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Metallurgy and Ceramics

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# KINETICS OF THE BETA TRANSFORMATION OF URANIUM

by

W. R. McDonell

Pile Materials Division

September 1957



E. I. du Pont de Nemours & Co.
Explosives Department - Atomic Energy Division
Technical Division - Savannah River Laboratory

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METALLURGY AND CERAMICS
(M-3679, 20th Ed.)

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## ABSTRACT

When uranium is heat treated at low beta-phase temperatures and air cooled, the rate of loss of preferred orientation is markedly less than the rate of coarsening of the grain structure. A coarse-grained structure cannot therefore be taken as an infallible indication of satisfactory heat treatment.

The final grain size of incompletely transformed metal is essentially equal to or larger than the starting grain size. Large-grained specimens can not be refined by interruption of the transformation at an intermediate stage.

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# KINETICS OF THE BETA TRANSFORMATION OF URANIUM

## INTRODUCTION

The term beta transformation designates a heat treatment applied to uranium fuel elements. Heating at beta-phase temperatures, 668 to 774°C, largely eliminates the preferred crystallographic orientation that is responsible for the dimensional instability of wrought uranium under irradiation[1]. Beta-transformed metal is in general stable with respect to gross dimensions under reactor exposure[1].

The beta transformation results, however, in a large-grained microstructure which is susceptible to a second type of dimensional instability, a surface roughening[1]. In fuel elements designed to close tolerance such a surface roughening may be detrimental. At high burnups the effect contributes to a gross distortion and mechanical deterioration of the fuel element[2].

The "ideal" structure for a uranium fuel element is thus one with a random crystallographic orientation and small grain size.

The purpose of the research which is reported here was to study the rate of the loss of preferred orientation and the rate of grain coarsening during beta heat treatment to determine if conditions exist for the production of this ideal structure. The study was prompted by the preliminary observation that during beta transformation, fine-grained, as-rolled metal passes through a transition stage of intermediate grain size. It was considered possible that such transition structures had lost the preferred orientation of the as-rolled metal.

## SUMMARY

The rate of loss of preferred orientation* during the heat treatment of uranium at low beta-phase temperatures was markedly less than the rate of coarsening of the grains. A coarse-grained structure cannot, therefore, be taken as an infallible indication of complete beta transformation. Short heating times at low beta-phase temperatures, followed by air cooling, produced metal which had the large-grained structure typical of beta-transformed metal, but which retained a high degree of preferred orientation that would make it dimensionally unstable on irradiation.

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* In this research, the anisotropy of the thermal expansion of the plate was taken as a measure of the preferred orientation.

To explain the difference between the rate of loss of preferred orientation and the rate of grain coarsening, it is postulated that halting the transformation at an intermediate stage by cooling leaves residual alpha-phase metal to act as nucleation centers for the regrowth of an oriented but large-grained structure.

Intermediate-sized grains were produced by halting the transformation of small-grained metal at an early stage. A similar interruption of the transformation of initially large-grained metal did not result in a significant refinement of grain size.

## DETAILS

### MATERIALS

The metal used in these studies was taken from wrought uranium plate, of 0.180-inch thickness, rolled at high alpha-phase temperatures at Superior Steel Company in August 1953 and February 1954[3]. This metal has been shown to have a strong preferred orientation, with a large fraction of (010) planes perpendicular to the rolling direction[4]. In the as-rolled condition the metal was completely recrystallized to uniform, equiaxed grains of about 0.030-mm diameter.

Standard beta transformation, heating for two minutes at 720°C in a salt bath and cooling in air, produced a structure which was nearly randomly oriented with grains ranging in diameter from 0.010 to greater than 0.600 mm. In this structure, over half the volume of metal was contained in grains greater than 0.300 to 0.400 mm in diameter[5]. This structure was designated a "large-grained" structure. Microstructures of typical as-rolled and beta-transformed metal are shown in Figure 1.

### EXPERIMENTAL PROCEDURE

The rate of loss of preferred orientation and the rate of grain growth were measured as functions of the beta-transformation temperature. Preferred orientation was characterized by a dilatometric technique which will be described. Grain size was characterized metallographically.

The specimens used were standard dilatometer specimens, cylinders 3.5 mm in diameter and 50 mm in length.

#### Heat Treatments

The specimens were beta transformed by immersion in a molten salt bath of eutectic $Li_2CO_3$-$K_2CO_3$. The salt temperature was measured by a chromel-alumel thermocouple in a stainless steel well immersed in the molten salt. The precision of the absolute temperature measurements was ±3°C, but relative measurements were accurate to ±1°C.

The specimens were suspended in the salt by means of nichrome wire. The time necessary for the specimen to reach salt temperature was less than one minute.

Following beta treatment, the specimens were air cooled through the transformation temperature, then quenched in tap water.

### Characterization of Preferred Orientation by the Dilatometric Technique

In the dilatometric technique, preferred orientation is characterized by the degree of anisotropy of the thermal expansion coefficients[4]. The thermal expansions of specimens oriented longitudinally and transversely to the rolling direction of the plate are measured. The highly oriented as-rolled plate, with a strong (010) texture, shows a low thermal expansion coefficient in the longitudinal direction and a correspondingly high coefficient in the transverse direction. Beta transformation of the plate produces metal with nearly the same thermal expansion coefficient in longitudinal and transverse specimens.

The thermal expansion was measured with a Leitz HVT dilatometer, using a standard technique previously described[4]. Dilatometer curves that recorded specimen expansion versus temperature were obtained over a temperature range of 25 to 550°C.

