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DIMENSIONAL CHANGES IN URANIUM UNDER THERMAL CYCLING

AEC RESEARCH AND DEVELOPMENT REPORT

By: L. R. Kelman

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Dimensional Changes in Uranium Under Thermal Cycling

By L. R. Kelman

Argonne National Laboratory

Report ANL-FF-54

ABSTRACT

Uranium undergoes permanent dimensional changes when thermal cycled in the alpha-temperature range without passing through any phase transformations. This deformation is attributed to the anisotropy of thermal expansion inherent in noncubic metals. Alpha uranium, which is orthorhombic, is especially susceptible because of its high anisotropy of thermal expansion. Uranium rods prepared by a wide variety of fabrication and heat-treat methods were thermal cycled between 100°C and 550°C. Observations were made on the effects of fabrication technique, heat treatment, and thermal cycling on the dimensions, surface roughening, microstructure, and preferred orientation of these rods.

INTRODUCTION

This investigation was started about 1½ years ago as a study of the effects of thermal cycling upon the warp of 8-in. long, 0.364-in. diameter rolled uranium rods then under consideration for use in reactor rod assemblies for the CP-4 pile.[1] It soon became evident that permanent dimensional changes other than warp resulted from thermal cycling. It was found that heating and cooling of rolled uranium rods in the alpha temperature range (without passing through any phase transformations) resulted in a permanent increase in length with a corresponding decrease in diameter, and that continued thermal cycling of these rods resulted in continued elongation with no indication of an appreciable change in rate of elongation.[2]

Previous investigators have noted very small dimensional changes as a result of heating and cooling in the alpha temperature range. Battelle Memorial Institute was the first to note anomalous dilatometric behavior of uranium which they attributed to the anisotropy of the metal.[3,4] They were actually heating through the alpha-beta transformation, but obtained some indications that uranium undergoes permanent deformation in the high alpha region. Further experiments were made to check this point, but dimensional changes on heating and cooling through the transformation plus surface roughening of the specimens confused the results.[5,6,7,8] They also studied causes of surface roughening,[6] but in no case did they attribute dimensional instability or surface roughening to plastic deformation during heating and cooling in the alpha range.

In a study of the dimensional changes during annealing of 8-in. long W-slugs, Foote and Van Echo[9] found that annealing alpha-rolled slugs in the alpha region resulted in an increase in length

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and a decrease in diameter and that on one or two subsequent anneals these dimensional changes continued in the same direction at approximately the same rate. Although they claimed that the changes in dimensions increase with increasing time and temperature of annealing, their data show a definite temperature effect but no appreciable time effect. One of their 8-in. long slugs elongated 12.9 mils on annealing four hours at 585°C and 7.6 mils on a subsequent anneal of 24 hr at 465°C. Several slugs were given three successive anneals at 465°C for times of 6, 6, and 16 hr, and on each anneal the specimens elongated approximately 5 mils. They found that, on the average, diameter changes were such as to just compensate for the elongation, giving a constant density. Annealing gamma-extruded slugs at 465°C resulted in small but erratic dimensional changes. They offered no mechanism explaining elongation of alpha-rolled slugs on annealing in the alpha region.

In a recent series of papers Boas and Honeycombe[10,11,12,13] have shown that plastic deformation occurs when certain noncubic metals are repeatedly heated and cooled. They attribute this deformation to the anisotropy of thermal expansion inherent in these metals. They first noted that the surface of tin-base bearings roughened on heating and cooling whereas lead-base bearings showed no signs of surface roughening. They then thermal cycled the noncubic metals, tin, zinc, and cadmium along with a cubic metal, lead, and found that only lead was unaffected by this cyclic treatment of 30°C-150°C. In the noncubic metals they observed deformation marks, a roughening of the smooth surfaces of the specimens with accentuation of grain boundaries, and migration of grain boundaries as a result of thermal cycling.

From their test results they proposed the mechanism diagrammatically shown in Fig. 1 in which the stress between adjacent grains of different orientations is plotted against temperature of a thermal cycle. On heating, thermal expansion results in a stress between adjacent grains which is proportional to the increase in temperature until plastic deformation begins. Further heating results in plastic deformation which relieves any further stress except for the slight increase in stress due to work hardening. On cooling, elastic deformation in the reverse direction occurs until zero stress is reached at some elevated temperature. Further cooling results in stresses in the opposite direction which may exceed the yield point at this lower temperature resulting in plastic deformation in the opposite direction. Repeating this cycle results in stress reversals similar to those occurring when alternate external tensile and compressive stresses are applied. Because of this analogy, Boas and Honeycombe[10] named the phenomenon "thermal fatigue." This mechanism assumes a definite yield point and is very much simplified because it does not consider the effect of time on the creep of metals such as uranium.

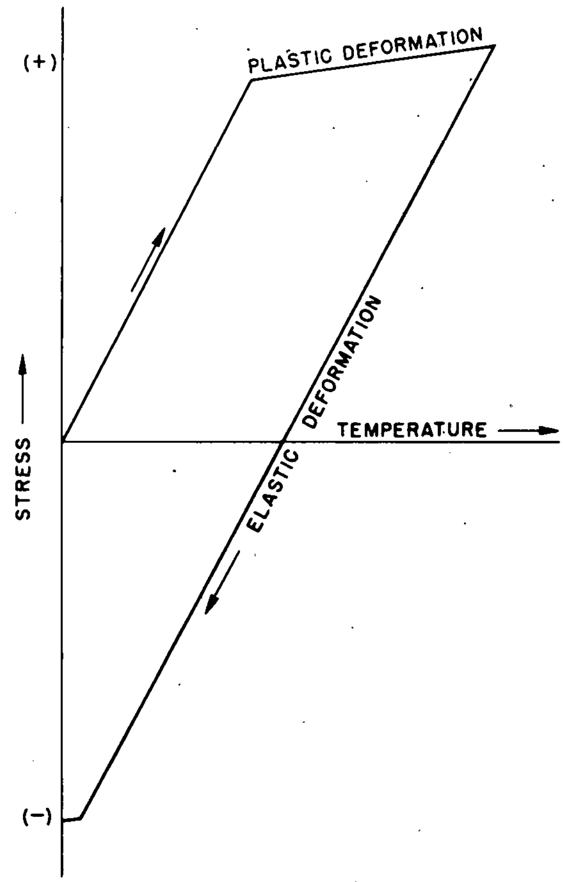

Fig. 1—Deformation with thermal cycling in anisotropic metals from Boas and Honeycombe.[11]

An interesting consequence of this phenomenon is that, no matter how stress free an anisotropic metal may be at an elevated temperature, cooling after annealing will result in new stresses. Therefore, polycrystalline aggregates of anisotropic metals cannot be obtained in a stress-free condition at room temperature by casting or annealing. This was first suggested by C. H. Desch[14] in 1923 during a controversy over the possibilities of grain growth on annealing noncubic cast metals without prior plastic deformation. He pointed out that anisotropy of thermal expansion gave rise to plastic deformation during cooling of the casting and on heating to the annealing temperature.

Boas and Honeycombe[11] studied factors influencing plastic deformation of noncubic metals on thermal cycling. They found that rate of cycling had a negligible effect and also that no plastic deformation occurred in lead from which they concluded that it is unlikely that nonuniform expansion due to temperature gradients was responsible for the observed deformation. They also found that the number and intensity of deformation lines as well as the extent of surface roughening increased with increasing number of thermal cycles, and also with the temperature range of cycling. Deformation of individual grains in a polycrystalline aggregate occurred regardless of grain size, but a single crystal of cadmium showed no evidence of deformation. They also found some indication that less plastic deformation occurred in individual grains of a specimen possessing a preferred orientation than in a specimen of randomly oriented grains. However, they made no measurements of the overall dimensions of the specimen.

Estimates of the magnitude of stresses produced during the heating of a noncubic metal have been made by Boas and Honeycombe[11,12] and Laszlo.[15] These calculations indicate that plastic deformation on thermal cycling is more dependent on the anisotropy of thermal expansion than on the anisotropy of Young's modulus or the critical shear stress.

Boas and Honeycombe[12,13] have also shown that the presence of hard second phases such as CuSn and SnSb in tin-base bearing alloys considerably reduces the deformation on thermal cycling, whereas deformation in tin-base alloys consisting only of a solid solution was almost as severe as in pure tin.

The phenomenon investigated by Boas and Honeycombe and the mechanism they propose to explain their findings appear to be related to the dimensional instability of uranium when thermal cycled. Alpha uranium is orthorhombic and highly anisotropic in thermal expansion. High temperature x-ray diffraction measurements at Battelle Memorial Institute[16,17] gave the following mean coefficients of expansion:

Crystallographic direction	25° to 300°C	25° to 600°C
[100]	23	28
[010]	−3.5	−1.4
[001]	17	22

Two of the coefficients are fairly large and positive whereas one is actually negative.

~~Some uranium rods, exposed in the Hanford piles, have shown extensive warping, surface roughening, and dimensional changes which may cause slugs to stick in the cooling tubes and which therefore results in a serious operating hazard.[18]~~ The similarity in dimensional changes of uranium due to thermal cycling and that due to pile exposure suggests a common cause. A consideration of the localized heating and cooling when fission occurs leads one to a plausible mechanism which appears to be related to the thermal cycling mechanism already discussed. For example, Siegel[19] has estimated that in natural uranium in a flux of 10^{13} neutrons/cm^2/second, each fission fragment produces lattice vibrations which in effect raises the temperature of 10^7 atoms to 1500°K. During any one second, a total fraction of 4×10^{-4} of the metal has been so heated for short intervals of time, ~~and during 100 days' exposure in the Hanford pile each portion of the metal has been so heated about 3300 times.~~ Even though there should be no relation between radiation and thermal cycling, a thorough investigation of thermal cycling effects is warranted. Uranium will be subjected to an appreciable number of heating and cooling cycles on starting up and shutting down the pile and to a very large number of small thermal cycles at pile operating temperatures. Finally, there is practical as well as fundamental interest in the problem of obtaining stress-free, dimensionally stable uranium and other anisotropic metals.

PROCEDURE

Since it was desired to apply the results of this investigation toward the design of the CP-4 fuel rods[20] the specimen dimensions and the thermal cycling setup were designed to simulate CP-4 conditions.

Fabrication and Heat Treatment of Specimens

Uranium rods were prepared by a wide variety of fabrication and heat treat methods resulting in a considerable variation in preferred orientation and grain size. In most cases alpha-rolled 1½-in. diameter Hanford slug metal was used for starting material. The methods of fabrication included casting, swaging, and extruding in the alpha and in the gamma temperature ranges, and rolling in the high alpha region and at 300°C (below the recrystallization temperature range). The heat treatments used included annealing at 575°C to recrystallize slightly above the maximum cycling temperature; beta annealing for 2 hr at 725°C to eliminate preferred orientation but with extreme grain coarsening; and fast-beta treating by dipping in a lead pot at 725°C, holding for a short time, and then water quenching in an attempt to obtain a fine-grained, randomly oriented structure. Also studied were some coarse-grained, highly oriented uranium rods prepared by the strain-anneal method[21,22] and a highly oriented plate rolled at 300°C.

Preparation of Specimens for Cycling

The same procedure was used to prepare all rods for cycling. The rods were fabricated to approximately ½ in. diameter at which size some of them were given a desired heat treatment. They were then machined to 0.384 in. diameter and either 4-in. or 2-in. lengths. At this stage they were electropolished and deep etched[23] and their macro grain structures were noted.

The rods were then annealed in a stainless steel container filled with NaK as a heat transfer medium for 2 hr at 575°C and furnace cooled. NaK has been used throughout these experiments as a heat transfer medium because it does not alloy with the bare uranium rods, it is liquid in the range of temperatures used for this study, it is a fairly good thermal conductor, and its high coefficient of thermal expansion produces convection currents that help to eliminate thermal gradients. The annealing temperature was arbitrarily chosen as 575°C, which is 25°C higher than the maximum cycling temperature. Thus dimensional changes which might be caused by recrystallization, grain growth, and possibly by transformation of unstable phases in the fabricated rods were eliminated prior to cycling.

Following the anneal, the rods were centerless ground to 0.364 in. diameter and the ends were ground square to eliminate warp due to annealing.

Thermal Cycling of Specimens

The rods were thermal cycled under conditions which roughly simulate pile conditions. They were supported freely in a vertical position with 20 mils clearance in retaining rings at each end of the rods. Hence, the rods stand on end and are free to elongate and warp without restraint other than that produced by their own weight. This assembly was held in a stainless steel container filled with NaK as a heat transfer medium as shown in Fig. 2. The container was heated by induction using a 20 kva Ecco converter. By locating thermocouples in the center of a rod and at several points along the length of a 10-in. column of NaK, it was found that heating from 100°C to 550°C in 5 min gave no appreciable thermal gradients. Cooling from 550°C to 100°C by means

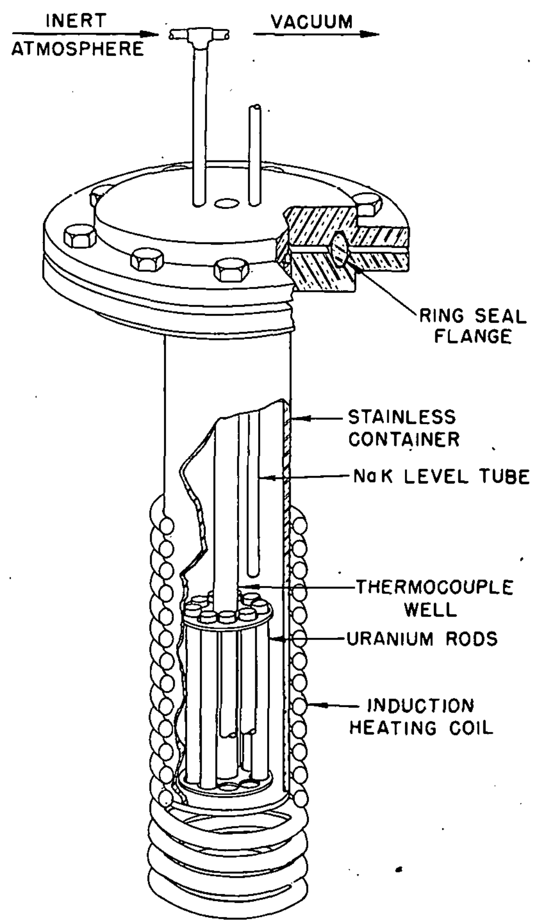

Fig. 2—Thermal Cycling set-up.

of several fans took from 20 to 25 minutes. The upper temperature of 550°C was arbitrarily chosen. It is somewhat higher than the maximum temperature initially contemplated in the center of the CP-4 pile fuel rods. The lower temperature was fixed at 100°C as a temperature which could be reached in a reasonable time by air cooling.

