49	-6.75	-47

* Plate irradiated to 0.077% burnup of total atoms at 50-150°C[16]

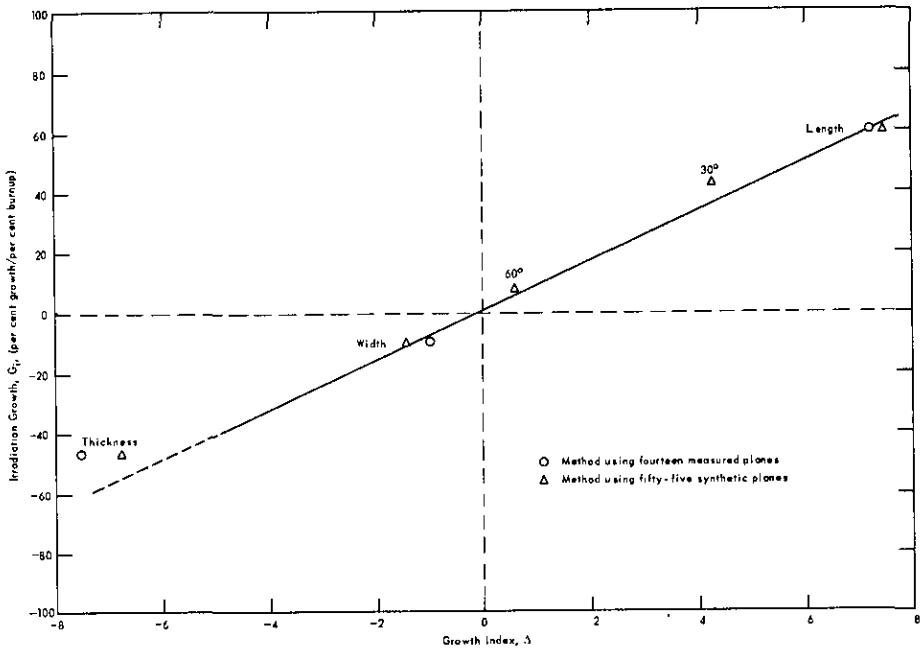


FIGURE 2 – CORRELATION OF GROWTH INDEX, Δ, WITH IRRADIATION GROWTH OF AS-ROLLED URANIUM PLATE

Correlations are shown for length, width, and thickness directions and for directions 30 and 60° from the length direction. The as-rolled plate was irradiated to 600 MWD/T (0.077 atom % burnup) at 50-150° C in the MTR. Note that use of the alternative procedure (fifty-five synthetic planes) for calculation of Δ produces only minor changes in the correlation values derived by the fourteen-plane procedure.

Application of the fifty-five-plane method for overcoming the difficulties introduced by nonsymmetrical distribution of the measured planes required that the intensities of an arbitrarily chosen set of symmetrically distributed planes be derived from a map of the plane intensities over all possible orientations. Such maps for the length, width, and thickness directions of the as-rolled plate are shown in Figure 3, as constructed from the intensities of the measured planes.