### Characterization of Grain Structure by Metallography

The grain structure of the beta-transformed specimens was studied by standard metallographic techniques. Polished specimens were electropolished and observed under polarized light.

## RESULTS

### Comparison of Rate of Loss of Preferred Orientation and Rate of Grain Growth

The loss of preferred orientation on beta transformation at low beta-phase temperatures occurred at a relatively slow rate. The dilatometer curves for longitudinal and transverse specimens indicated a gradually decreasing anisotropy of thermal expansion coefficient as the time of heating at a given temperature was increased. Typical dilatometer curves for as-rolled, partially transformed, and completely transformed metal are shown in Figure 2. Thermal expansion coefficients derived from the dilatometric curves of a series of specimens beta transformed at 675°C are given in Figure 3, and show that between 16 and 30 minutes were required to yield a nearly isotropic structure at this temperature.

At beta-transformation temperatures lower than 675°C, the loss of anisotropy proceeded at a decreased rate, while at temperatures higher than 675°C, the rate was increased. The anisotropy, as indicated by the difference in transverse and longitudinal thermal expansion coefficients at 300°C, is shown as a function of time for

various transformation temperatures in Figure 4. The temperatures and times for complete loss of preferred orientation are approximately as follows:

| Temperature, °C | Time, minutes |
|---|---|
| 670 | > 60 |
| 675 | 22 |
| 680 | 8 |
| 685 | 3 |

In contrast to this, the coarsening of grain size on beta transformation was fairly rapid. Microstructures of dilatometer specimens heated for various times at 675°C are shown in Figure 5 for comparison with the corresponding loss of preferred orientation shown in Figure 3. The temperatures and times necessary to obtain the large-grained structure characteristic of beta-transformed uranium were as follows:

| Temperature, °C | Time, minutes |
|---|---|
| 670 | 30-60 |
| 675 | 4 |
| 680 | 2 |
| 685 | 1 |

At 665°C, the grain coarsening was not complete after 60 minutes at temperature, but a 16-hour heating did produce the large-grained structure.

Intermediate structures showed that the grains progressively coarsened from the small equiaxed structure of the as-rolled metal to the large, irregular, and varying-sized grains of the beta-transformed state. The coarsening started at the outer edges of the specimen, but in general spread rapidly throughout the specimen to present a homogeneous intermediate-sized structure before the large-grained structure was obtained.

The heating to 550°C for the dilatometer run did not noticeably alter the grain structure of the specimens, as demonstrated by comparison with specimens not run in the dilatometer.

A comparison of the times and temperatures necessary for complete loss of preferred orientation and for complete grain coarsening during beta transformation is shown in Figure 6. This comparison showed that it was not possible by control of the degree of transformation to produce a randomly oriented structure without also producing a large-grained structure.

## Retransformation of Beta-Transformed Metal

An attempt was made to produce an intermediate-sized grain structure analogous to the transition structure of the partially transformed as-rolled metal, by interrupting the transformation of previously beta-transformed specimens at an intermediate point. Such specimens were initially isotropic so a partial retention of their crystallographic orientation would not be detrimental.

In no case could a significant refinement of the original large-grained structure be produced. Beta treatments were conducted over a range of times, one minute to sixteen hours, at temperatures 665 to 685°C. Evidence of a minor refinement of the previously beta-transformed structure was observed for times of 4 to 30 minutes at 670°C. Such a structure is shown in Figure 7. The effect was not a pronounced one, however, and may have been due to other conditions.

## Interrupted Transformation

A final experiment was undertaken to illustrate the impossibility of producing a refined grain structure by incomplete retransformation of previously transformed metal. Fine-grained, as-rolled specimens were heat treated for a series of two-minute intervals at low beta-phase temperatures. Between the beta-phase heatings, the specimens were allowed to cool in air for 30 seconds, a time sufficient for the sample to cool to alpha-phase temperatures. The heat treatments were conducted so as to produce a series of specimens with total beta-heating times of two to eight minutes, interrupted by cooling to the alpha phase and reheating every two minutes.

The microstructures of these specimens showed a progressive coarsening in grain size, in much the same fashion as the uninterrupted beta heatings (see Figure 8). This was true in spite of the fact that no specimen of the series had remained at beta-phase temperatures for more than two minutes at a time.

# DISCUSSION OF RESULTS

## Mechanism of Beta Transformation

A mechanism may be suggested to explain the retention of preferred orientation at beta-heating times and temperatures that produce large grain structures. It is hypothesized that the transformation from the highly oriented alpha phase to the beta phase is incomplete in short times at low beta-phase temperatures and that the residual alpha grains serve as nuclei for subsequent retransformation back to an oriented, but large-grained alpha phase.

The following sequence of events, illustrated in Figure 9, may occur. When the metal is heated to beta-phase temperatures, the nuclei of the beta phase form and start to grow at the expense of the surrounding alpha phase. If the transformation is interrupted by cooling at some time before completion, the residual alpha-phase grains can act as nuclei for the regrowth of alpha-phase metal. The residual

alpha nuclei preserve to some extent the preferred orientation of the as-rolled metal; their growth results in a large-grained structure possessing a preferred orientation similar to the as-rolled metal. This hypothesis is supported by the direct dilatometric observation that interruption of the transformation of a highly oriented specimen by cooling returns the specimen more or less to its original orientation[6].