To determine the effect of small temperature fluctuations at elevated temperatures, a few rods were cycled between 500°C and 550°C. Heating and cooling each took about 2½ min for this test.

Inspection and Measurement of Rods

In most cases, three duplicate rods were cycled for each type of fabrication and heat treatment. Specimens for metallographic study of microstructure and x-ray diffraction study of preferred orientation were cut from one rod after various numbers of cycles. The other two rods were used as duplicate specimens for length, diameter, surface roughening, and warp studies. Lengths and diameters were measured with a hand micrometer after 50, 100, 300, 700, and in some cases 1500 cycles. Measurements at the beginning of the tests were fairly accurate, but warp and surface roughening precluded accurate measurements of some rods as cycling progressed. Surface roughening was noted visually, and recorded by means of full size photographs, but no attempts were made to quantitatively measure the size, shape, or number of bumps. Warp measurements were made with the rod supported on V-blocks using a Federal dial gage graduated in 0.0001-in. divisions and eccentricity was taken to be half the total run-out.

Specimens were prepared for preferred orientation determination by machining to true cylinders and then electroetching a 0.005-in. layer from the surface. X-ray photograms were taken using copper K-alpha radiation, with 0.025-in. pinhole, 5-cm specimen to film distance, 3-cm pinhole to specimen distance, and one hour exposure. The settings used were: (1) the beam perpendicular to the rolling direction; (2) the beam at 18 degrees to the rolling direction. The specimens were rotated about their own axes at 1 rpm to get an integrated picture. The sharpness of the texture was measured by the length of the Debye arc of the (002) plane which indicates the degree of scatter of the "C" axis from the mean orientation. For further details see references 24 and 25.

DISCUSSION OF RESULTS

No attempt is made in this report to thoroughly discuss every specimen that has been cycled in the course of this investigation. However, the elongation data of most of the specimens are tabulated and discussed briefly to show how they fit into the general cycling program. Representative specimens are discussed in detail to illustrate the effects of fabrication technique, heat treatment, and thermal cycling on elongation, surface roughening, microstructure, and preferred orientation of the specimens. Within the accuracy of a hand micrometer in measuring rods which did not warp and whose surfaces did not bump or oxidize during cycling, diameter changes were such as to just compensate for the length changes. Diameter measurements included large errors due to surface roughening of many rods and due to surface oxidation of some rods and are, therefore, not given. Some density determinations were made which will be discussed later. Warp determinations are not reported because it is thought that the amount of warp was strongly influenced by the way the specimens were supported during the test. This is especially true in the case of rods which elongated so much that their upper ends were well above the retaining ring. Also, the 4-in. long specimens warped considerably more than the 2-in. long specimens. Photographs of some of the rods after cycling (see Figs. 5 and 6a) give an idea of the extent of warp.

Rolled Rods—Alpha Annealed

Uniformity and Reproducibility

Uranium rods that have been rolled in the alpha temperature range by a variety of methods have been thermal cycled up to 1500 times between 100°C and 550°C. The length changes of these rods are tabulated in Tables 1, 2, and 3 where the results are arranged according to the type of rolling. The only heat treatment these rods were given before cycling was a 575°C anneal which is slightly above the maximum cycling temperature.

Hot-Rolled Rods.—When this investigation was started, the standard method for alpha rolling uranium was to heat to 600°C and then roll to finished size without returning the rods to the furnace. The rods tend to become hotter as rolling proceeds, so they are held in air between

Table 1 — Elongation of standard alpha-rolled uranium rods due to thermal cycling between 100°C and 550°C

Fabrication and heat treatment	Rod No.	Rod length (in.)	Per cent elongation $\frac{L-L_0}{L_0} \times 100$: Due to anneal	Number of cycles (N): 100	300	700	1500	Elongation coefficient $\frac{1}{L}\frac{dL}{dN} \cdot 10^6$
575°C annealed	5	4	0.01	2.09	5.27	10.85		160
	8	4	−0.03	1.93	4.57	7.84		130
	9	4	−0.05	2.31	5.88	12.01		180
575°C annealed	40	4	−0.03	2.45†	5.46	10.20		200
	41	4	0.15	1.95†	4.25	9.00		160
	42	4	0.01	3.60†	8.77	20.25		270
Hanford slug — 575°C annealed	997	4	0.00	2.13	4.4	12.0		160
575°C annealed — 5 days in liquid nitrogen	15	4	−0.06	0.83	1.65	4.57		61
	16	4	−0.04	2.92	9.50	20.55		250
	17	4	−0.07	2.33	7.10	14.82		210
725°C annealed	4	4	−0.34	−0.04†	0.07	0.40	0.78	(‡)
	6	4	−0.28	−0.05	(*)	(*)	(*)	(‡)
	10	4	−0.28	−0.02†	0.31	1.00	1.79	(‡)
600°C rolled, 15 min at 725°C between passes, 75% reduction from std alpha-rolled rod, 575°C annealed	82	4	0.03	0.74	1.88	5.42		71
	83	4	0.02	0.60	1.90	4.63		63
	84	4	−0.01	0.61	1.80	4.50		61

*Rod cut up for samples at various stages of cycling.
†Rod cycled 2200 times between 500°C and 550°C before further cycling between 100°C and 550°C, but recorded elongations are corrected for the elongation that occurred on 500°C-550°C cycling.
‡Coefficients too small to be measured accurately.

Table 2 — Elongation of 600°C-soak-rolled uranium rods due to thermal cycling between 100°C and 550°C

Fabrication and heat treatment*	Rod No.	Rod length (in.)	Per cent elongation $\frac{L-L_0}{L_0} \times 100$: Due to anneal	Number of cycles (N): 100	300	700	1500	Elongation coefficient $\frac{1}{L}\frac{dL}{dN} \cdot 10^6$
30 min at 600°C after every pass, 575°C annealed	53	4	0.00	1.79	(†)	(†)	(†)	180
	55	4	0.00	1.77	4.48	9.75	20.22	130
	57	4	−0.03	1.75	4.41	8.80	17.04	120
30 min at 600°C after every pass, not annealed	54	4		1.71	4.57	10.05		140
	56	4		1.71	4.53	9.88		140
	58	4		1.72	4.16	9.98		150
15 min at 600°C after every 2 passes, 575°C annealed	69	4	0.00	2.24	6.91	14.48		220
	70	4	0.02	1.95	6.40	13.24		210
	71	4	0.04	1.45	5.60	11.60		160
15 min at 600°C after every 2 passes, 2 min at 717°C, water quenched, 575°C annealed	88	4	−0.05	0.13	0.48	0.89		14
	89	4	−0.03	0.20	0.63	1.82		24
	90	4	−0.05	0.15	0.64	0.99		16
15 min at 600°C after every 2 passes, 2 min at 687°C, water quenched, 575°C annealed	91	4	−0.04	0.36	0.99	1.80		28
	92	4	−0.04	0.19	0.50	0.90		15
	93	4	−0.03	0.35	0.94	1.86		28

*All rods reduced 75% by 600°C soak rolling from standard alpha-rolled rod.
†Rod cut up for samples at various stages of cycling.

passes until they appear to be about 600°C. Nonuniformity in rolling texture and grain size along the length of standard alpha-rolled rods results from this poor temperature control, and there are indications that the rods are sometimes heated through the alpha-beta transformation.

The erratic structure of these rods was reflected in their behavior when thermal cycled (Table 1). Three sets of specimens consisting of 3 supposedly duplicate specimens in each set—(5, 8, and 9), (40, 41, and 42), and (15, 16, and 17)—showed large variations in elongation. In general it was found that the rods which elongated the least (8, 41, and 15) had the roughest surfaces after cycling and had the largest grain size. One set of the standard alpha-rolled rods, (15, 16, and 17), were submerged in liquid nitrogen (−195.8°C) for 5 days in an effort to transform any retained beta phase. The cold treatment resulted in no perceptible changes in dimensions and the cycling results were erratic.

A 4-in. length of a standard alpha-rolled W-size slug received from Hanford (No. 997 from G material of billet No. 1735) was found to elongate 12.0% in 700 cycles which is about the same elongation as the similarly fabricated small diameter rods 5, 8, and 9.

In an effort to obtain reproducibility of structure in rolled uranium rods, several rods were rolled at 600°C and soaked in a furnace at that temperature between passes. Better reproducibility in structure and in growth on cycling was

Table 3—Elongation of cold-rolled uranium rods due to thermal cycling between 100°C and 550°C

			Per cent elongation $\frac{L-L_0}{L_0} \times 100$						Elongation coefficient
		Rod		Number of cycles (N)					
Fabrication and heat treatment	Rod No.	length (in.)	Due to anneal	50	100	300	700	1500	$\frac{1}{L}\frac{dL}{dN} \cdot 10^6$
Room temp rolled—18% reduction	1	4	−0.01		2.22	7.19	19.70		250
from std alpha-rolled rod,	2	4	0.05		2.34	8.10	21.80		260
575°C annealed	3	4	−0.06		2.08	7.20	21.05		260
300°C Soak-rolled—75% reduction	47	4	0.25		3.11	9.48	21.85	60.25	320
from std alpha-rolled rod,	48	4	0.22		3.20	(*)	(*)	(*)	320
575°C annealed	49	4	0.21		3.18	9.86	25.20	64.0	320
300°C Soak-rolled—89% reduction	94	4			(*)	(*)	(*)		(*)
from std alpha-rolled rod,	95	4	0.52			11.27	29.5		360
575°C annealed	96	4	0.54			10.45	30		360
300°C Soak-rolled—89% reduction	1-4	2		1.35	2.83	(*)	(*)		280
from std alpha-rolled rod,	5-8	2		1.27	2.64	9.02	26.47		290
575°C annealed	11-14	2		1.61	3.31	11.26	31.41		360
	15-18	2		1.65	3.41	11.48	31.83		370
300°C Soak-rolled—89% reduction	21-24	2		0.10	0.31	0.72	1.68		24
from std alpha-rolled rod,	25-28	2		0.19	0.41	1.09	2.42		32
725°C annealed	31-34	2		0.28	0.47	(*)	(*)		50
575°C annealed	35-38	2		0.13	0.20	1.25	1.45		21
300°C Soak-rolled—89% reduction	41-44	2		0.00	0.01	(*)	(*)		(†)
from std alpha-rolled rod,	45-48	2		0.00	0.03	0.25	0.99		14
½ min at 725°C, water quenched,	51-54	2		0.15	0.08	0.25	1.24		18
575°C annealed	55-58	2		0.02	0.08	0.35	1.24		18
300°C Soak-rolled—75% reduction from std alpha-rolled rod;	50	4	−0.02		0.04	0.49	1.23	2.58	17
2 min at 723°C, water quenched,	51	4	0.00		0.02	0.25	0.80	2.71	13
575°C annealed	52	4	0.19		(*)	(*)	(*)	(*)	(*)

*Rods cut up for samples at various stages of cycling.
†Coefficients too small to be measured accurately.

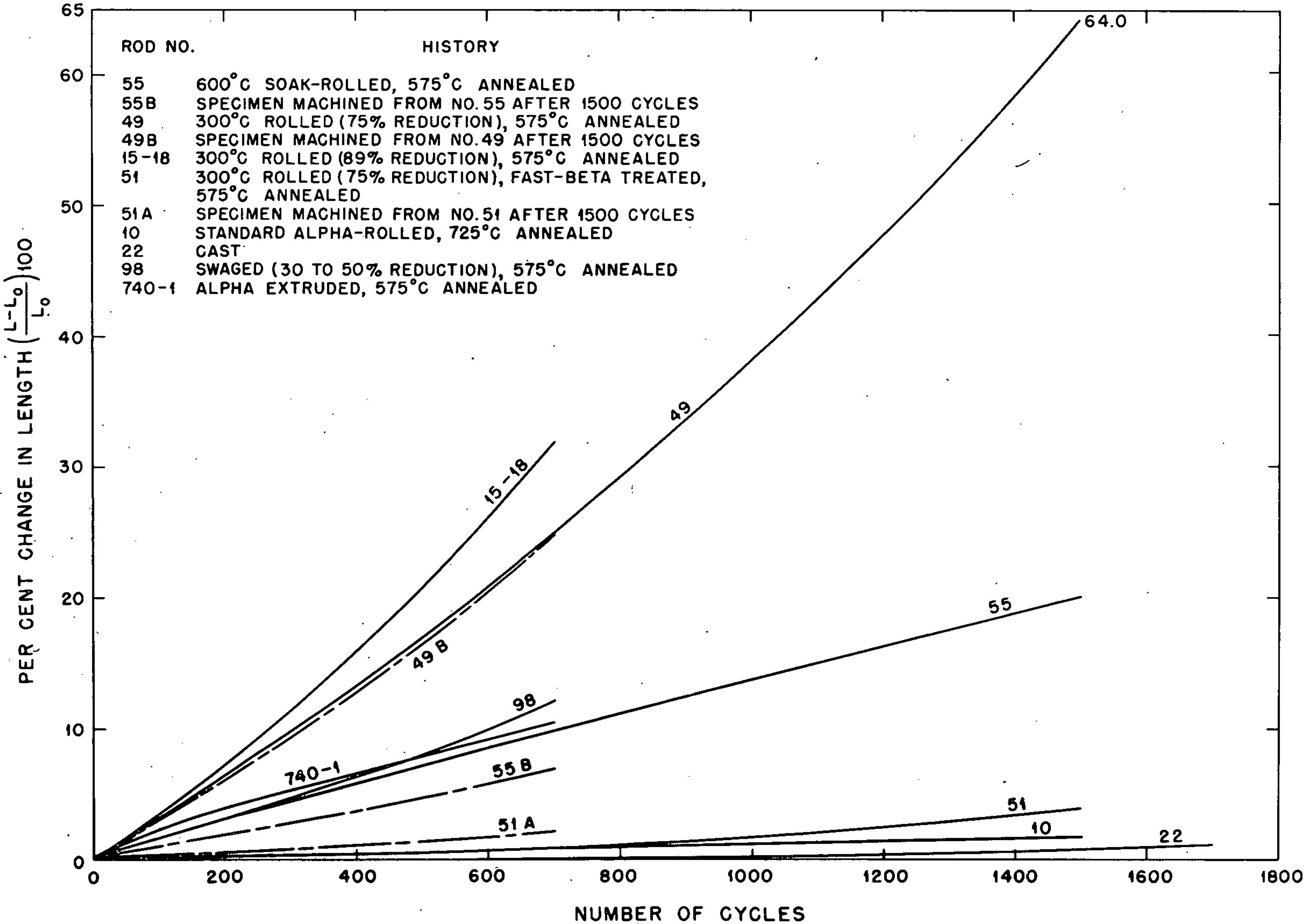

Fig. 3 — Per cent elongation of uranium rods due to cycling between 100 °C and 550 °C.