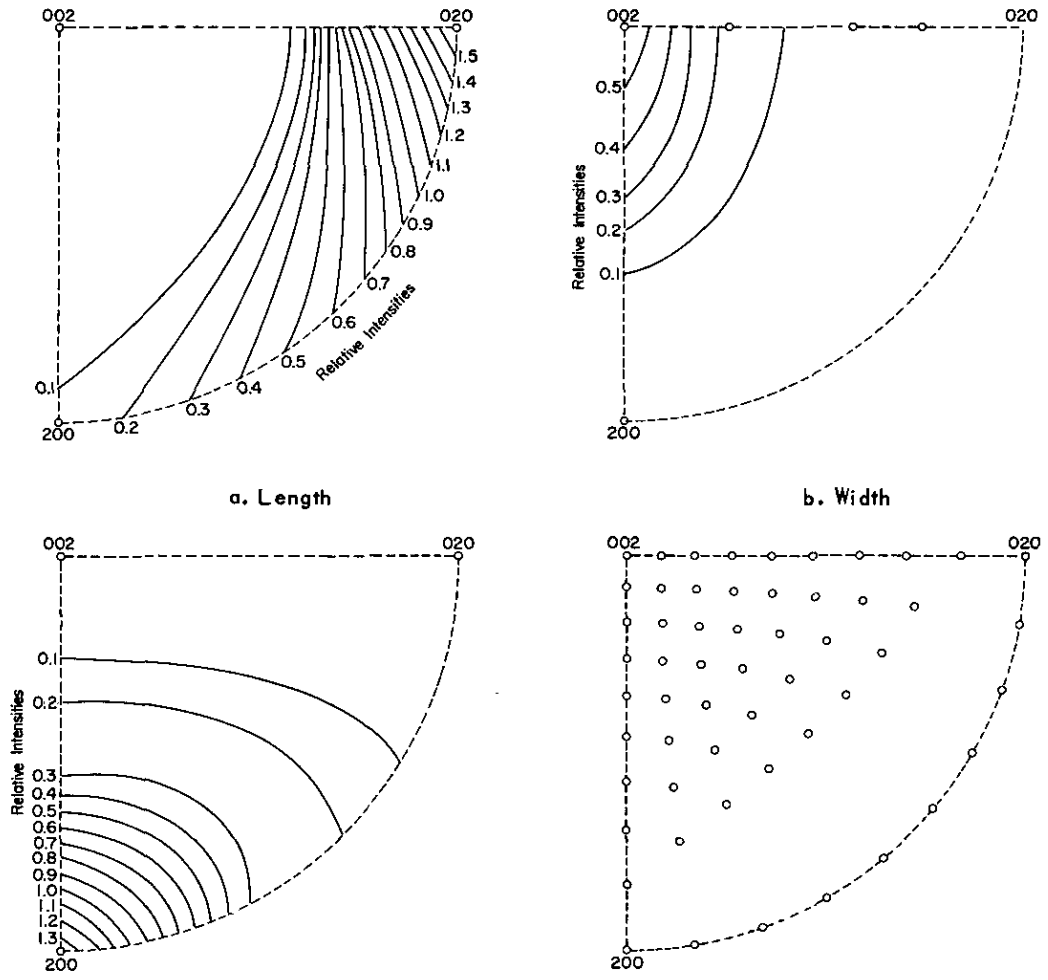


FIGURE 3 – INTENSITY DISTRIBUTION OF PLANES IN AS-ROLLED URANIUM PLATE
The intensities of planes perpendicular to the length, width, and thickness directions in the plate are mapped on inverse pole charts in a, b, and c. In d is shown a symmetrical distribution of fifty-five synthetic planes at 10° intervals used for the alternative calculation of the orientation parameter, Δ.

In this alternative calculation, fifty-five planes were chosen at 10° intervals over the pole chart quadrant (Figure 3d), and the intensities of the corresponding orientations were taken from the pole chart maps. The orientation parameters that were obtained, normalized to conform with the earlier calculations, are shown in Figure 2 for comparison with those obtained using the standard method. It is apparent that, for the specimen characterized, the possible unsymmetries involved in the balanced set of fourteen measured planes induce only small discrepancies from the values calculated using the completely symmetrical set of fifty-five synthetic planes.

A similarly derived correlation between the irradiation growth and preferred orientation for directions in the plane of the plate at 30° and 60° from the length direction is indicated in Figure 2. The diffraction data were taken from plate specimens that were formed similar to, but not identical to, the irradiated plate. Dimensional changes, ΔD, in the 30° and 60° directions were calculated from the measured dimensional changes, ΔL and ΔW, in the length and width directions, respectively, using the relation:

$$\Delta D_\theta = \Delta L \cos^2\theta + \Delta W \sin^2\theta; \quad \theta = 30^\circ \text{ or } 60^\circ \qquad (6)$$

Though use of this relation assumes an effectively perfect alignment of b- and c-axes with the length and width of the plate, the resulting points do not deviate greatly from the principal curve of Figure 2.

These results of the SRL diffraction method may now be compared with alternative techniques - the AECL diffraction method and an SRL dilatometric method.