Competing with untransformed alpha grains as nucleating centers for alpha-phase material are nuclei recrystallizing directly from the beta phase. The greater the degree of the original transformation to the beta phase, that is, the longer the time and the higher the temperature of beta heating, the more effectively would the direct nucleation process compete, and the lesser would be the retained preferred orientation. This corresponds to the facts observed. The conditions for complete transformation would be those which resulted in essentially complete loss of preferentially oriented metal.

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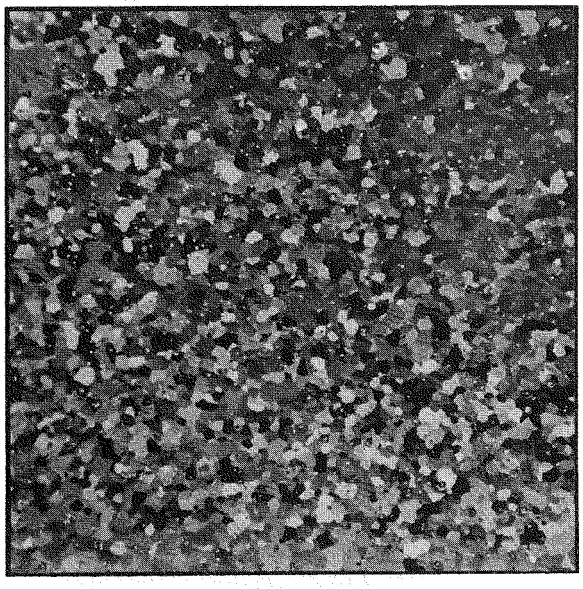
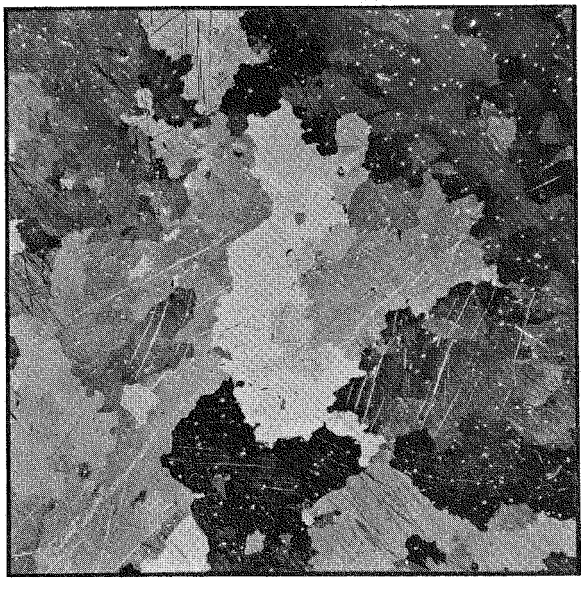
FIGURE 1