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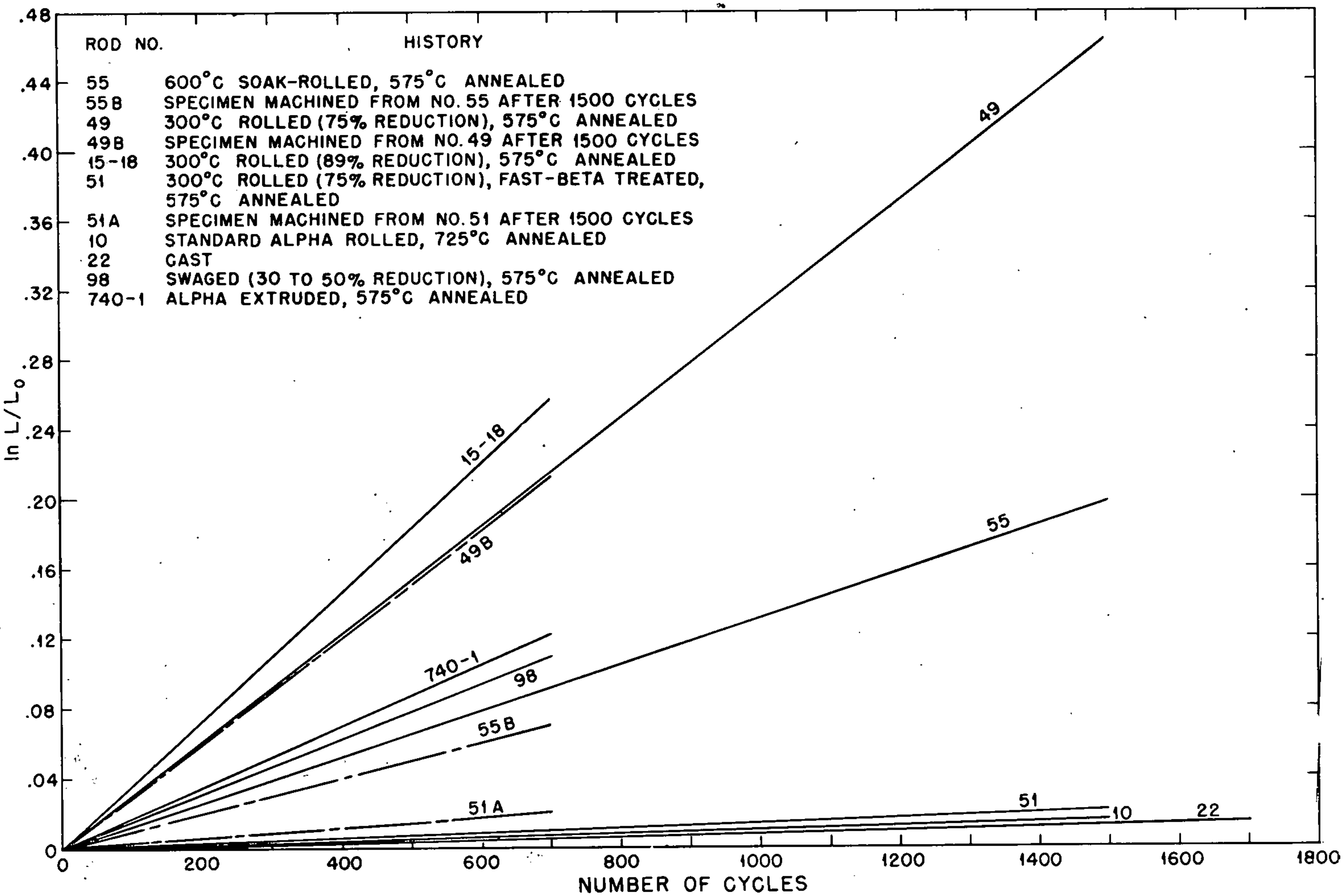

Fig. 4 — True elongation of uranium rods due to cycling between 100 °C and 500 °C.

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achieved by this method (Table 2), but the method is cumbersome, time consuming, and not always successful. The use of a liquid-metal bath to keep the rods at constant temperature during rolling might make this a feasible production technique.

The effect of the anneal before cycling was examined by annealing one set of 600°C-soak-rolled rods (53, 55, and 57) and not annealing an identical set of rods (54, 56, and 58). These rods all elongated between 8.8% and 10% in 700 cycles regardless of whether or not they were annealed prior to cycling.

Cold-Rolled Rods.—Rolling below the uranium recrystallization temperature was another possible method of obtaining reproducible rolling results. One attempt was made to roll at room temperature, but only 18% reduction was attained before fracture occurred. However, cycling of 3 of these rods (1, 2, and 3) resulted in almost identical elongations in 700 cycles (Table 3).

Rolling at 300°C with a 5-min soak at 300°C between every other pass to insure maintaining the proper temperature during rolling resulted in rods of very uniform grain structure and strong preferred orientation which reacted similarly to thermal cycling. Also, this method of rolling results in considerably less oxidation of the uranium than does 600°C rolling which is an important consideration in the rolling of U^{235}. It was, therefore, decided to use 300°C-soak-rolled rods for the major part of this investigation.

Elongation

The amount of longitudinal growth on thermal cycling was found to increase directly as the number of cycles. In Figs. 3 and 4 are plotted elongation data for at least one rod of every type that was cycled. A plot of per cent elongation based on the original length against number of cycles, as shown in Fig. 3, shows an apparent acceleration of elongation as cycling proceeds. This type of plot does not represent the instantaneous growth of the rods and, therefore, gives a false rate of growth. If the instantaneous change in length, dL/L, is integrated between the original length, L_0, and the instantaneous length, L,

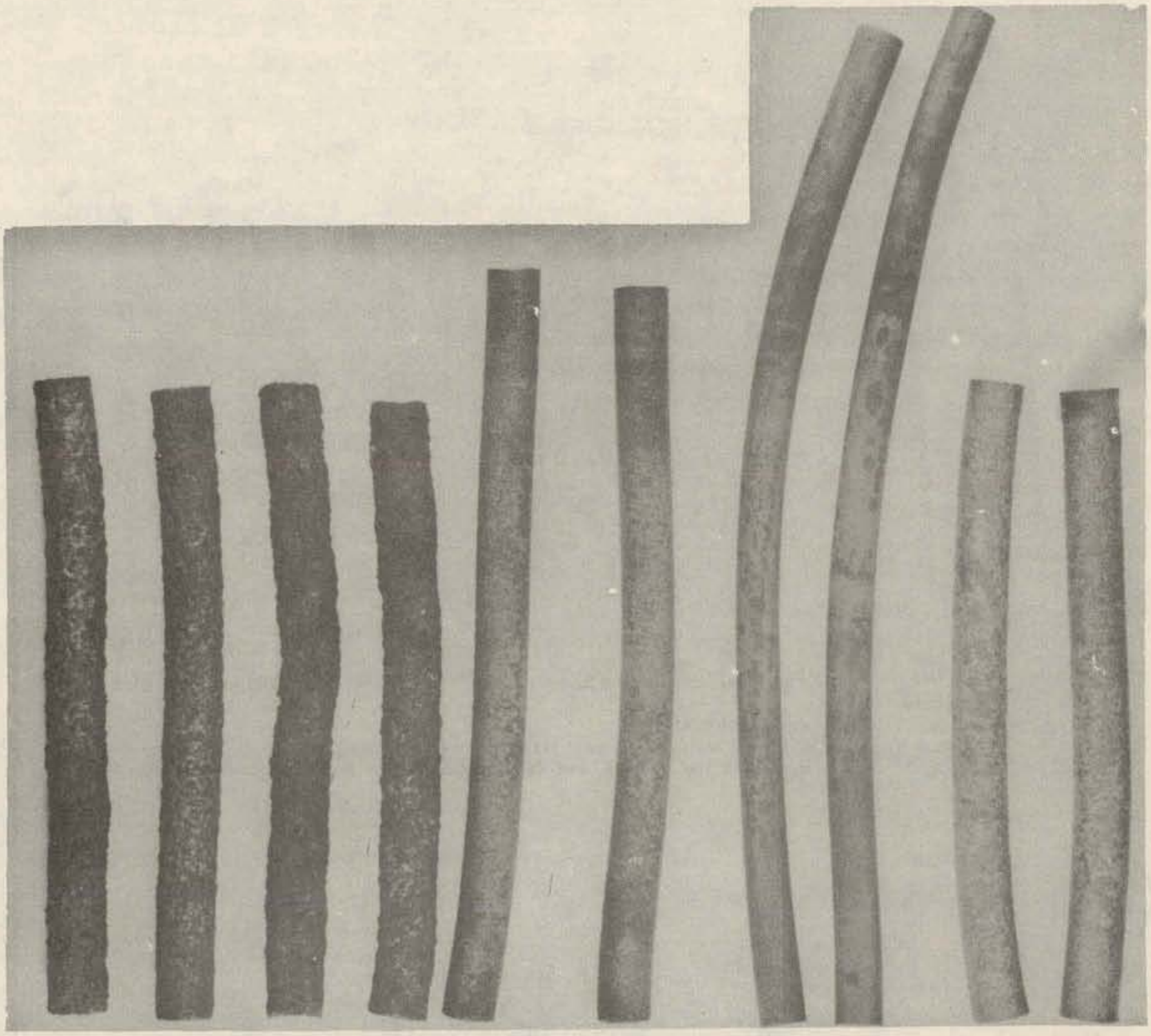
$$\int_{L_0}^{L} \frac{dL}{L} = \ln \frac{L}{L_0}$$

and the resulting expression, $\ln L/L_0$, is plotted against the number of cycles, a true rate of growth is obtained.[26] The slope of the curve gives a coefficient of elongation on cycling representing the increase in length per unit length in one cycle. Coefficients of elongation, henceforth referred to as kappa (K), were obtained in this manner for all rods cycled and are included in all tables showing growth on cycling. Warp and surface roughening were not appreciable near the beginning of the cycling tests, but, as can be seen from the cycled rods shown in Figs. 5 and 6, they introduced large errors in length measurements in the latter stages of the cycling tests. Therefore, in drawing the curves, more weight was given to the earlier points. In all tests, length measurements after 50 or 100 cycles were sufficient to predict fairly accurately the rate of elongation on further cycling.

Study of the data in Tables 1, 2, and 3 and of Figs. 3 and 4 shows that, in general, the greater the reduction in area during rolling, the greater the rate of growth. Further, rods rolled at lower temperatures seem to grow at higher rates. A rod (15-18) that had been rolled at 300°C to an 89% reduction in area showed the greatest rate of elongation on cycling—$K = 370 \times 10^{-6}$ or approximately 30% elongation in 700 cycles (Table 3). Continued cycling to 1500 cycles of rods (47, 49) that had been 300°C rolled to 75% reduction resulted in as much as 64% elongation ($K = 320 \times 10^{-6}$). These rods are shown in Fig. 5 after cycling. Specimens approximately ½ in. diameter × 1 in. long were machined from these cycled rods and were recycled 700 times. The recycled rods elongated at about the same rate as did the original rods (Figs. 3 and 4).

To examine the possible effect of the dimensions of the rods on elongation due to cycling, 0.364-in. diameter rods of lengths of ⅛ in., ¼ in., ½ in., 1 in., and 2 in., shown in Fig. 7, were machined from the same highly oriented 300°C-rolled rods and thermal cycled together. The test results (Table 4) show that varying the length of these rods had no appreciable effect on the amount of growth after 300 cycles.

Within the accuracy of the measurements of length and diameter, density appeared to be unaffected by thermal cycling. Density determinations made before and after 1500 cycles of rod 49 which elongated 64% ($K = 320 \times 10^{-6}$) indicated a slight decrease in density. The method consisted



22	23	4	10	55	57	47	49	50	51
*Cast		600°C rolled 725°C anneal for 2 hr		600°C soak-rolled		300°C soak-rolled		300°C soak-rolled Pb pot dipped at 723°C	

*Cast slugs were cycled 1700 times.

Fig. 5—Uranium rods fabricated by various methods after 1500 cycles between 100°C and 550°C. Approx. 1×

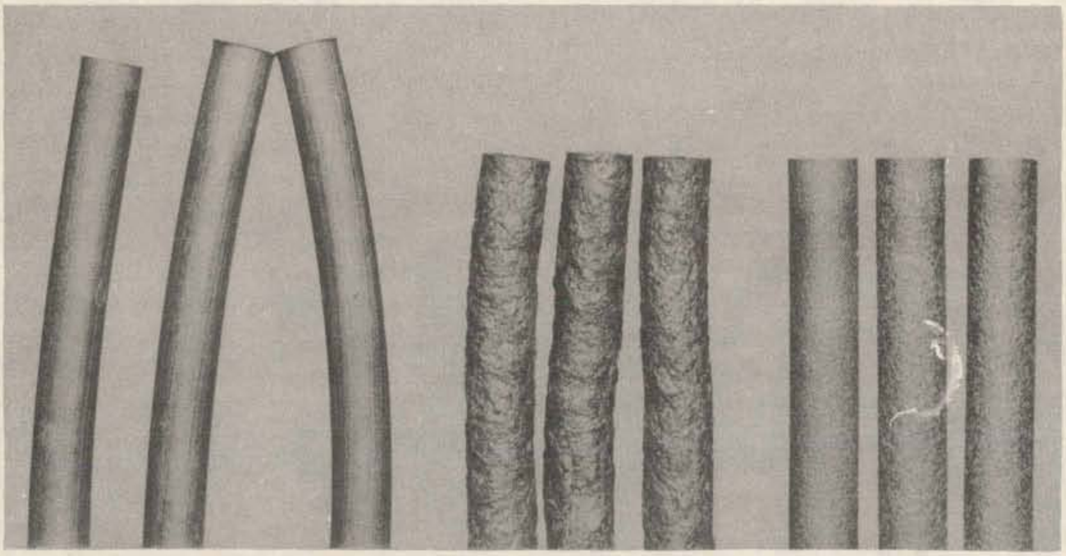


Fig. 6a — 300°C-rolled rods of various thermal histories after 700 cycles between 100°C and 550°C (from M-4315). 1×

Group I — Annealed 2 hr at 575°C
Group II — Annealed 2 hr at 725°C, then 2 hr at 575°C. (slow-beta treatment).
Group III — ½ min dip in lead pot at 725°C and water quenched, then annealed 2 hr at 575°C. (fast-beta treatment).