AECL DIFFRACTION METHOD

Procedure and Theory

An alternative diffraction technique was developed by Barss of the AECL to quantitatively characterize preferred orientation in uranium.[7] In this technique the intensity of a given plane, usually (020), is measured as a function of angle Φ from the reference direction, with X-ray or neutron diffraction techniques on appropriate spherical or cylindrical specimens. The intensities of the measured planes at a given angle Φ are averaged over all angles of rotation about the reference direction by rotation of the specimen during measurement, or, in specimens from rod metal, by assuming a radially constant texture. Under these conditions the relative amount of metal oriented with the measured plane (hkl) at an angle Φ to the reference direction is given by $I_{(hkl)} \sin\Phi_{(hkl)}$.

In a formalism analogous to that outlined above, the irradiation growth, γ_d, in the reference direction, d, of an incremental volume of metal

in an orientation such that the b-axis is inclined at an angle Φ_b to the reference direction is expressed by equation (6).[7]

$$\gamma_d = \gamma_b - \gamma_b(1 + A) \sin^2\Phi_b \tag{6}$$

In this equation, A is a measure of the relative contribution of the a-axis in the reference direction, as compared with the contribution of the c-axis, and may vary from 0 to 1*.

The volume fraction of metal in orientation Φ_b is given by equation (7).

$$\left(\frac{\Delta V}{V}\right)_{\Phi_b} = \frac{I_{(020)} \sin\Phi_b \, d\,\Phi_b}{\int_0^{\pi/2} I_{(020)} \sin\Phi_b \, d\,\Phi_b} \tag{7}$$

Weighting the irradiation growth of an incremental volume in orientation Φ_b by the amount of metal in this orientation and integrating over all orientations yield the net growth, G_i, of the specimen in the reference direction, d, as shown in equation (8a).

$$G_i = \gamma_b - \gamma_b(1 + A)\Phi_{(020)};$$

$$\Phi_{(020)} = \frac{\int_0^{\pi/2} I_{(020)} \sin^3\Phi_b \, d\,\Phi_b}{\int_0^{\pi/2} I_{(020)} \sin\Phi_b \, d\,\Phi_b} \tag{8a}$$

For the purposes of the integration, A is assumed constant over all values of Φ_b. In the final equation (8a) then, A represents an effective contribution of a-axis orientations to the irradiation growth in the reference direction as compared to the contribution of the c-axis orientations.**

---

* The symbol A represents the value of $\cos^2\Phi'_a$ where Φ'_a is the angle between the a-axis of the crystal and the perpendicular to the b-axis in the plane defined by the b-axis and the reference direction.

** E. F. Sturcken has pointed out that a complete characterization of the preferred orientation of the specimen may be obtained using the AECL technique by actual measurement of the intensity of a-axis orientations, $\Phi_{(200)}$, as a function of angle Φ. The orientation parameter obtained is identical to the parameter, Δ, derived by the SRL technique, and is given in equation (8b) below, where $\Phi_{(200)}$ is defined analogously to $\Phi_{(020)}$ in equation (8a).

$$\Delta = \frac{\Phi_{(200)} - \Phi_{(020)}}{n} \tag{8b}$$

Correlation with Observed Irradiation Growth

The AECL diffraction technique was applied to rod irradiated in the NRX to obtain values for $\Phi_{(020)}$ for comparison with the observed irradiation growth of the rod.[7] A least squares fit to the data produced the correlation shown in Figure 4, where A = 0.6, in agreement with the reported tendency for the c-axis to take somewhat preferentially a direction perpendicular to the rolling direction in hot-rolled rod.[7] The value of γ_b, derived from the intercept at $\Phi = 0$, is equal to 610% per atom % burnup, somewhat greater than the single crystal value, and much greater than the value obtained for as-rolled plate using the SRL diffraction technique. This discrepancy will be further considered in the comparison of results in a later section.