As-Rolled

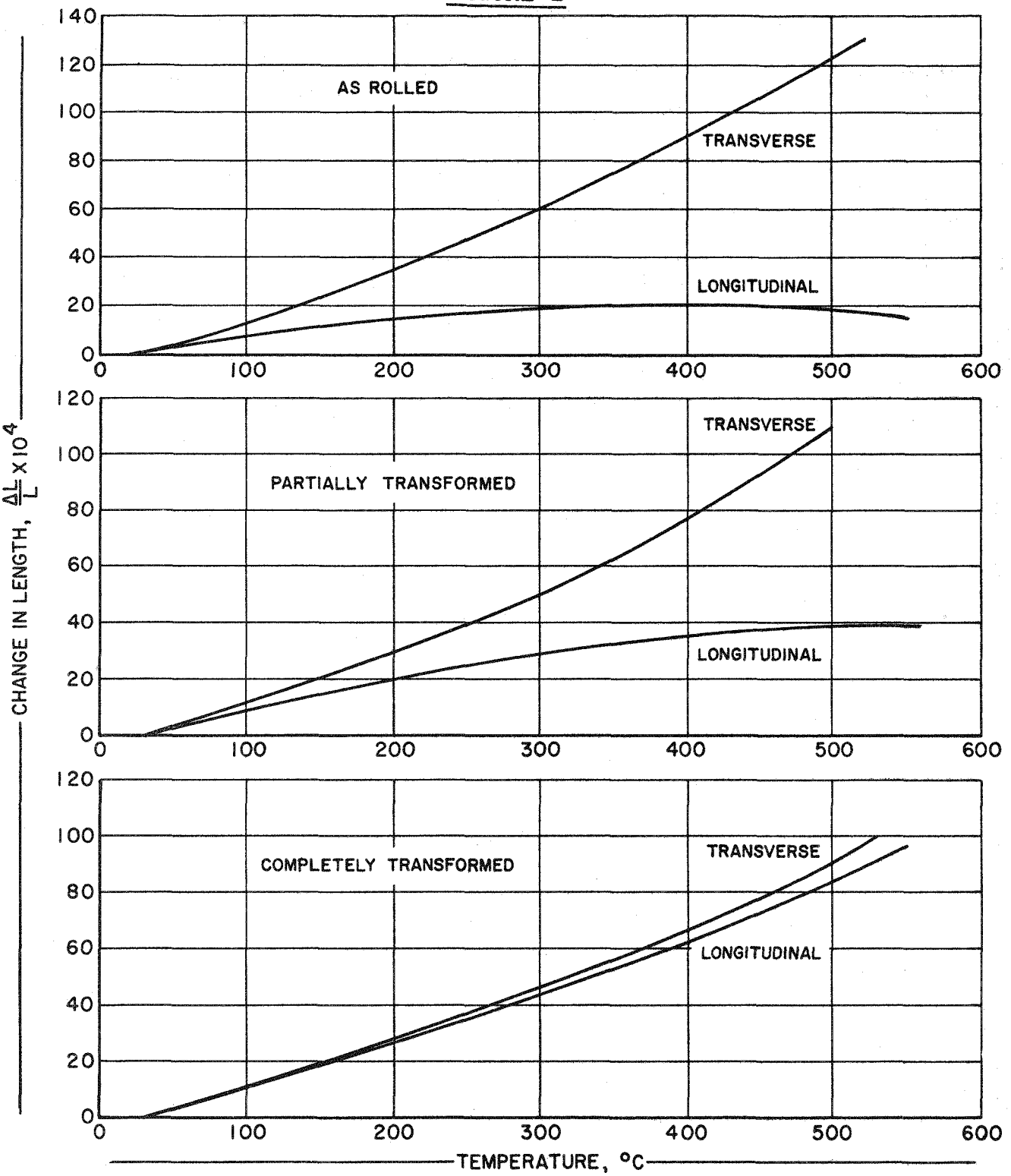


Beta-Transformed

GRAIN STRUCTURE OF AS-ROLLED
AND BETA-TRANSFORMED (AIR-COOLED) URANIUM PLATE

Mag. 50X

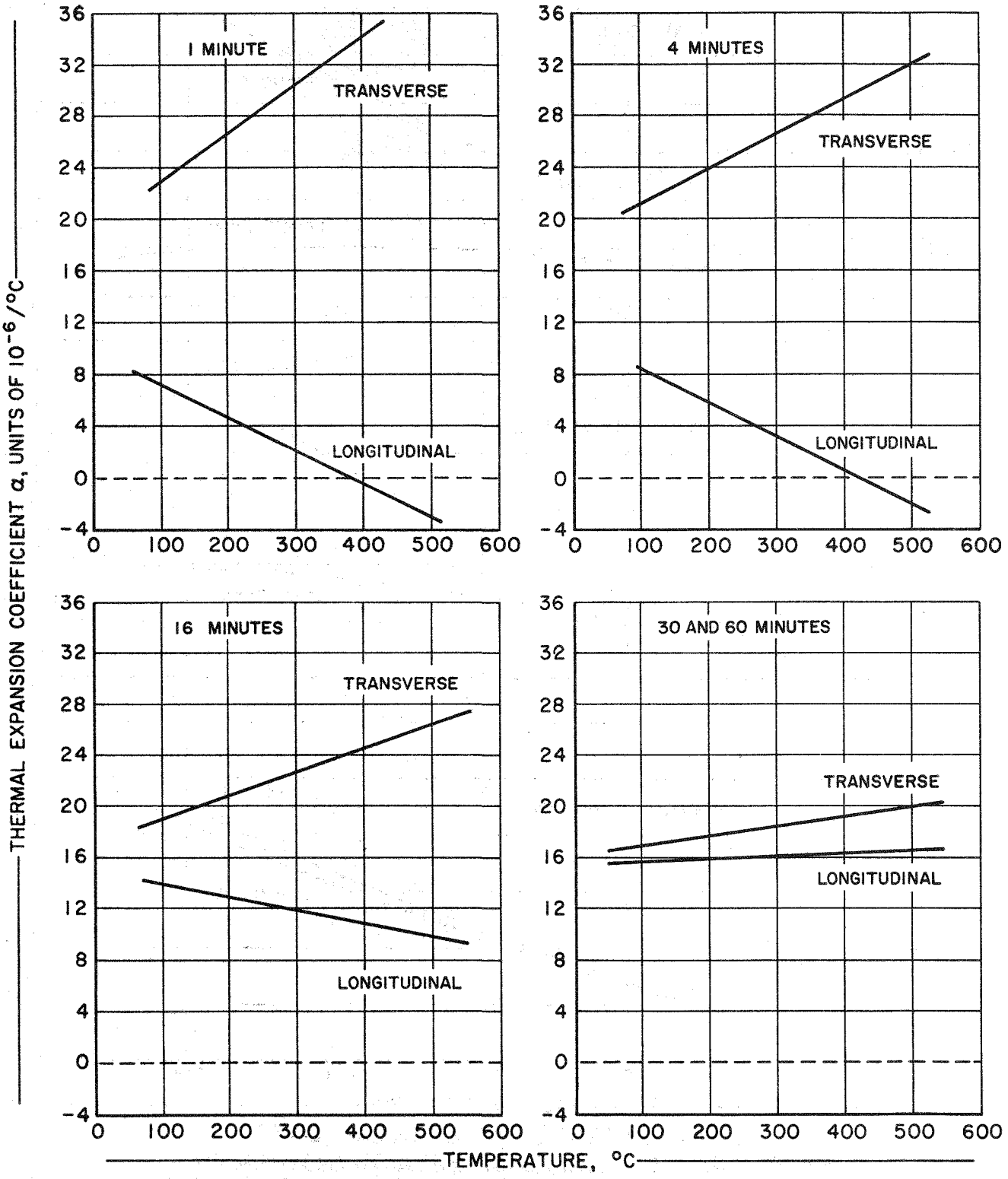
FIGURE 2