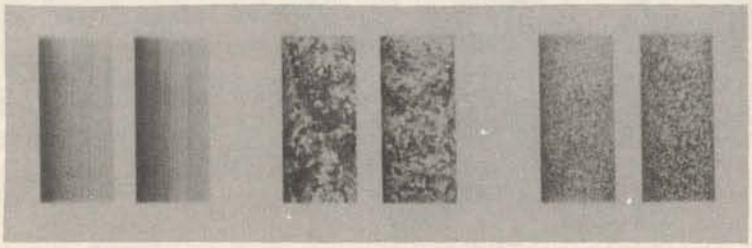
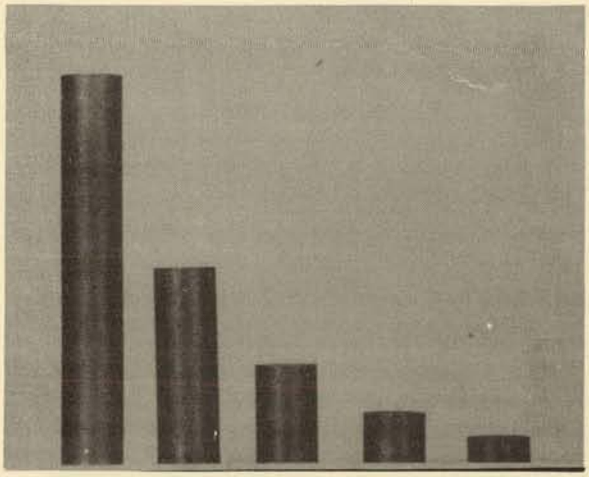


Fig. 6b — Macrostructures of the rods shown in Fig. 6a before cycling (from M-4383). 1 ×

of accurately weighing and measuring ¼-in. diameter by ½-in. long polished cylinders. Checks of duplicate specimens showed a decrease in density of 0.17 g/cc due to thermal cycling. Making liberal allowances for possible errors in each step of the density measurement procedure, the calculated maximum and probable errors were 0.039 and 0.038 g/cc.



2 in. 1 in. ½ in. ¼ in. ⅛ in.

Fig. 7—Specimens used to study the effect of rod dimensions on growth due to cycling. Rods were 0.364 in. diameter and 2 in., 1 in., ½ in., ¼ in. and ⅛ in. long (from M-4267). 1×

Table 4—Effect of rod dimensions on growth of 300°C rolled uranium rods when cycled 300 times between 100°C and 550°C

Rod dimensions (in.) Length	Diameter	Elongation (per cent)	Elongation coefficient (K) $\frac{1}{L}\frac{dL}{dN}\cdot 10^6$
⅛	0.364	9.05	290
¼	0.364	9.05	290
½	0.364	8.77	280
1	0.364	8.84	280
2	0.364	8.99	290

Surface Roughening

In general, uranium rods rolled in the alpha temperature range and then annealed at 575°C remained smooth on cycling. As was previously pointed out, the nonuniformity in rolling texture and grain size of hot rolled uranium rods was reflected in the erratic elongation results on cycling. The rolled rods with the least preferred orientation elongated the least. At the same time, these rods had the largest grain size of all the rolled rods, and their surfaces were roughened slightly on cycling. Typical surface appearance of rolled and annealed rods after cycling is shown in Fig. 5 (47, 49 and 55, 57) and in Fig. 6a (Group I).

Preferred Orientation

It is well known that plastic deformation of metals produces texture in which the individual grains take up preferred orientations with respect to the direction of working. Three general types of preferred orientation have been found in rolled uranium rods depending upon the temperature of rolling and the amount of reduction. The mean orientation textures are as shown in Table 5.

From Table 5 it may be seen that the c-axis is radial to the rod axis while the b-axis orientation varies with amount and temperature of cold work. Room temperature rolling is characterized by a b-axis orientation parallel to the rolling direction. The least amount of working occurs with 600°C-soak rolling where the (110) pole tends to line up with the rolling direction, placing the (010) (b-axis) as much as 45 degrees from the rolling direction in the most oriented condition. The texture produced by 300°C rolling is characterized by a duplex orientation of the axis which orients in the rolling direction and also at an angle to the rolling direction. This texture may be described by a major and minor mean orientation with a scatter as may be seen from the pole figures (Fig. 8).

Major texture: c-axis radial with a scatter of 15 degrees toward the rod axis and b-axis at 31 degrees from the rolling direction with a scatter of 15 degrees to and away from the rolling direction.

Minor texture: c-axis same as above, and b-axis parallel to the rolling direction with a scatter of about 10 degrees away from the rolling direction.

Thermal cycling was found to sharpen the preferred orientation texture as may be seen from the x-ray data in Table 6. The scatter of the (001) pole decreases from 15 degrees dark and 19 degrees light to 10 degrees dark and 15 degrees

Table 5 — Preferred orientations in rolled uranium rods

Rod No.	X-ray No.	Fabrication	Radial Pole	Radial Scatter Dark	Radial Scatter Light	Rolling direction Pole	Rolling direction Scatter
1	453	18% Reduction — room temp	(001)	20	32	(010)	48
53	475	Soak-rolled — 600°C	(001)	42	70	(010)*	35
1-4	633	89% Reduction — 300°C,	(001)	15	25	(010)†	30
		duplex	(001)	15	25	(010)	10

*(010) pole 35 degrees from rolling direction.
†(010) pole 31 degrees from rolling direction.

Table 6 — The effect of 700 thermal cycles on the preferred orientations of 300°C-rolled uranium rods

Heat treatment	Rod No.	X-ray No.	Degree of scatter (poles) 001	010	110*
575°C anneal					
Before cycling	1-4	633	15 D 19 L	45	35 L 20 D 45
After cycling	5-8	634	10 D 15 L	40	20 30

D — dark intensity. L — light intensity. *Duplex structure.

light after 700 cycles. The decrease may be due to better ordering of the lattice upon cycling inasmuch as the width of the lines decreases as well as the length of arc. A similar decrease in scatter was noted on other rods[25] and, in some cases, indication of preferred orientations of the b-axis occurred where none could be detected previous to cycling.

Microstructure

The effect of thermal cycling on the microstructure of a 300°C-rolled and 575°C-annealed rod (1-4) is shown in Fig. 9. The fine grain structure resulting from the recrystallizing anneal was not appreciably changed after 100 cycles, was slightly coarsened after 300 cycles, and was appreciably coarsened to a mixed grain size after 700 cycles as shown in Fig. 9d. Cycling 1500 times of rods 47 and 49, which were similar to those shown in Fig. 9, resulted in a somewhat larger grain size. However, 700 more cycles of specimens machined from these 1500-cycled rods resulted in no further change in the microstructure.

There was some evidence that grain size or some other unknown factor contributes to the rate of growth on cycling. For example, standard alpha-rolled rod 41 grew 9.00% ($K = 160 \times 10^{-6}$) and rod 42 grew 20.25% ($K = 270 \times 10^{-6}$) in 700 cycles, as shown in Table 1. These were supposed to have been duplicate specimens from the same rolled rod, and x-ray diffraction studies showed no appreciable difference in preferred orientation. However, rod 41 was found to have about twice

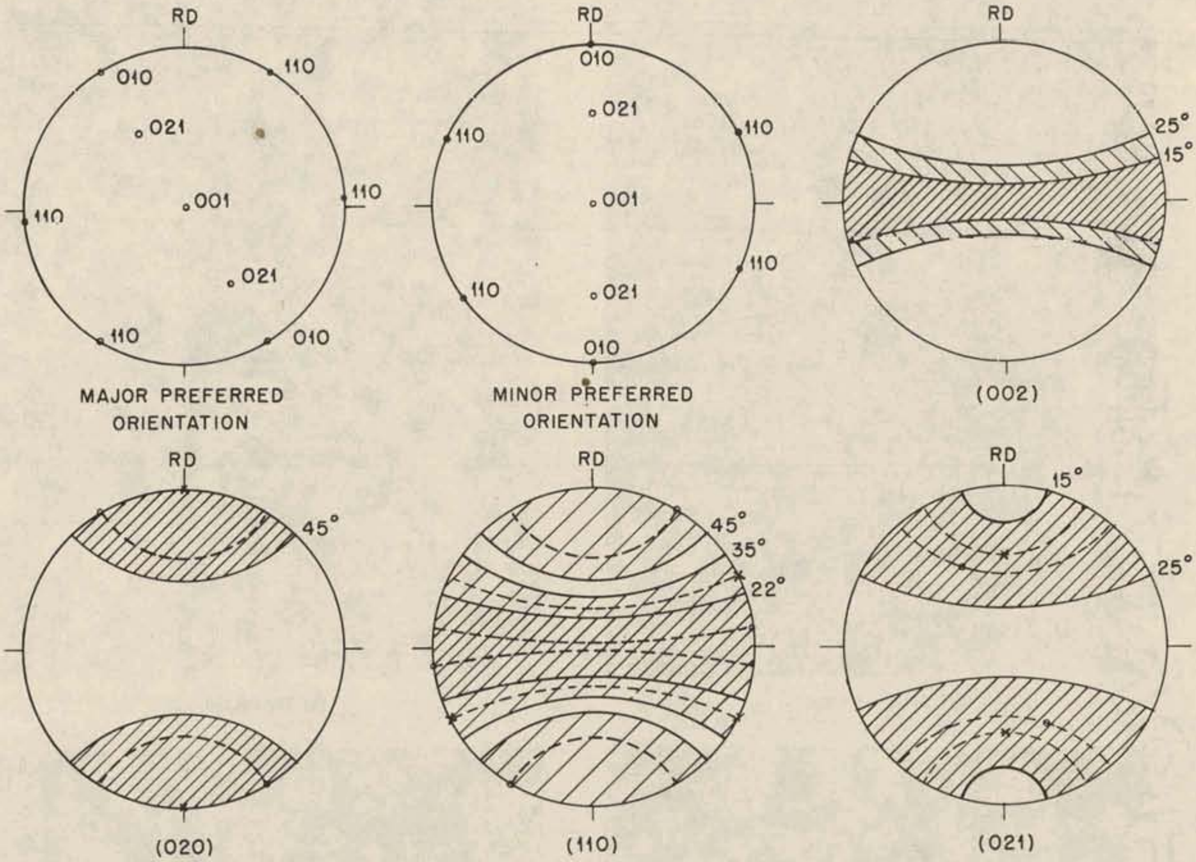


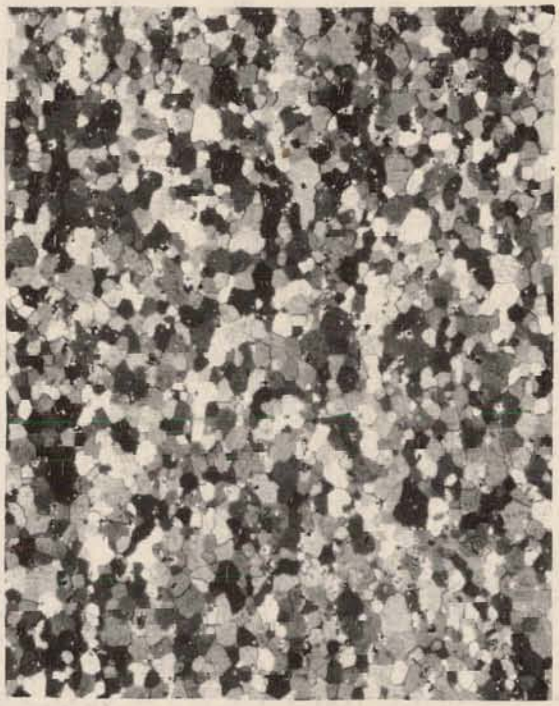
Fig. 8—Pole figures of 300°C-rolled and 575°C-annealed rod No. 1-4 showing a duplex preferred orientation. (X-ray specimen No. 633)

the grain size of rod 42. This same apparent grain size effect was found in another set of rods, and numerous tests by Schwope, et al,[27] have been interpreted by them to show the same effect. To divorce the grain size effect from the effect of preferred orientation, very coarse-grained, highly oriented specimens were cycled along with specimens having similar preferred orientations but comparatively finer grains. The coarse-grained specimens were obtained from the gage length and the finer grained specimens from the ends of the tensile bars shown in Fig. 10 prepared by the strain-anneal method.[21,22] The coefficients of thermal expansion of these specimens, as measured by Schwope and coworkers at Battelle Memorial Institute, are given in Table 7 along with the elongation on cycling.

The coarse grains were larger than that of any of the rods cycled during this investigation as shown in Fig. 11a. Nevertheless, they elongated 15.2% ($K = 200 \times 10^{-6}$) and 13.8% ($K = 180 \times 10^{-6}$) in 700 cycles. The finer grained rods were slightly less anisotropic than the coarse-grained rods and grew 21.0% ($K = 270 \times 10^{-6}$) and 22.0% ($K = 280 \times 10^{-6}$) in 700 cycles. This greater elongation may reflect a grain size effect, however, the results of this test confirm previous indications that the degree of preferred orientation is the major factor influencing the rate of elongation on thermal cycling. The extreme surface roughening of the coarse-grained specimens may account for the lower rate of elongation of the coarse than the fine-grained specimens. The grain size in the coarse-grained specimens was large compared to the diameter of the specimens.

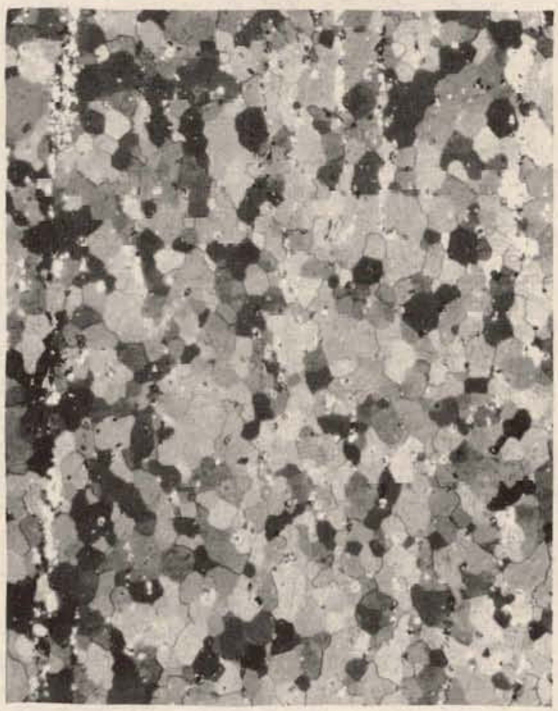
The microstructures of the coarse and fine-grained specimens before and after 700 cycles are shown in Fig. 11. Cycling of the coarse-

(a) Before cycling



(b) 100 cycles

(c) 300 cycles



(d) 700 cycles

Fig. 9a-d.—The effect of thermal cycling between 100°C and 550°C on the microstructure of a 300°C-rolled rod (1-4). Polarized light at 150×.

grained specimen resulted in the development of small similarly oriented grains clustered in areas approximately the same size as the original large grains (Fig. 11b). Further discussion of this grain structure which was developed on cycling will be reserved for a later section concerned with the thermal cycling of other coarse-grained specimens. The finer grained specimen actually had a mixed grain structure before cycling (Fig. 11c) which reacted to cycling in much the same manner as the 300°C-rolled, 575°C-annealed rod (1-4) shown in Fig. 9.