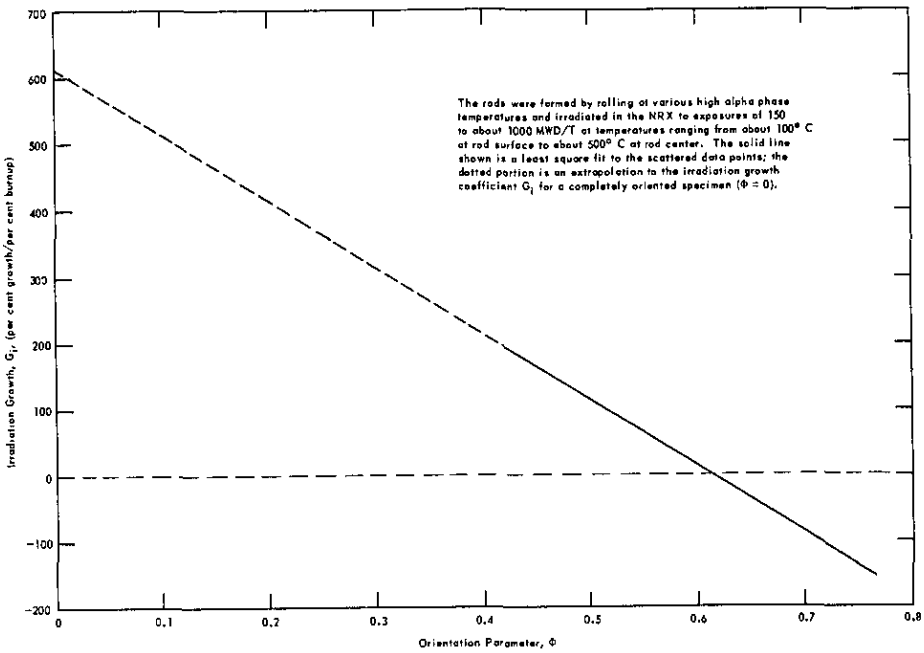


FIGURE 4 - CORRELATION OF AECL ORIENTATION PARAMETER, Φ, WITH IRRADIATION GROWTH OF URANIUM RODS
(From Reference 6)

THERMAL EXPANSION METHOD OF SAVANNAH RIVER LABORATORY

Procedure and Theory

The large anisotropy of thermal expansion of the alpha uranium crystal,[17] illustrated in Figure 5, makes the thermal expansion coefficient of a uranium specimen a sensitive measure of its preferred orientation. Thermal expansion in the b-axis direction of the crystal is considerably lower than in the a- or c-axes directions, and, thus, measured coefficients lower than the mean value of the three crystallographic axes (about 15 x 10^{-6}/°C at room temperatures) indicate an (010) type texture that will elongate on irradiation.

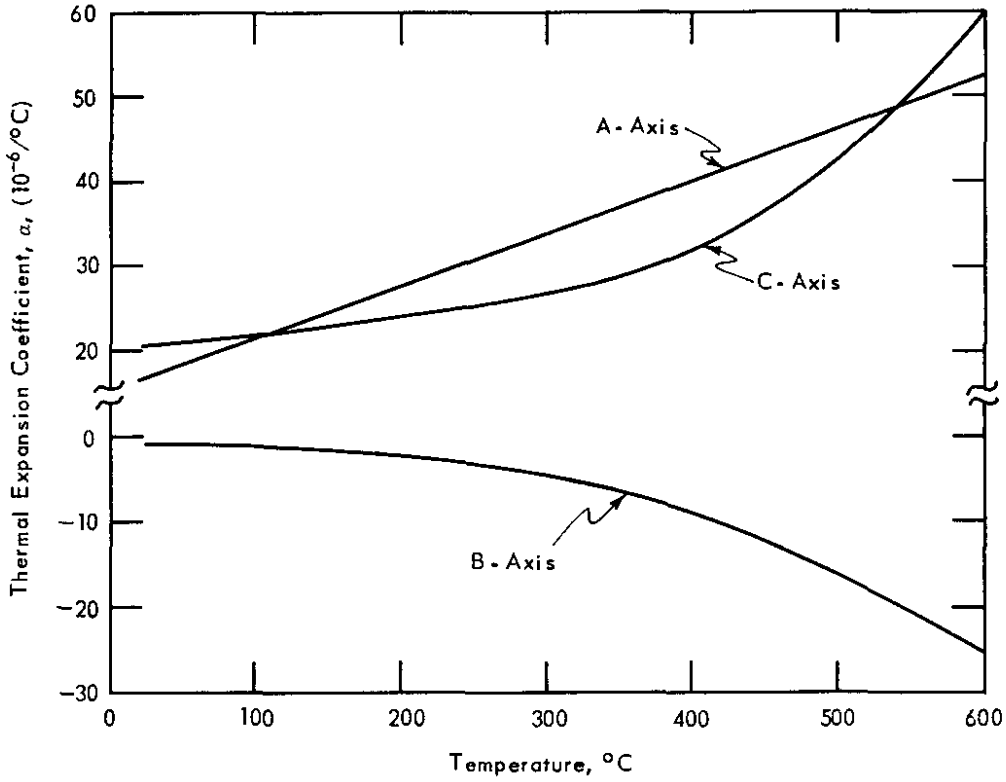
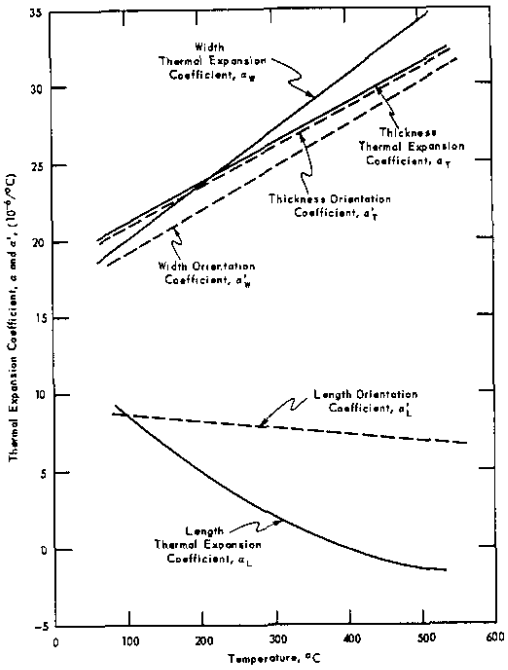