THERMAL EXPANSION OF URANIUM PLATE
AFTER VARIOUS DEGREES OF BETA TRANSFORMATION

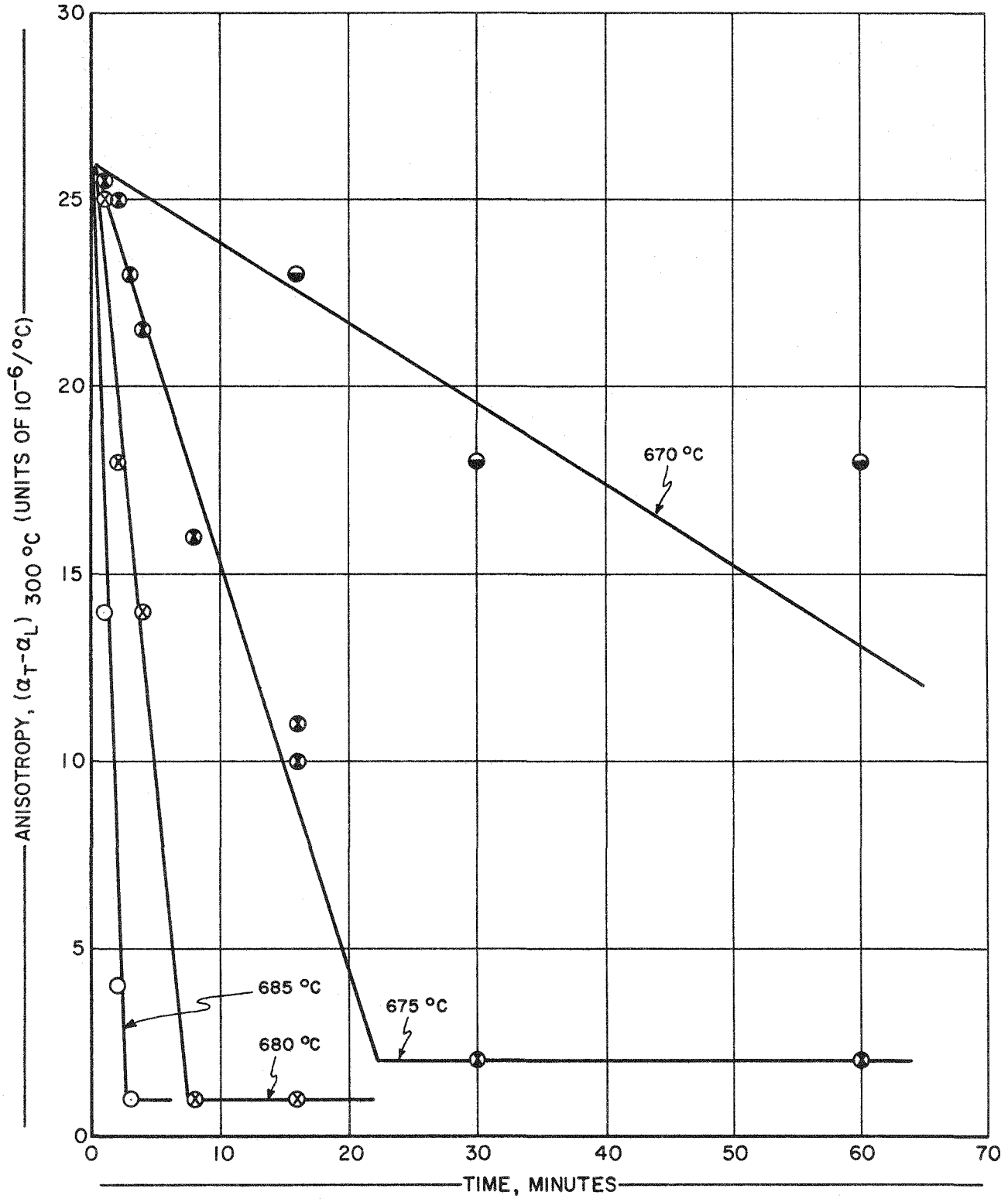
The expansions in the longitudinal and transverse directions of the plane of the plate are shown.