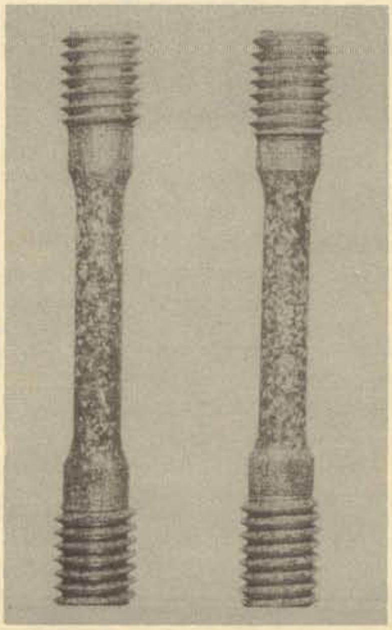


Fig. 10—Strain-anneal tensile bars used to compare the effect of cycling on coarse and fine-grained uranium of similar preferred orientation (from M-4344). 1×

Rolled Rods—Beta Heat Treated

Elongation

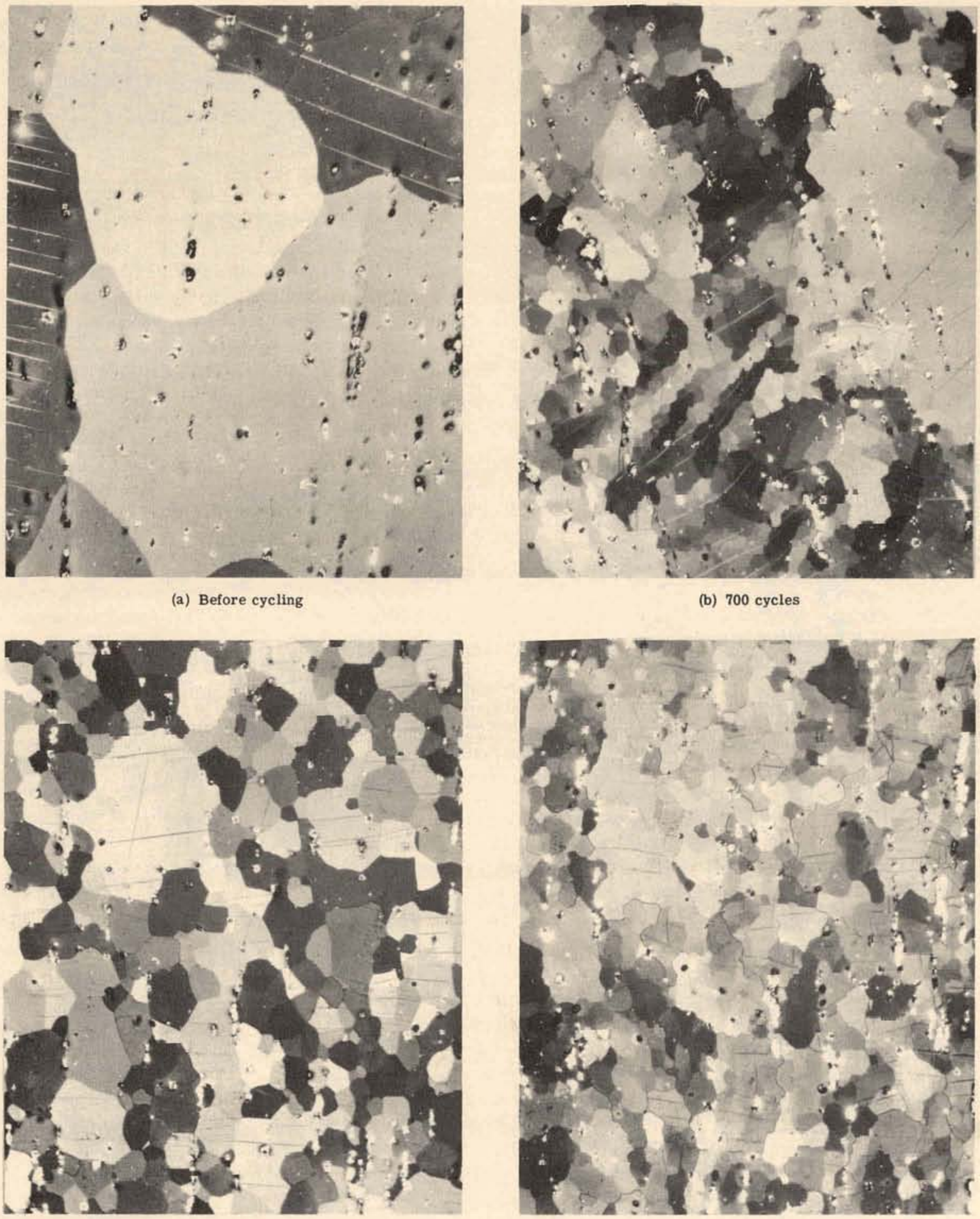
Early test results indicated that the elongation of rolled uranium rods on thermal cycling was a function of the degree of preferred orientation and that any treatment which produced a random orientation would minimize the tendency to elongate. To examine this possibility, several standard alpha-rolled rods (4, 6, and 10) and 300°C-rolled rods (21-24, 25-28, 31-34, and 35-38) were annealed for 2 hours at 725°C and slow cooled. As shown in Tables 1 and 3, their tendency to grow on cycling was practically eliminated by the beta anneal. However, the anneal produced extremely large grains as shown in Fig. 15a and the rods were badly warped and twisted and had rough gnarled surfaces after cycling as shown in Figures 5 and 6a.

Another possible method of obtaining a random structure in rolled uranium rods consisted of heating into the beta temperature range between passes while rolling at 600°C. Rods 82, 83, and 84 were rolled at 600°C with a 15-min soak at 725°C after every two passes. A more random structure was obtained, and the elongation on cycling was about half of that found in standard alpha-rolled rods as shown in Table 1.

It was apparent from the above tests that a fine-grained random structure was desirable to minimize both growth and surface roughening on cycling of uranium rods. Therefore, a method was sought whereby the grain growth could be

Table 7—Thermal expansion coefficients and elongation on cycling of coarse and fine grained specimens of similar orientation

Tensile bar No.	Grain size	Thermal expansion coef $\times 10^6/°C$ ∥R.D.	⊥R.D.	Elongation (per cent) No. of 100 to 550°C cycles 58	100	300	700	Elongation coefficient $\frac{1}{L}\frac{dL}{dN}\cdot 10^6$
1	Coarse	4.8	17.3-20.3	1.11	2.03	5.80	15.2	200
	Fine	6.05	19.0-19.2	1.65	2.78	7.96	21.0	270
2	Coarse	4.05	19.1-19.8	0.97	2.21	5.07	13.8	180
	Fine	5.8	18.0-18.3	1.69	2.82	8.11	22.0	280



(a) Before cycling

(b) 700 cycles

(c) Before cycling

(d) 700 cycles

Fig. 11a-d—Microstructures of coarse and comparatively fine-grained specimens of similar orientation before and after cycling. Polarized light at 150×.

minimized during a beta heat treatment. An approach to the desired results was obtained by heating rapidly into the beta temperature range by dipping into a lead pot at 725°C, holding for a short time (2 min and ½ min were tried) and water quenching.

Several highly oriented rods were given this fast beta heat treatment, and in all cases, the growth on cycling was drastically reduced. A 2-min fast beta treatment of a rod (51) similar to the ones that grew as much as 64% in 1500 cycles ($K = 320 \times 10^{-6}$) resulted in only 2.7% elongation in the same number of cycles ($K = 13 \times 10^{-6}$) as shown in Figs. 3, 4, and 5. In another case, a ½-min fast beta treatment decreased the tendency of the highly oriented rods shown in Fig. 6a to grow from 31.8% ($K = 370 \times 10^{-6}$) to 1.24% ($K = 18 \times 10^{-6}$) in 700 cycles.

Since highly oriented rods that had been fast beta treated still showed a slight tendency to grow on cycling, a study was made to determine the effects of successive fast beta treatments. Highly oriented 300°C-rolled rods (75% reduction) were given 1, 2, 4, and 8 fast beta treatments to approximately 720°C for 2 min. Also, one of these rods was given one fast-beta treatment to 744°C. Growth results are given in Table 8. It is apparent that more than one fast beta treatment did not decrease the tendency for rods to elongate on cycling. The higher temperature fast beta treatment resulted in slight if any improvement in tendency to grow.

Table 8 — Effect of multiple fast-beta treatments and beta temperature on growth of uranium rods when cycled 700 times between 100°C and 550°C

Rod No.	Fast-beta treatments: Number	Fast-beta treatments: Temp (°C)	Elongation (per cent), Number of cycles: 50	100	300	700
5-7	1	744	0.16	0.38	0.83	1.62
10-12	1	722	0.22	0.46	1.16	2.28
15-17	2	718	0.23	0.50	1.28	2.71
20-22	4	718	0.30	0.59	1.46	3.19
31-33	8	718	0.26	0.47	1.29	2.69

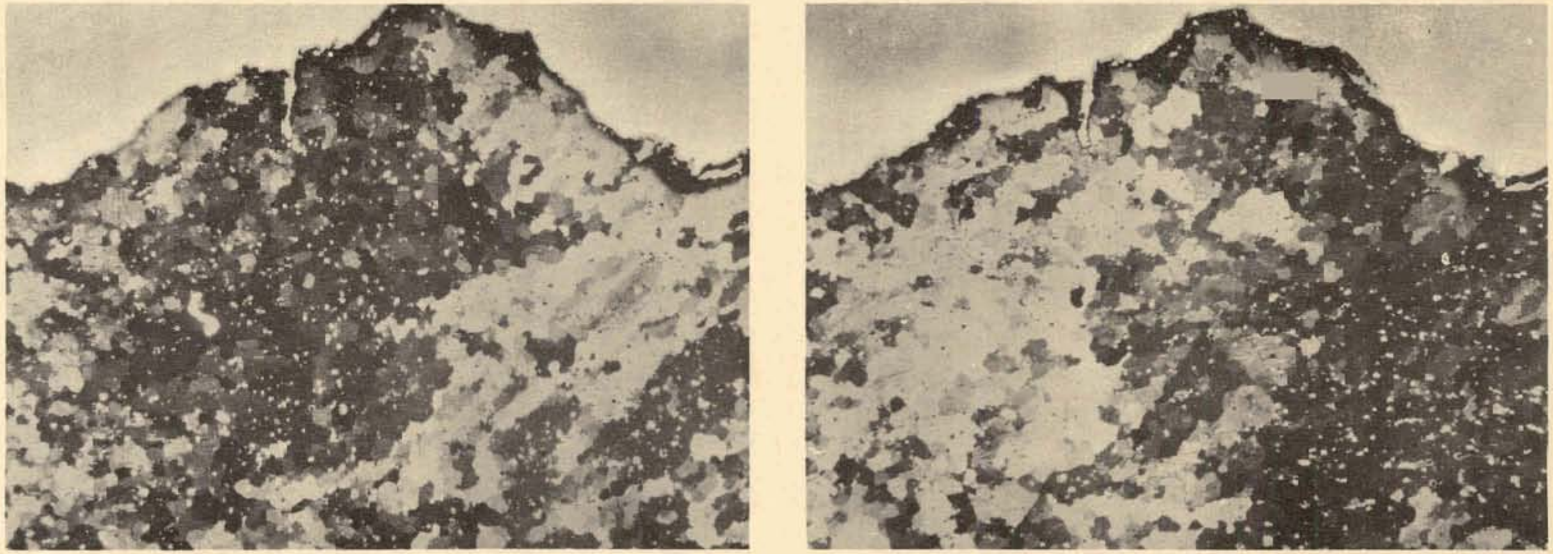
Surface Roughening

Comparison of Fig. 6a with Fig. 6b shows the general relationship between the amount of surface roughening on cycling and the grain structure of all rods examined during this investigation. The coarser the grain structure, the greater the surface roughening. This criterion can be used to predict whether or not a rod will roughen on cycling. Metallographic sections through surface bumps (Figs. 12 and 13) show that large clustered areas of small grains are directly associated with the bumps. Although inspection under low magnification gives a general idea of the size of clusters, it is often difficult to relate one cluster to a surface bump. As shown in Figs. 12a and 12b, rotation of the stage under polarized light causes groups of grains within a cluster to shift to adjacent clusters resulting in new apparent cluster boundaries. The cluster behaves mechanically like a large grain. This was found to be the case even after rods had been cycled many times. For example, cycling coarse-grained castings 1700 times resulted in the extremely fine-grained clustered structure shown in Fig. 18b. However, specimens that were machined from the cycled cast rods and then recycled 700 times surface roughened as badly as the original rods.

The surfaces of all fast beta treated rods showed a slight tendency to roughen after a large number of cycles which can be attributed to the coarsening of the grain structure as shown in Figs. 6b and 16a.

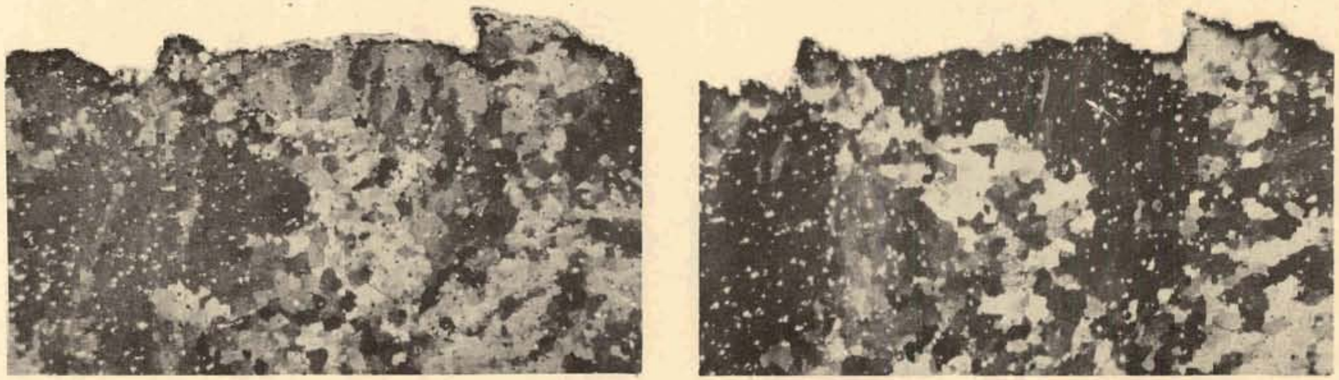
Preferred Orientation

The effect of heating alpha-rolled uranium rods into the beta region is two-fold: (1) the preferred orientation is almost completely removed, and (2) the grain size is increased and a new type of texture described as "clustering" appears. The



(a) (b)

Fig. 12a-b — Section through a large surface bump showing a cluster of similarly oriented subgrains associated with the bump. Note the similarity in the size of the bump and the cluster. Micros (a) and (b) represent different stage rotations under polarized light at 50×.