FIGURE 5 – THERMAL EXPANSION COEFFICIENTS OF CRYSTALLOGRAPHIC AXES OF ALPHA URANIUM

The curves were derived graphically from X-ray data of Schwartz and Vaughn[17]

Quantitative characterization of such textures using thermal expansion coefficients is complicated by extraneous deformations of a specimen during heating or cooling that are due to mechanical interactions of neighboring grains of differing orientations.[18] As neighboring grains expand differentially, the stresses that are built up between them may result in plastic deformation of one or the other, an effect that is reflected in a change in the apparent thermal expansion coefficient of the specimen. In a highly oriented specimen, such as the as-rolled plate considered previously, the apparent thermal expansion coefficient obtained during simple heating may differ markedly from that characteristic of the net thermal expansions of the individual grains, as indicated in Figure 6. For analogous reasons, the thermal expansion behavior of a polycrystalline uranium specimen may be markedly dependent upon its previous mechanical and thermal history, to an extent much greater than would be anticipitated from the changes in preferred orientation that may be produced by the



The large difference between length and width or thickness directions indicates the strong preferred orientation of the plate. The dotted lines designate "orientation coefficients" that are obtained just following a reversal in temperature and represent approximately the thermal expansion of the specimen independent of plastic deformation effects produced by interactions between grains of differing orientations.

FIGURE 6 - THERMAL EXPANSION COEFFICIENTS OF AS-ROLLED URANIUM PLATE

working or heating operation.[19] This is illustrated in Figure 7, which shows the large change in thermal expansion coefficient that may result on annealing of a mildly cold-worked specimen under conditions where recrystallization effects, and thus preferred orientation changes, appear negligible. Thus, the measured thermal expansion coefficient cannot, in general, be used for quantitative comparisons of the degree of preferred orientation in specimens of different mechanical or thermal history.

These difficulties may in some measure be overcome by a technique in use at SRL that is designed to yield a coefficient of expansion that is uniquely dependent upon the net preferred orientation of the specimen.[19] This technique involves measurement of the thermal expansion of the specimen just following a reversal in temperature. Under these conditions, the stresses between neighboring grains that produce plastic flow are relaxed, allowing the specimen to deform largely in accord with the net thermal expansion of its grains, and perturbed only by small elastic readjustments that are in themselves functions of the preferred orientation of the specimen. In practice, secondary effects not subject to precise interpretation may be encountered in some cases, but generally a coefficient can be obtained that appears to be a consistent representation of the preferred orientation of specimens under several differing mechanical and thermal conditions.[19,20] The "orientation coefficients" shown in Figure 6, taken as indicative of the preferred orientation of the as-rolled plate, were obtained in this fashion.