FIGURE 3




THERMAL EXPANSION COEFFICIENTS OF URANIUM PLATE
BETA TRANSFORMED FOR VARIOUS TIMES AT 675° C AND AIR COOLED

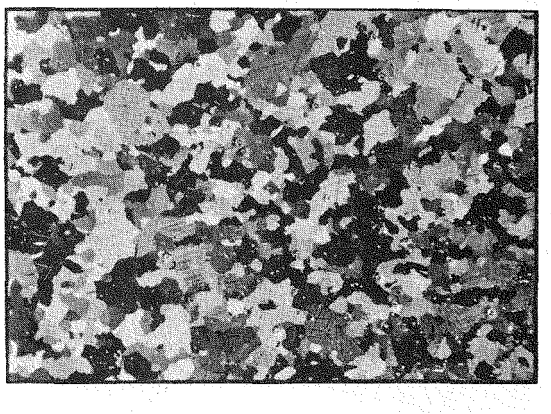
FIGURE 4



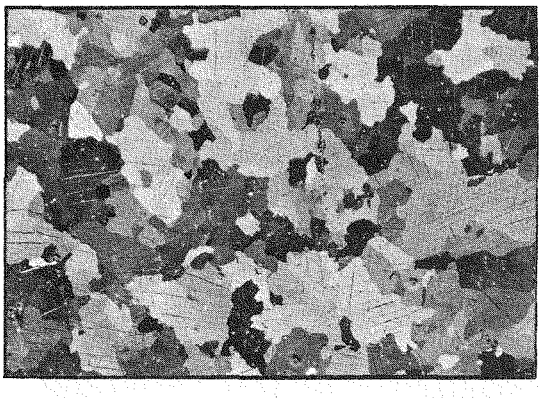

LOSS OF ANISOTROPY OF THERMAL EXPANSION COEFFICIENT AT VARIOUS BETA-TRANSFORMATION TEMPERATURES

FIGURE 5

1 minute 0.030 mm

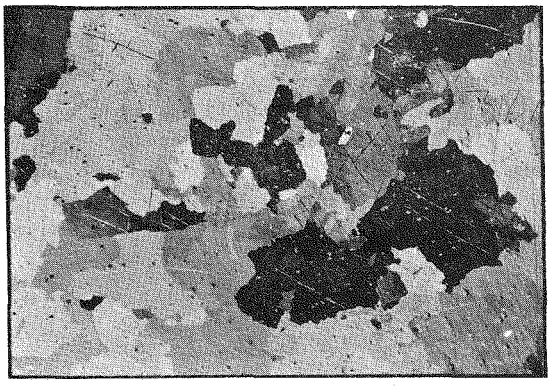


2 minutes 0.030-0.100 mm



3 minutes 0.030-0.200 mm

4 minutes 0.030-0.300 mm

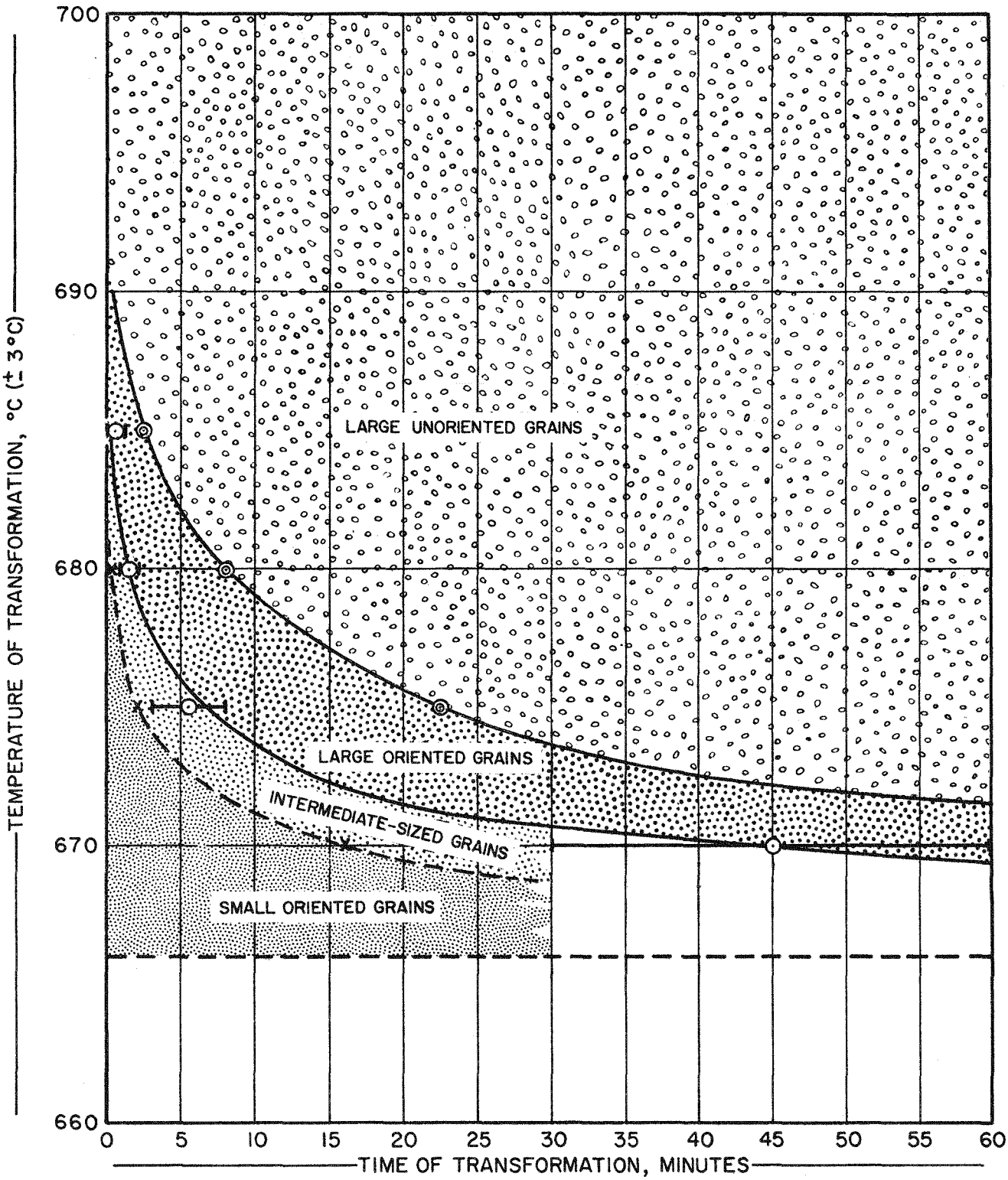


8 minutes 0.030-0.600 mm

16 minutes 0.030-0.600 mm

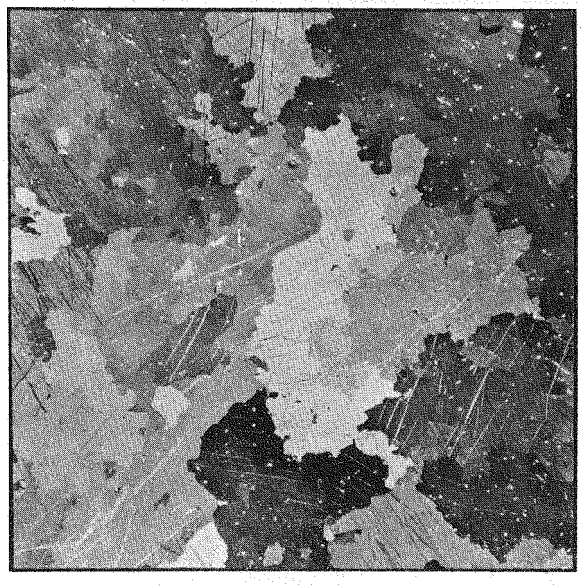
GRAIN STRUCTURES OF URANIUM PLATE