(a) (b)

Fig. 13a-b — Section through two small surface bumps. Note the cluster of subgrains between the bumps, also that each bump appears at the end of a long cluster. Micros (a) and (b) represent different stage rotations under polarized light at 50×.

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fast beta heat treatment resulted in a moderate to coarse alpha grain structure with a random orientation, but within each alpha grain a number of subgrains of nearly parallel orientation existed. This type of structure produced discontinuous Debye rings, the individual diffraction spots were closely grouped but the groups occurred at random around the ring (Fig. 14e). The slow-beta heat treatment resulted in a much coarser grain structure as shown in Fig. 14c.

The effect of thermal cycling was to increase the degree of scatter within each individual cluster as shown by the greater uniformity of the Debye rings (Figs. 14d and 14f).

Table 9 —Thermal expansion coefficients of uranium rods representing the heat treated 300°C-rolled uranium rods shown in Fig. 6a

Heat treatment	Rod No.	Thermal expansion coef × 10⁶/°C ‖R.D. Length	⊥R.D. Diam #1	⊥R.D. Diam #2	Elongation coefficient $\frac{1}{L}\frac{dL}{dN}\cdot 10^6$
Annealed 2 hrs at 575°C	1-4	4.9	16.5	17.1	280
	11-14	5.4	16.1	16.4	360
Annealed 2 hrs at 725°C, then annealed 2 hrs at 575°C	21-24	12.9	16.0	15.2	24
	31-34	14.3	11.9	13.4	50
½ min at 725°C, water quenched, then annealed 2 hrs at 575°C	41-44	13.2	15.4	12.1	(*)
	51-54	12.6	14.6	12.6	18

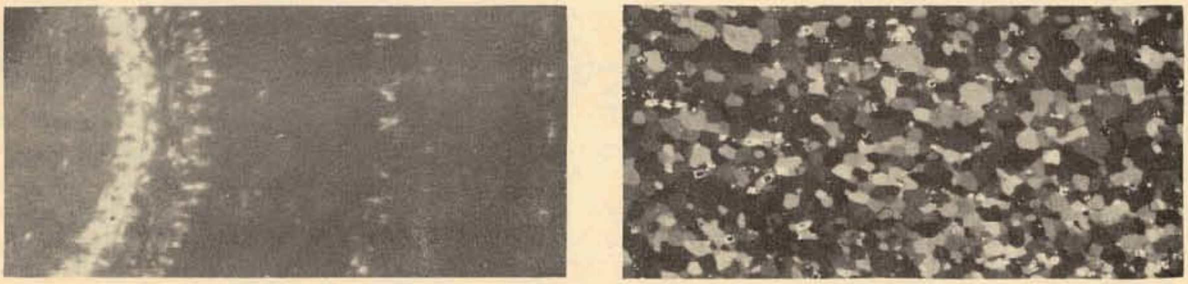
*Coefficients too small to be measured accurately.

As an average measure of preferred orientation, dilatometric measurements of the anisotropy in the coefficients of thermal expansion of specimens representing the 300°C-rolled and heat-treated rods, shown in Fig. 6, were made by Schwope and co-workers at Battelle Memorial Institute. The expansion coefficients along the length of the rods (the rolling direction) and across two diameters (perpendicular to the rolling direction) are given in Table 9 along with average elongation coefficients on thermal cycling. These results indicate that the high anisotropy of the annealed rolled rods as well as their tendency to grow on cycling was greatly reduced by the slow and fast beta treatments.

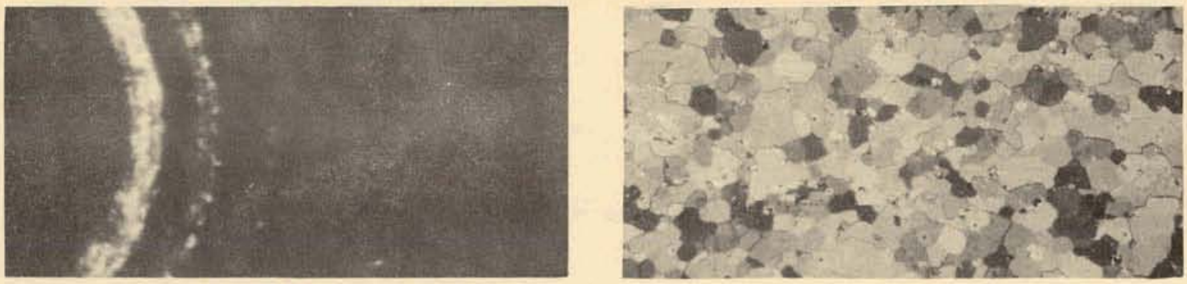
Microstructure

The effect of thermal cycling on the microstructures of the slow beta treated and fast beta treated 300°C-rolled uranium rods of Fig. 6a are shown in Figs. 15 and 16. A 300°C-rolled rod that had been annealed at 725°C (31-34) had a coarse grain structure before cycling. Rotation of the stage under polarized light revealed a macromosaic pattern of subgrains within the large grains which were so closely oriented that they were hardly distinguishable. After 100 cycles it was much easier to distinguish between the small subgrains in some of the original large grains, but most of the structure was not changed as shown in Fig. 15b. After 300 cycles the new grains were apparent throughout the structure, Fig. 15c, and after 700 cycles they were so prominent that it was difficult to determine the original large grains. However, at low magnification, it was apparent that the new grains were still clustered in areas of about the same size as the original grains. Study of these new grains formed by cycling indicates that, even though they became more randomly oriented within patches as cycling progressed, their size was not much affected by cycling and they appeared simultaneously across entire areas rather than at grain boundaries. This suggests that they were not formed by nucleation and growth.

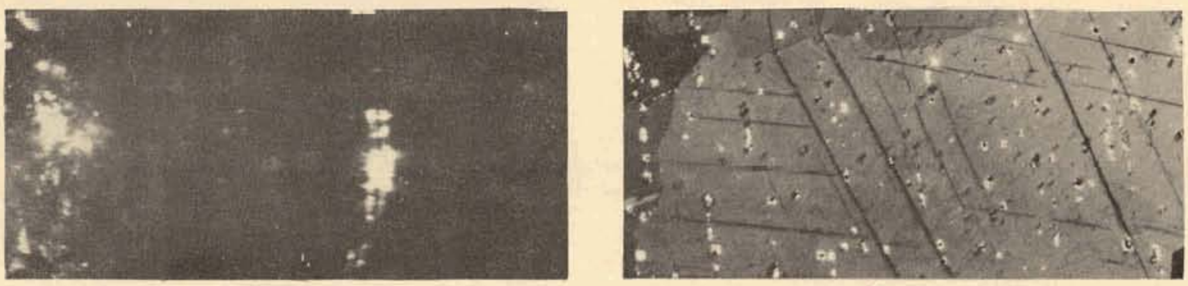
Water quenching a 300°C-rolled rod (41-44) after a 30-sec dip in a lead pot at 725°C and then 575°C annealing resulted in the confused looking structure shown in Fig. 16a. The boundaries of large areas were extremely jagged, and rotation under polarized light revealed numerous subgrains of various sizes and orientations clustered



(1-4) (a) 575°C anneal—before cycling (1-4)



(5-8) (b) 575°C anneal—after 700 cycles (1-4)



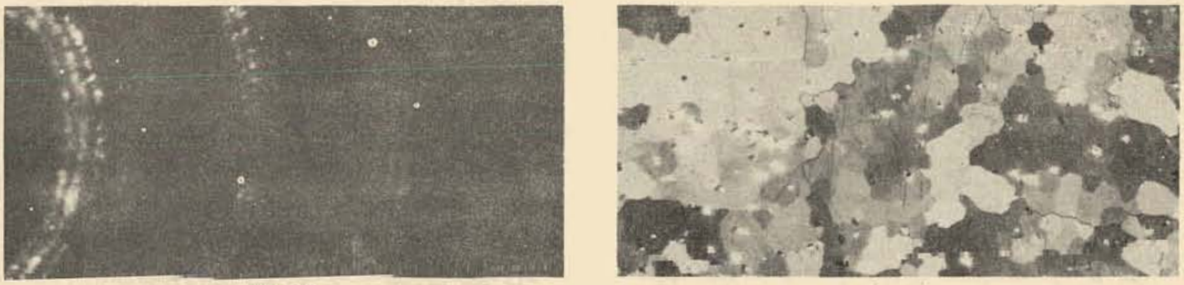
(31-34) (c) Slow beta treated—before cycling (31-34)

Fig. 14a-c —Comparison of x-ray diffraction structures and microstructures before and after cycling of rods shown in Fig. 6a. Photomicrographs under polarized light at 150×. X-ray photograms 2×.

within these areas. There was no appreciable change in this structure after 100 cycles, but small equiaxed subgrains were easily distinguished across entire areas after 300 cycles. Further cycling accentuated this new structure to give the fine-grained clustered structure shown in Fig. 16d.

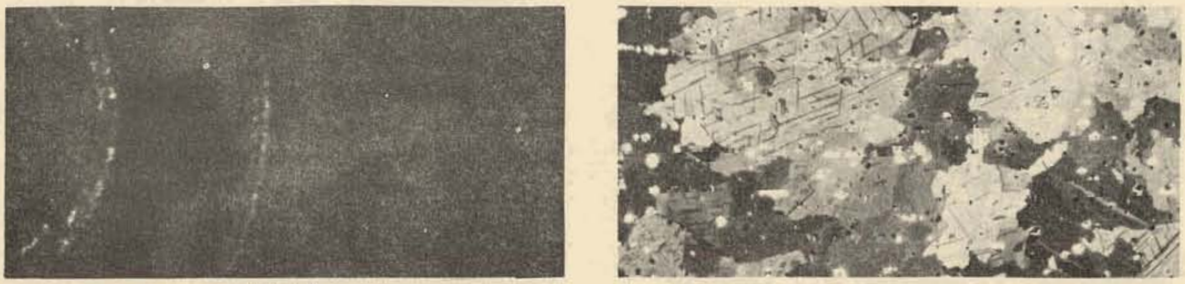
Further cycling of uranium rods similar to the slow beta treated and fast beta treated rods discussed above did not cause any further change in their microstructures. This was evident from the microexamination after 700 cycles of specimens which had been machined from rods that had already undergone 1500 cycles.

The above-described subgrain structure was also found after cycling coarse-grained uranium formed by the strain-anneal method as shown in Fig. 11b. The maximum temperature during the strain-anneal process was 650°C and the microstructure of the specimen before cycling as well as x-ray diffraction studies showed no indications of a subgrain structure. The formation of the substructure seen in cycled rods is, therefore, not dependent upon prior heating of the uranium into the beta region. Fig. 11b also shows the formation of similarly oriented parallel rows of subgrains within the large clustered areas. These rows are not parallel to rows of inclusions, but



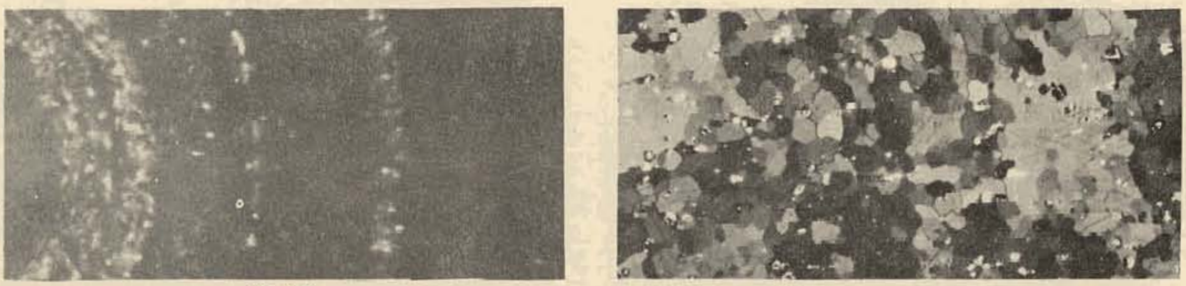
(35-38) (31-34)

(d) Slow beta treated—after 700 cycles



(41-44) (41-44)

(e) Fast beta treated—before cycling



(45-48) (41-44)

(f) Fast beta treated—after 700 cycles

Fig. 14d-f —Comparison of x-ray diffraction structures and microstructures before and after cycling of rods shown in Fig. 6a. Photomicrographs under polarized light at 150×. X-ray photograms 2×.

they are lined up parallel to deformation lines.

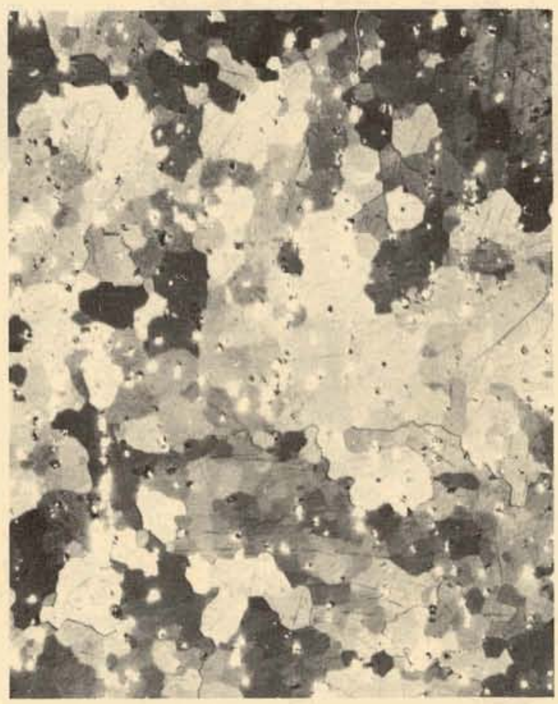
The subgrain structure described has been shown to form on cooling from the beta temperature region. Slow cooling from 725°C resulted in fairly large subgrains so closely oriented that careful metallographic technique was required to bring them out under polarized light (Fig. 15a). Water quenching from 725°C resulted in smaller subgrains which were easily observed under polarized light because of the greater scatter between adjacent subgrains (Fig. 16a). A test was made to determine whether or not the substructure could be formed by a single cooling from the high alpha region without cooling through the beta-alpha transformation. A uranium specimen containing both the coarse and fine-grained parts of a strain-annealed tensile bar similar to those shown in Figs. 10 and 11 and known to be free of the subgrain structure, was dipped in a lead pot at 633°C for ½ min and then quenched in water. The quenched specimen showed no signs of a substructure, and the only change in the microstructure was a slight increase in the number of straight deformation lines.