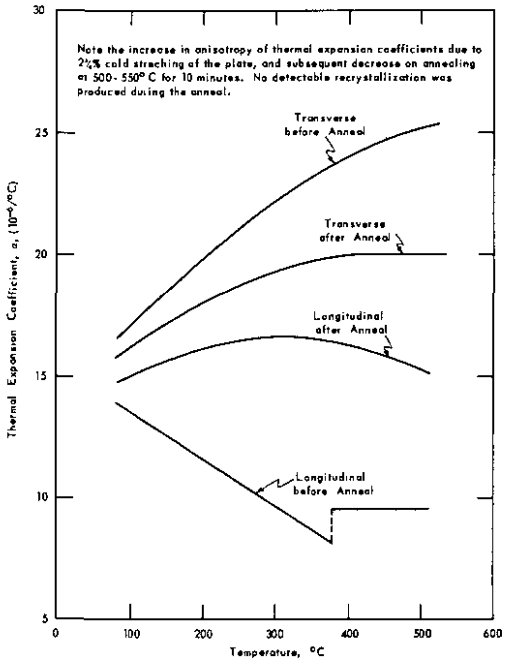


FIGURE 7 - EFFECT OF COLD WORK ON THERMAL EXPANSION COEFFICIENTS OF BETA-TREATED URANIUM PLATE

It does not appear possible to establish analytically a unique correspondence between the thermal expansion coefficients (or "orientation coefficient") of an arbitrary specimen and its corresponding irradiation growth. However, comparisons between specimens with qualitatively similar preferred orientation often can be employed to predict dimensional stability. An example of this is afforded by the comparisons made by Chiswik of thermal expansion coefficient and dimensional stability of rod specimens that were rolled at 300°C to various reductions.[8] A plot of the data obtained yields a smooth curve, as shown in Figure 8, which can be used to predict the stability of the specimens formed in a similar manner.

A more detailed consideration of the relationship between thermal expansion and irradiation growth can be made when the qualitative features of the preferred orientation of a specimen can be established by alternative techniques, such as the X-ray characterization described in a previous section. In analogy with the considerations of Barss, as previously described, the irradiation growth, γ_d, and thermal expansion coefficient, α_d, of an increment of metal with the b-axis at angle θ_b to the reference direction d may be described by equations (9a) and (9b), respectively.

$$\gamma_d = \gamma_b - \gamma_b(1+A)\sin^2\theta_b \qquad (9a)$$

$$\alpha_d = \alpha_b + [A\alpha_a + (1-A)\alpha_c - \alpha_b]\sin^2\theta_b \qquad (9b)$$

In these equations, A and 1-A are the relative contributions of the a- and c-axes orientations respectively to irradiation growth and thermal expansion in the reference direction. The constant γ_b is again the growth of the incremental metal in the b-axis direction, and α_a, α_b, and α_c are thermal expansion coefficients in the directions of the a-, b-, and c-axes, respectively. Combining these equations yields a relationship between irradiation growth, G_i, of the metal in reference direction d, and the thermal expansion coefficient in that direction, as indicated by equation (10a).

$$G_i = \gamma_d = \gamma_b + \frac{(A+1)\alpha_b\gamma_b}{A\alpha_a + (1-A)\alpha_c - \alpha_b} - \frac{(A+1)\gamma_b}{A\alpha_a + (1-A)\alpha_c - \alpha_b}\alpha_d \quad (10a)$$

This equation is most appropriately applied to a direction, d, in which b-axis orientations make a large contribution. In a direction d', in which a-axis orientations are predominant, an analogous equation (10b) may be most conveniently applied.

$$G_i = \gamma_{d'} = -\gamma_b - \frac{(B+1)\alpha_a\gamma_b}{B\alpha_b + (1-B)\alpha_c - \alpha_a} + \frac{(B+1)\gamma_b}{B\alpha_b + (1-B)\alpha_c - \alpha_a}\alpha_{d'} \quad (10b)$$

In this equation, B and (1-B) are the relative contributions of the b- and c-axes orientations, respectively, to irradiation growth in the reference direction.