BETA TRANSFORMED FOR VARIOUS TIMES AT 675°C AND AIR COOLED
Mag. 40X

FIGURE 6

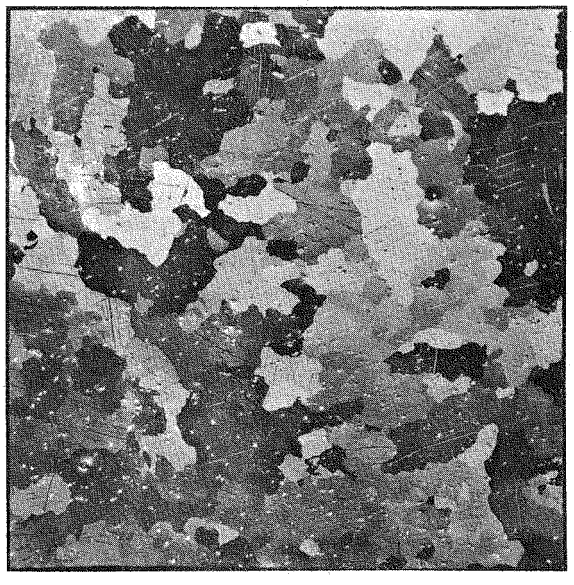



TIMES AND TEMPERATURES FOR TRANSFORMATION OF URANIUM PLATE

FIGURE 7



Initial Beta-Transformed Structure

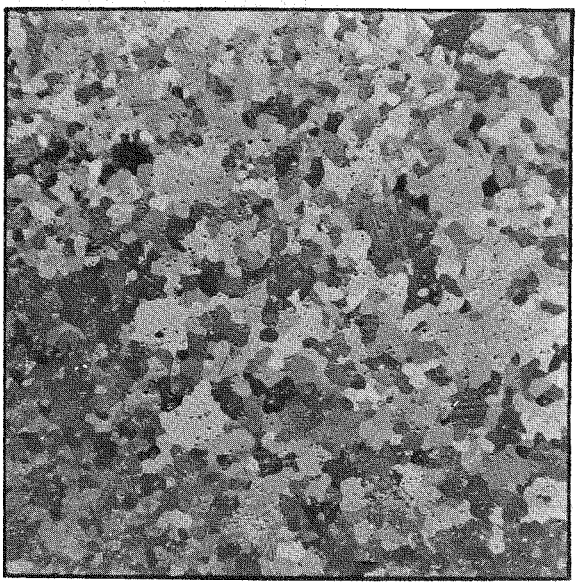


Partially Retransformed Structure

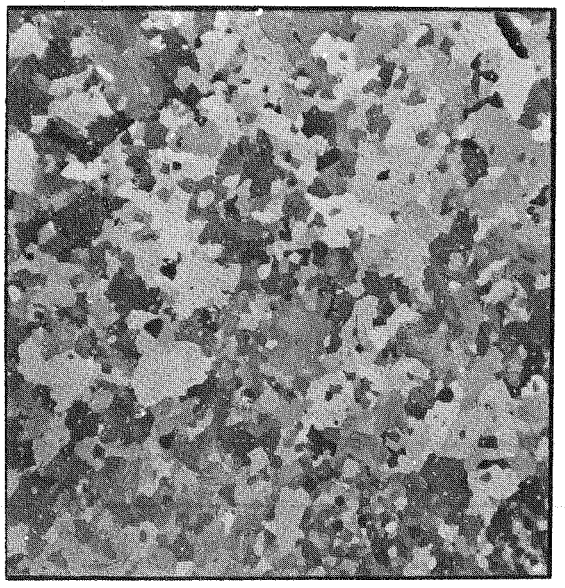
GRAIN STRUCTURE OF PARTIALLY RETRANSFORMED URANIUM PLATE

Beta-transformed metal was heated at 670°C for 8 minutes and air cooled. The grain size of the retransformed metal is slightly refined as compared to the original beta-transformed metal. Mag. 50X.