There is a marked similarity between the conditions which form subgrains in uranium and the conditions necessary to produce veining in alpha iron. Veining is the subboundary pattern often

(a) Before cycling

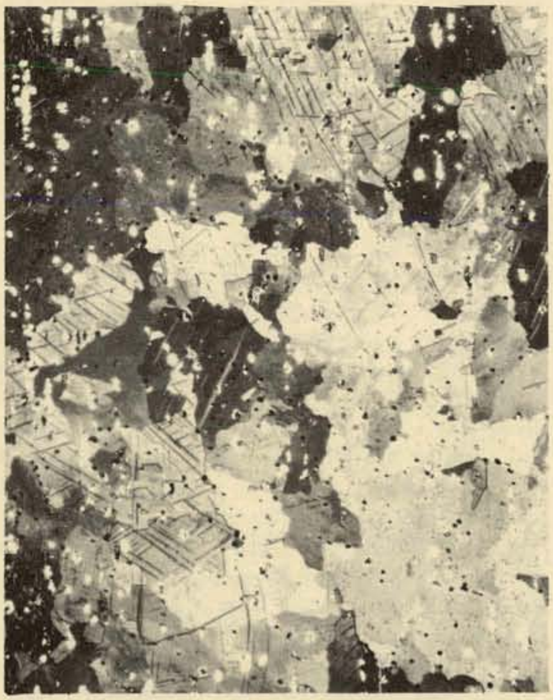
(b) 100 cycles

(c) 300 cycles



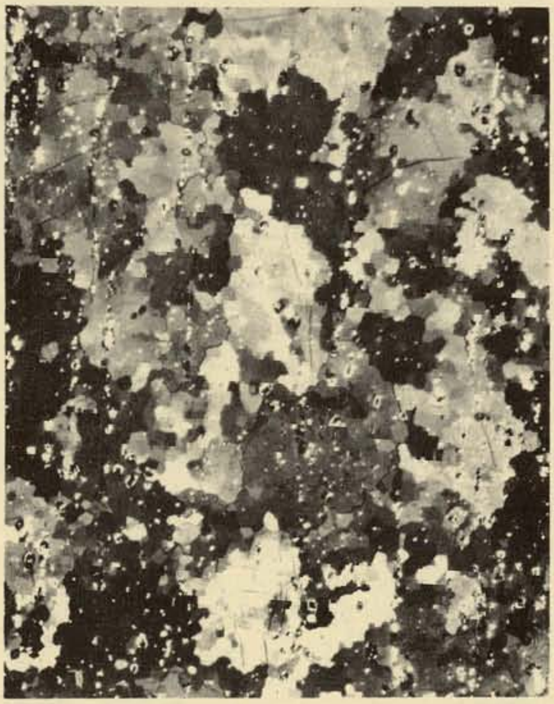
(d) 700 cycles

Fig. 15a-d — The effect of thermal cycling between 100°C and 550°C on the microstructure of a 300°C-rolled rod (31-34) that had been slow-beta treated. Polarized light at 150×.



(a) Before cycling

(b) 100 cycles



(c) 300 cycles

(d) 700 cycles

Fig. 16a-d — The effect of thermal cycling between 100°C and 550°C on the microstructure of a 300°C-rolled rod (41-44) that had been fast-beta treated. Polarized light at 150×.

found in ferrite if very careful metallographic technique is used. Greninger[28] has described the macromosaic structure in alpha iron and has pointed out that it is composed of many mosaic lattices which show a scatter in orientation from 1 to 10 degrees. The veining is attributed to the boundary lines of these closely oriented areas which can be considered as subgrains. Hultgren and Herrlander[29] have shown that veining is a deformation phenomenon resulting from plastic deformation of ferrite in the high alpha region. These investigators also caused veining in iron by rapidly cooling through the gamma-alpha transformation. In this case stresses due to the transformation were considered to cause plastic deformation of the ferrite already formed. They showed that the number of veins formed can be considerably reduced or veining altogether prevented by very slow cooling through the transformation.

700 cycles

Fig. 17—Typical curved deformation lines formed in 600°C-soak-rolled rod (54) as a result of cycling. Polarized light at 150×.

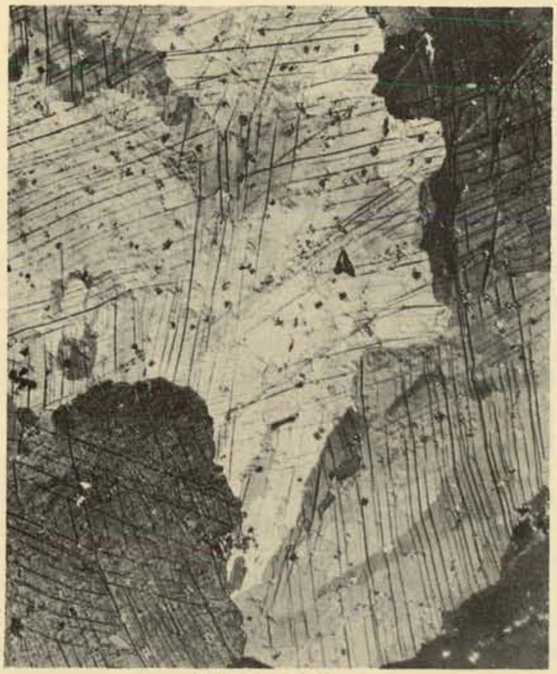
In an anisotropic metal such as uranium, a macromosaic structure can be brought out by the differences in intensity of reflected polarized light from adjacent subgrains. Therefore, recourse to tedious metallographic techniques to develop subgrain boundaries is not necessary as in the case of isotropic ferrite. The stresses necessary to produce plastic deformation at elevated temperatures resulting in veining in ferrite can be induced by external loading or by cooling through the gamma-alpha transformation. However, anisotropy of thermal expansion is a possible source of these stresses on thermal cycling of uranium at elevated temperatures. Hundreds of fairly slow cycles to 550°C produced a substructure in uranium which became more prominent with continued cycling. One rapid cycle to 633°C did not result in any perceptible substructure. It remains to be seen whether the formation of subgrains in uranium is a function of the rate of heating and cooling on cycling and whether it can be induced by external loading at elevated temperatures as in the case of ferrite.

An interesting effect of thermal cycling was the distortion of deformation lines in most of the cycled rods. Before cycling, these were straight lines crossing entire grains as shown in Figs. 11a and 15a. Cycling caused them to become peculiarly curved as they crossed clustered areas of subgrains. This is especially clear in Fig. 17 which represents a 600°C-soak-rolled rod (54) after 700 cycles. Closer inspection shows these curved lines to be made up of several short fairly straight lines each related to one of the subgrains formed on cycling.

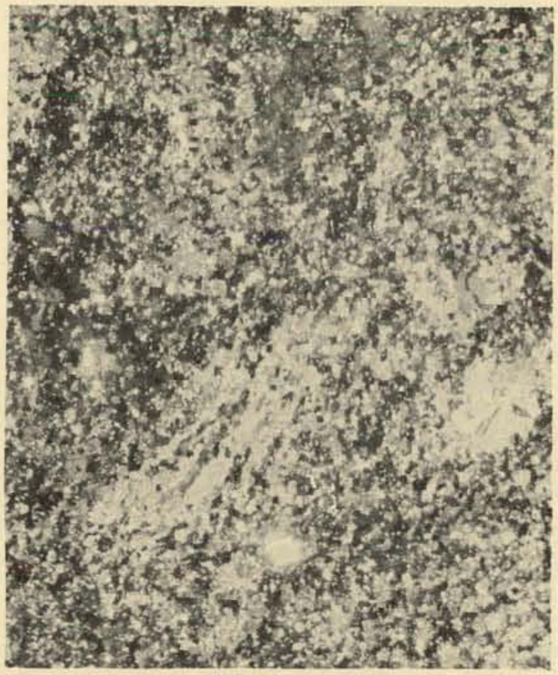
Cast, Swaged, and Extruded Rods and Rolled Plate

Cast, Swaged, and Extruded Rods

Other methods of fabrication used to produce rods for cycling included casting, swaging, and extruding. Elongation data showing the effect of cycling on these rods are given in Table 10. The cast rods (18 through 23) had a very coarse-grained structure as did a set of rods (59 through 66) which were supposed to have been alpha extruded but had apparently overheated during extrusion. Where a very large grain size occurred, no preferred orientation was assumed because the arcs occurred in random positions on the x-ray photograms. The accuracy of measuring the small dimensional changes noted for these rods was greatly influenced by extreme surface bumping and warping as shown in Fig. 5.



(a) Before Cycling



(b) 1700 cycles

Fig. 18a-b —The effect of cycling between 100°C and 550°C on the microstructure of a cast rod (22). Polarized light at 150×.

(a) Before cycling

(b) 700 cycles

Fig. 19a-b —The effect of cycling between 100°C and 550°C on the microstructure of a coarse-grained extruded rod (59). Polarized light at 150×.

A cast rod (22) had a coarse-grained structure with a secondary structure of fairly large subgrains which were easily distinguished as shown in Fig. 18a. In this case the structure was not appreciably changed after 300 cycles, but the extremely fine-grained structure shown in Fig. 18b was formed after 1700 cycles. These fine grains were clustered in areas about the size of the original large grains as could be seen by rotating the stage under polarized light. Continued cycling to 700 more cycles of rods machined from these 1500 cycled cast rods resulted in no further change in microstructure.

A coarse-grained extruded rod (59) (Fig. 19a) also contained small grains clustered in large areas after 700 cycles as shown in Fig. 19b. In this specimen some areas showed none of the small grains. The curved deformation lines described in connection with Fig. 17 were also apparent in this specimen.

A fast beta treatment might serve a dual purpose; that of randomizing an oriented structure and that of refining a coarse-grained structure. To examine the latter possibility, a fast beta treatment was given to cast rods and to coarse-grained alpha extruded rods similar to those described. The fast beta treatment resulted in much less surface roughening on thermal cycling, but some roughening was still apparent after 700 cycles.

Another set of alpha-extruded rods (740-1, 2, and 3) that were cycled were extremely fine grained and highly oriented. They elongated considerably (Table 10 and Figs. 3 and 4) and their surfaces remained smooth on cycling. The mean orientation and pole figures of these rods are shown in Fig. 20.

Several swaged rods (97, 98, 99, LA-3 and LA-4) were also cycled. These rods also had considerable preferred orientation as shown by the mean orientation and pole figures in Fig. 21. They elongated considerably on cycling (Table 10), and here again a fast beta treatment reduced the elongation in 700 cycles to a small amount, (LA-1 and LA-2).

Cycling of 300°C-Rolled Plate

To study the effect of thermal cycling on rolled uranium plate, two square specimens ¾ in. × ¾ in. × ⅛ in., which had been machined from a 300°C rolled plate (81.7% reduction—rolled in one direction), were cycled 700 times between 100°C and 550°C. The square specimens were cycled by the same procedure as that used for cycling rods except that they rested flat on a square face.

The specimens are shown in Fig. 22 after 700 cycles and dimensional changes during cycling are summarized along with coefficients of thermal expansion in Table 11. Flaring at the edges, which had been noted in cycling of rods, was very prominent in these specimens. In 700 cycles, the specimens elongated as much as 44.1% ($K = 500 \times 10^{-6}$) in the direction of rolling and contracted 14.6% ($K = -230 \times 10^{-6}$) perpendicular to the direction of rolling and 16% ($K = -210 \times 10^{-6}$) perpendicular to the plane of rolling.

The mean orientation of the rolled plate differs from that of the rolled rods in that the c-axis is at an angle of approximately 17 degrees to the normal to the plate with a small scatter in the rolling direction and complete scatter in the cross direction. The b-axis is at an angle to the rolling direction corresponding to 90 degrees from the c-axis with a scatter in both directions.

Effect of Cycling Variables

Temperature Interval

Most of the cycling tests for this investigation were made between 100°C and 550°C. However, to determine the effect of small thermal fluctuations at elevated temperatures, several rods were cycled 2200 times between 500°C and 550°C. These rods included the standard alpha-rolled and 575°C-annealed rods 40, 41, and 42, the standard alpha-rolled and 725°C rods 4 and 10 shown in Table 1 and the cast and 575°C-annealed rods 18, 19, and 20 shown in Table 10. Most of the rods had been previously cycled 100 times between 100°C and 550°C and growth due to this cycling is included in Table 12 for comparison with the results of cycling between 500°C and 550°C. It is apparent that rods which tend to grow on thermal cycling between 100°C and 550°C will also grow when cycled between 500°C and 550°C and that rods which tend to grow the most when cycled over a large temperature range will also grow the most when subjected to small thermal fluctuations at elevated temperatures. The rate of elongation on cycling over the large range was approximately 30 to 70 times as great as the rate on cycling over the small range.

The effect of low temperature cycling was studied by cycling 2000 times between 20°C and

300°C of 300°C-rolled rods that had been heat treated in three different ways. The heating and cooling was accomplished by mechanically transferring the rods in NaK-filled containers between hot and cold oil baths. Heating and cooling each took 5 minutes. The elongation data for this test time allowed at temperature and essentially the same rate of heating and cooling for each test. Equipment has been designed for the purpose of studying time-temperature relationships during cycling.[30] The role creep plays in deformation during cycling is being studied in this equipment.