In the derivation of these equations, interaction effects between grains during irradiation growth have again been neglected.

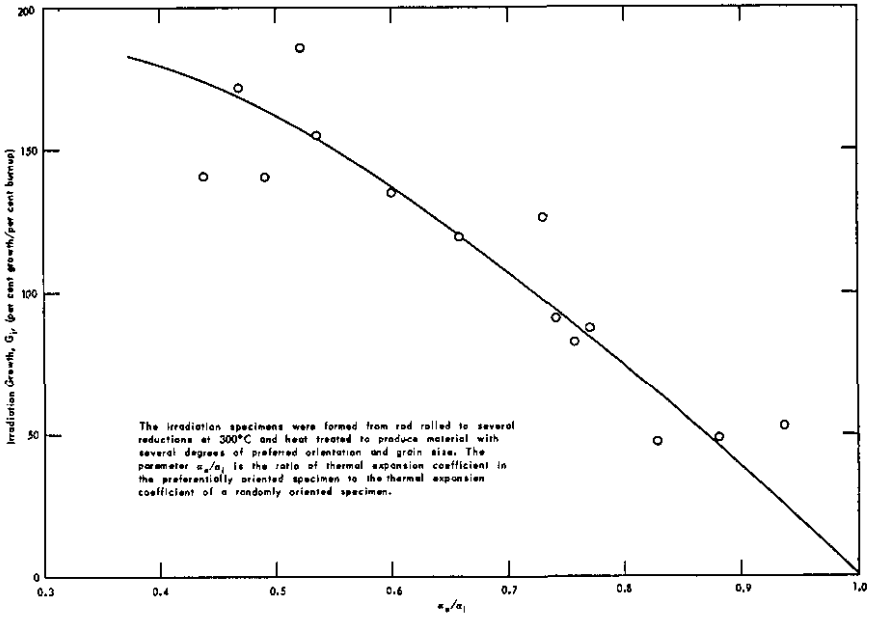


FIGURE 8 - CORRELATION OF THERMAL EXPANSION COEFFICIENTS AND IRRADIATION GROWTH OF ROLLED URANIUM ROD (ANL)
(From Reference 8)

Correlation with Observed Irradiation Growth

Comparison of the above equations with empirical results can be made by reference to the as-rolled plate considered in an earlier section. Thermal expansion coefficients, as well as orientation coefficients obtained for this plate by the cycling technique, are shown in Figure 6, for the length, width, and thickness directions in the plate. Plots of the orientation coefficients at 300°C versus the observed irradiation growth in the corresponding direction (Table IV) yield the curves shown in Figure 9, which correspond to various values of A and B in equations (10a) and (10b).* The coefficients at zero irradiation growth were derived from these equations for each value of A or B by setting G_1 equal to zero. The best values of A and B for the as-rolled plate may be approximated by comparison of the relative amount of metal in predominantly a- or b-axes orientations with that in predominantly c-axis orientations, as given in the data of Figure 3, which yields $A \tilde{=} 0.7$ for the contribution of the a-axis orientations in the length direction and $B \tilde{=} 0.5$ for the contribution of the b-axis orientations in the thickness direction. Corresponding to these values of A and B are values of γ_b equal to 140 and 100% growth per atom % burnup for length and thickness directions, respectively, obtained by extrapolation to single crystal values of the thermal expansion coefficients. The average value of γ_b equal to 120 agrees with the analogous value obtained exclusively from X-ray data and is again considerably lower than the single crystal value.