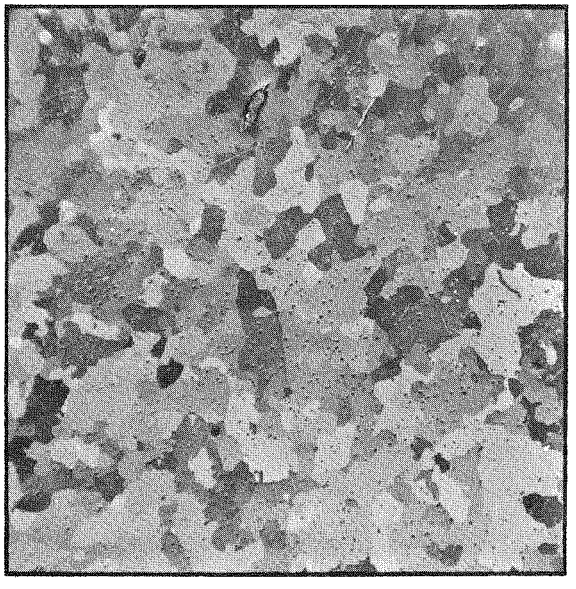
FIGURE 8



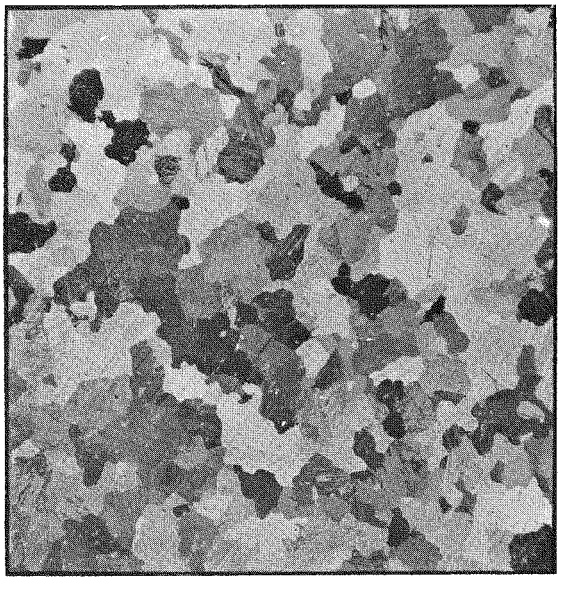
2 minutes



4 minutes in 2-minute intervals



6 minutes in 2-minute intervals

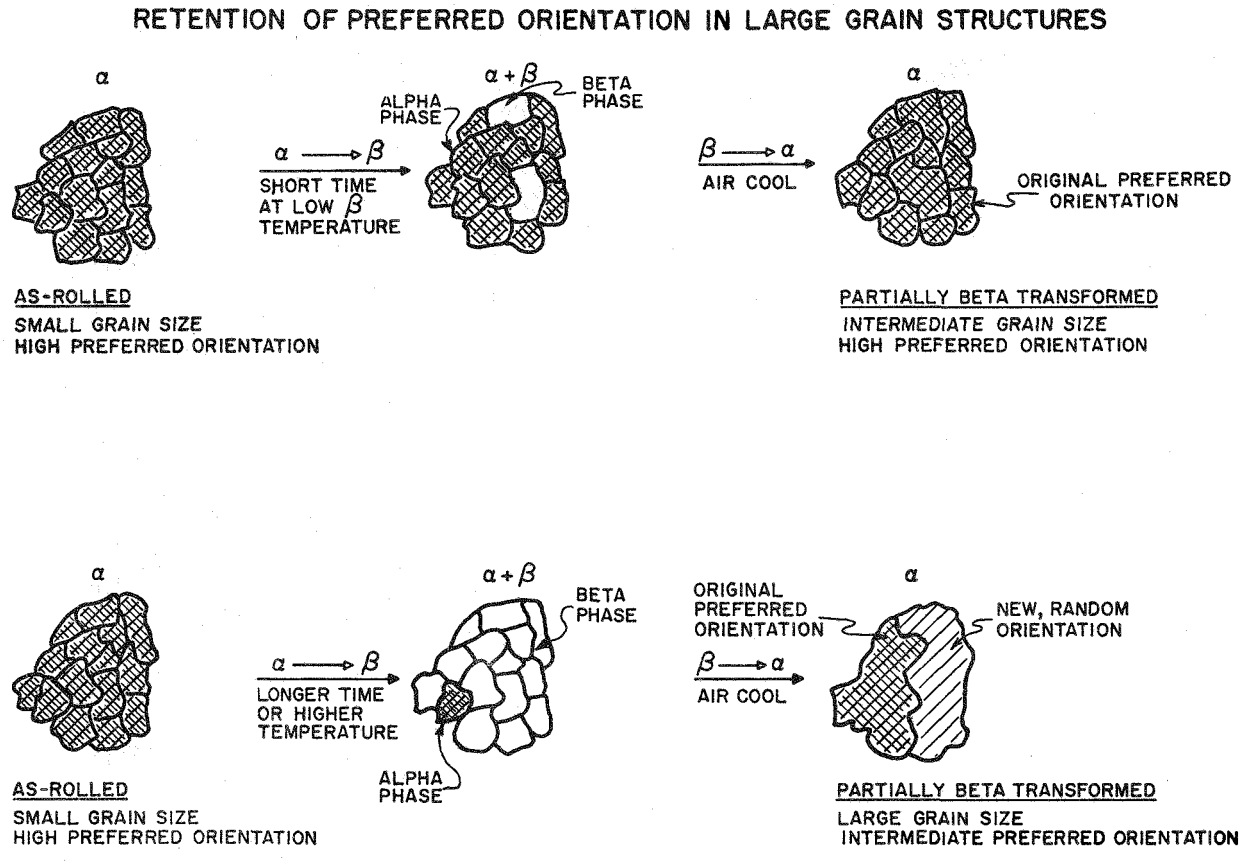


8 minutes in 2-minute intervals

GRAIN STRUCTURES OF URANIUM PLATE
BETA TRANSFORMED AT 670°C BY INTERRUPTED HEATINGS

Note the progressively increasing grain size. Mag. 50X.

FIGURE 9




## MECHANISM OF BETA TRANSFORMATION