Table 10—Elongation of cast, swaged, and extruded uranium rods due to thermal cycling between 100°C and 550°C

Fabrication and heat treatment	Rod No.	Rod length (in.)	Per cent elongation $\frac{L-L_0}{L_0} \times 100$: Due to anneal	Number of cycles (N): 50	100	300	700	Elongation coefficient $\frac{1}{L}\frac{dL}{dN} \cdot 10^6$
Cast—575°C-annealed	21	4	−0.02		−0.03	−0.08	(‡)	(††)
	22	4	−0.01		0.01	−0.02	(*)	(††)
	23	4	0.01		0.00	0.00	(*)	(††)
Cast—2 min at 723°C†, 575°C annealed	18	4	−0.03		(†)	−0.26	−0.58	(††)
	19	4			(†)	(‡)	(‡)	(††)
	20	4	−0.08		(†)	−0.26	−0.56	(††)
Swaged tapered castings, 30% to 50% reduction—575°C annealed	97	4		0.92	1.63	4.70	11.80	160
	98	4		0.91	1.67	4.78	12.03	160
	99	4			(‡)	(‡)	(‡)	(‡)
Swaged castings, 50% reduction (by LA)—575°C annealed	LA-3	2		1.01	1.96	5.38	16.7	200
	LA-4	2		0.95	1.83	5.09	16.0	200
Swaged castings, 50% reduction (by LA), 2 min at 725°C, water quenched—575°C annealed	LA-1	2	−0.08			0.31	1.37	17
	LA-2	2	0.00			0.59	1.80	23
Alpha extruded (by MIT), fine-grained—575°C annealed	740-1	2	0.34		2.44	5.23	9.72	180
	740-2	2	0.18		2.19	4.83	8.38	180
	740-3	2			2.12	(‡)	(‡)	210
Alpha-extruded (by MIT), coarse-grained—575°C annealed	59	4	−0.01		0.11	0.37	1.07	14
	60	4	0.02		−0.12	0.30	0.99	13
	61	4	0.03		0.17	0.29	0.86	12
Alpha-extruded (by MIT), coarse-grained—2 min at 723°C, water quenched, 575°C annealed	64	4	0.01		0.16	0.66	1.64	23
	65	4	0.01		0.12	0.57	1.41	19
	66	4	0.01		(‡)	(‡)	(‡)	(‡)

*

Rod No.	No. cycles: 500	900	1700
22	−0.05	0.13	0.91
23	0.10	0.35	0.73

†Cycled 100 times between 100° and 550°C and then 2200 times between 500° and 550°C before fast beta treatment.

‡Rod cut up for samples at various stages of cycling.

††Coefficients too small to be measured accurately.

are compared in Table 13 with the elongation of similar rods when cycled between 100°C and 550°C. The rate of elongation on cycling between 100°C and 550°C is seen to be approximately 100 times as great as when cycled between 20°C and 300°C.

Time-Temperature Relationships

For all previously described thermal cycling tests a saw-tooth type of cycle was used with no Also, very high thermal gradients can be induced by fast heating and cooling in this equipment, and their effects are under investigation.

SUMMARY

The similarity in dimensional changes of uranium due to thermal cycling and that due to pile exposure suggests a common cause. This, together with the practical and fundamental interest in the problem of obtaining stress-free, dimensionally stable uranium and other anisotropic metals has motivated this investigation of thermal cycling effects.

Uranium rods prepared by a wide variety of fabrication and heat treat methods were thermal cycled between 100°C and 550°C in a vertical position under conditions roughly simulating CP-4 pile conditions. Rolled uranium rods elongated when thermal cycled, and their diameter changes were such as to just compensate for the length changes. In general, the greater the reduction in area during rolling and the lower the rolling temperature, the greater the rate of growth. The amount of longitudinal growth increased directly as the number of thermal cycles with no indication of change in rate of growth after 1500 cycles. Rods rolled at 300°C elongated as much as 64% in 1500 thermal cycles between 100°C and 550°C.

The degree of preferred orientation was the major factor influencing the rate of elongation of rolled uranium rods on cycling. There were some indications that grain size or some other unknown factor influenced the rate of growth, but these had minor effects. Fast heating into the beta temperature range followed by water quenching almost completely removed preferred orientation in rolled uranium rods and greatly decreased the tendency for growth on cycling. However, a moderate to coarse-grained structure resulted from this treatment, and the rods roughened slightly on cycling. Highly oriented rods that were given a fast beta heat treatment still had a slight tendency to elongate on cycling, and as many as eight such treatments on the same rod did not remove this tendency to elongate.

Thermal cycling of coarse-grained uranium rods resulted in rumpled and sometimes badly

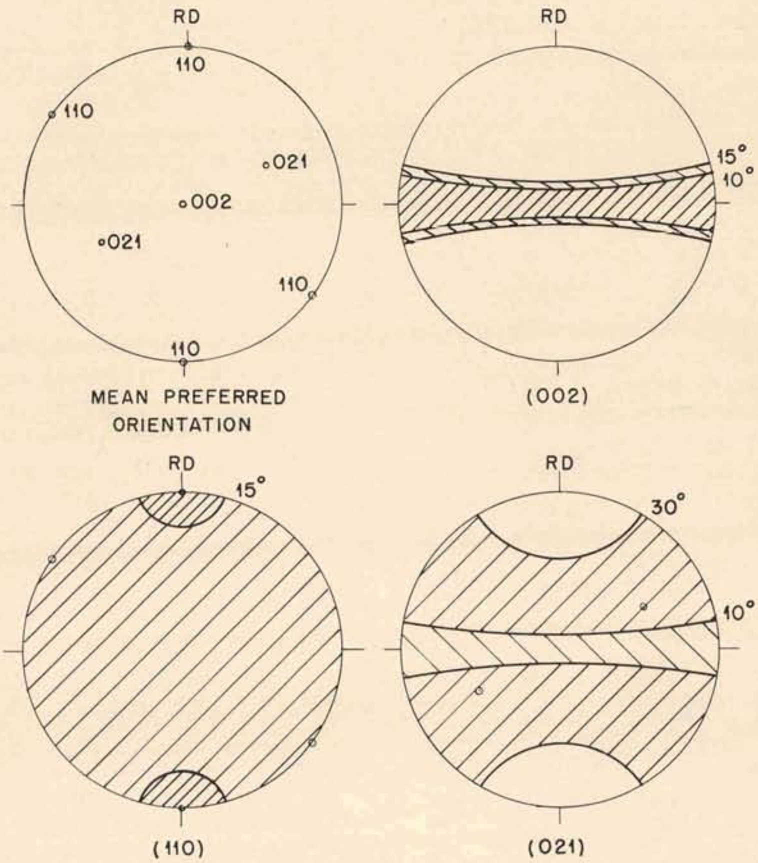


Fig. 20—Pole figures of alpha-extruded and 575°C-annealed uranium rod No. 740. (X-ray specimen No. 622)

bumped surfaces whereas fine-grained rods remained smooth on cycling.

A substructure of similarly oriented grains clustered within larger grains was noted in uranium that had been heated into the beta region and also in uranium that had been thermal cycled in the alpha region. Slow cooling after annealing in the beta region resulted in subgrains that were very closely oriented, but a fast beta heat treatment resulted in considerably greater scatter of adjacent grains within each cluster. This substructure was not developed by water quenching from 633°C of a coarse-grained uranium specimen prepared by the strain-anneal technique. However, the subgrains were found in all medium and coarse-grained uranium after thermal cycling, and the scatter between adjacent subgrains increased as cycling progressed. These clustered areas behaved mechanically as individual grains and were directly associated with surface bumps. The conditions which cause the formation of a substructure in uranium are very similar to the conditions necessary to produce veining in alpha iron.

Specimens prepared by other fabrication methods which caused preferred orientation also elongated on thermal cycling. These included alpha-extruded rods, swaged rods, and rolled plate.

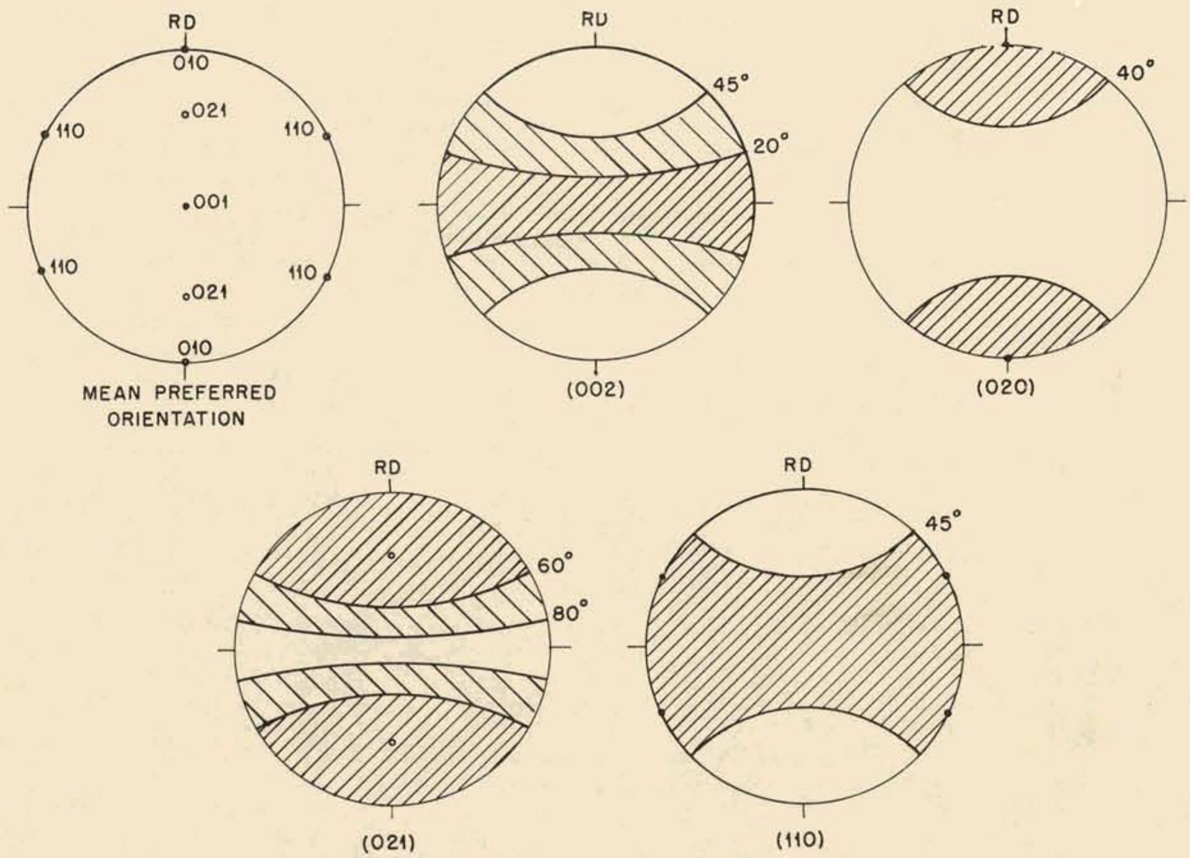


Fig. 21—Pole figures of swaged and 575°C-annealed uranium rod No. 99. (X-ray specimen No. 622)

Small thermal fluctuations at elevated temperatures caused elongation of oriented uranium rods. The rate of elongation on cycling between 500°C and 550°C was considerably less than that due to cycling between 100°C and 550°C. Thermal cycling between 20°C and 300°C also caused these rods to elongate but at a slower rate than when cycled between 100°C and 550°C.

From the results of this investigation, it is apparent that a randomly oriented, fine-grained

Table 11 — Dimensional changes of 300°C-rolled uranium plate due to thermal cycling between 100°C and 550°C

Direction	Sample No.	Exp coef $\times 10^6/°C$	Elongation (per cent) Number of cycles 50	100	300	700	Elongation coefficient $\frac{1}{L}\frac{dL}{dN} \cdot 10^6$
Parallel rolling direction	2	6.6	2.44	5.03	15.7	44.1	500
	3		2.50	4.93	15.4	43.0	470
Perpendicular rolling direction	2	15.7	−1.38	−2.49	−6.99	−14.6	−230
	3		−1.30	−2.62	−7.04	−14.0	−240
Perpendicular rolling plane	2	18.6	−0.72	−2.10	−6.16	−16.0	−210
	3		−0.79	−1.93	−5.86	−15.0	−200

Table 12 — Growth of uranium rods due to small thermal cycles at elevated temperatures

Fabrication and heat treatment	Rod No.	100-550°C (100 cycles) Elongation (per cent)	100-550°C (100 cycles) Elongation coefficient $\frac{1}{L}\frac{dL}{dN} \cdot 10^6$	500-550°C (2200 cycles) Elongation (per cent)	500-550°C (2200 cycles) Elongation coefficient $\frac{1}{L}\frac{dL}{dN} \cdot 10^6$
Standard alpha-rolled	43			0.69	3.1
	44			0.56	2.5
	45			0.36	1.6
575°C-annealed	40	2.45	200	0.66	3.0
	41	1.95	160	0.74	3.4
	42	3.60	270	1.89	8.5
Standard alpha rolled and 725°C-annealed	4	−0.04	(*)	0.06	(*)
	10	−0.02	(*)	0.05	(*)
Cast and 575°C-annealed	18	−0.04	(*)	0.00	(*)
	19	0.00	(*)	0.00	(*)
	20	0.00	(*)	0.00	(*)

*Coefficient too small to be measured accurately.

Table 13 — Growth of uranium rods due to low temperature cycling

Fabrication and heat treatment	Rod No.	100-550°C (100 cycles) Elongation (per cent)	100-550°C (100 cycles) Elongation coefficient $\frac{1}{L}\frac{dL}{dN} \cdot 10^6$	Rod No.	20-300°C (2000 cycles) Elongation (per cent)	20-300°C (2000 cycles) Elongation coefficient $\frac{1}{L}\frac{dL}{dN} \cdot 10^6$
575°C-annealed	7B	2.84	280	2A	0.543	2.7
				5A	0.495	2.4
				6A	0.493	2.4
725°C-annealed, then 575°C-annealed	7A	0.31	31			
	9B	0.26	26	3A	0.020	(*)
	9C	0.52	56	4A	0.024	(*)
Fast-beta treated, then 575°C-annealed	7C	0.22	22			
	8B	0.12	12	1A	0.000	(*)
	8C	0.06	06			

*Coefficients too small to be measured accurately.

structure is desirable to minimize both growth and surface roughening of uranium on thermal cycling. The desired results are approached by a fast beta treatment of 300°C-rolled rod.

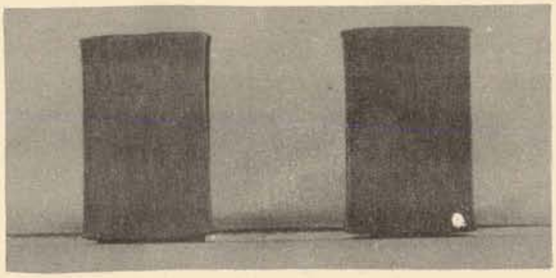


Fig. 22—Rolled uranium plates after 700 cycles between 100°C and 550°C. The specimens were square before cycling (from M-4312). 1×.

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