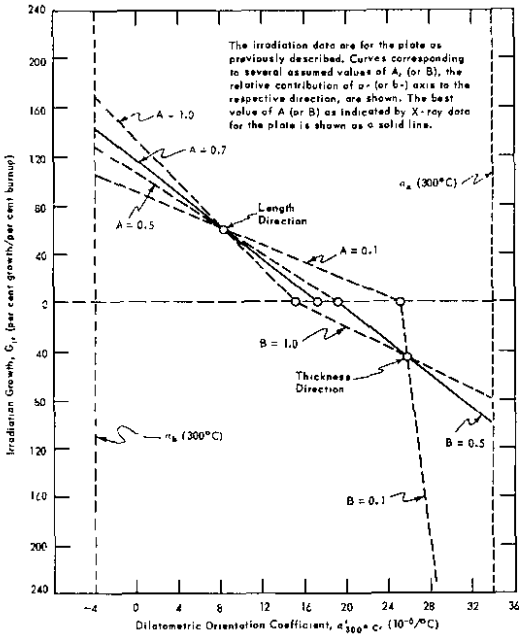


FIGURE 9 - CORRELATION OF DILATOMETRIC ORIENTATION COEFFICIENTS AND IRRADIATION GROWTH OF AS-ROLLED URANIUM PLATE

* Since the single crystal thermal expansion coefficients α_a, α_b, and α_c are still subject to some uncertainties, the data of Figure 9 should be taken as an illustrative approximation.

INTERPRETATION OF RESULTS

Values of γ_b obtained by the several techniques considered may be summarized along with pertinent conditions of the specimen irradiation, to illustrate the difference in intrinsic b-axis growth of the uranium crystal under the conditions presented.

Specimen and Correlation Technique	Irradiation Growth Coefficient, γ_b, % elongation per atom % burnup	Grain Size, mm	Irradiation Temperature, °C	External Restraint
Single Crystal				
Direct Irradiation (ANL)	420 ±20	-	<150	None
As-Rolled Rod				
Diffraction Method (AECL)	610	0.015 to 0.035[21] (Some rods un-recrystallized)	80 to 500[21]	Clad in[21] 0.080-inch Al, unbonded; Al/U = 0.06
As-Rolled Plate				
Diffraction Method (SRL)	114	0.015 to 0.050	50 to 150	Clad in 0.022-inch Al with Ni bond; Al/U = 0.3
Thermal Expansion Method (SRL)	120			

No simple correlation of the γ_b values with the given conditions is apparent, but their relative importance is indicated in a qualitative fashion in the following paragraphs.

Both the SRL plate and the AECL rod were of production-grade uranium, with relatively minor impurity contents (carbon 300-500 ppm), while the ANL single crystals were of high purity metal. The SRL plate was recrystallized to grains of about 0.030 mm; the AECL rod specimens were apparently either recrystallized to grains of about 0.030 mm or not recrystallized, depending on specific rolling conditions. Although grain interaction effects dependent upon state of recrystallization may have acted to enhance the growth of the AECL rod, no general correlation of irradiation growth with the above structural variables is evident.

Likewise, the specimen temperature and neutron flux and exposure do not appear to be the primary cause of the differences observed. The temperatures of the ANL crystals and the SRL plates were uniformly low, while the elevated internal metal temperatures of the AECL rod would be expected to decrease the growth rate of the specimen, rather than increase it, as was observed. The flux levels of the various irradiations were equivalently high; exposures of the SRL plate (0.077 atom % burnup) and the AECL rod (0.02 - 0.10 atom % burnup)

were comparable, while ANL crystals in the range 0.1-0.3 atom % burnup showed no major dependence of G_1 on exposure.

It would appear then that mechanical restraint effects were of major significance in determining the magnitude of dimensional changes of the specimens, especially the SRL plate. As indicated in the above table, the plate was clad with a considerable thickness of aluminum, capable of exerting a restraint up to possibly 3000-4000 psi on the uranium core. Such restraints have been demonstrated to suppress considerably the irradiation growth of uranium and uranium alloys.[4] The AECL rod with less cladding restraint and the unrestrained ANL crystal would show a greater growth than the SRL plate, though a precise correlation was not maintained.

Finally, the effects of specimen geometry, especially those associated with the resolution of cladding and other restraining forces into given directions in the specimen may play a role in the magnitude of dimensional instability encountered during irradiation, though no explicit assessment of their importance can as yet be made.

ACKNOWLEDGMENT

The development and application of SRL X-ray techniques for use in the correlation of irradiation growth and preferred orientation given in equation (5) are largely due to Dr. E. F. Sturcken of this Laboratory and have been previously reported.[15]

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