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M E M O R A N D U M

March 10, 1965

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FROM: G. R. CASKEY, JR.
G. R. Caskey, Jr. /C.L.A.

MICROSTRUCTURAL CHANGES ACCOMPANYING ANNEALING OF COLD-WORKED URANIUM

INTRODUCTION

Recovery of the capacity for plastic deformation by annealing previously cold-worked uranium plays a key role in the mechanism proposed for the cavitational swelling observed in irradiated uranium.(1) Consequently, an investigation of recovery of yield strength was undertaken for unalloyed uranium and several selected alloys. During the course of this study, variations in the volume fraction of twins in the various specimens of unalloyed uranium suggested that twinning might be a mechanism of the recovery process. This question was studied by detailed microscopic examination of annealed specimens of both prestrained and unstrained uranium and by observation of structural changes during annealing of specimens in a hot-stage microscope. Results of four experiments in this study are described in this memorandum.

SUMMARY

The evidence supports the hypothesis that deformation twinning is a mode of recovery during annealing of cold-worked uranium at temperatures of 400-500°C. The principal observations were:

- In unstrained specimens, the volume fraction of twins remained essentially constant or decreased slightly at all annealing temperatures.

- In prestrained specimens the volume fraction of twins decreased slightly for annealing temperatures below 350°C, increased for temperatures of 400-500°C, and decreased again at higher annealing temperatures.

- These behavior patterns were essentially the same for tests on a series of separate specimens or a test in which one specimen was annealed at successively higher temperatures.
- New twins were observed by hot-stage microscopy during the annealing of a deformed specimen at temperatures of 250 and 360°C.
- New twins were observed to form during annealing at 400°C of a specimen previously strained approximately 1%.

The present work, which is believed to be the first recognition of the phenomenon of twinning as a mode of recovery, still needs an independent confirmation. In the specific case of uranium, such a behavior would imply that twinning is intimately associated with cavitational swelling and that other deformation modes such as slip are relatively more difficult than twinning at these temperatures (400-500°C). The effect of alloying on twinning needs to be investigated to determine if twinning and swelling susceptibility are correlated.

DISCUSSION

Description of Experiments

Tensile specimens of unalloyed uranium (dingot 993), composition listed in Table I, 1/8-inch diameter with a 1/2-inch gage length, were extended 0.8% at room temperature and unloaded. This prestrain gave a permanent deformation of about 0.5%. This is below the 0.8-1.2% deformation required for recrystallization. Duplicate specimens were annealed for 1.5 hours at temperatures ranging from 150 to 650°C along with blanks from the same stock that had not been prestrained. One specimen from each annealing temperature was pulled to fracture at room temperature to measure the recovery temperature; recovery was shown to occur at about 300°C.(2) The second specimen along with the blanks was examined by optical microscopy. A point count of the volume fraction of twins was made on a longitudinal section from each blank and annealed specimen. (3)

The volume fraction (f_v) of twins in the unstrained blanks remained essentially constant at about 7% for all annealing temperatures except 300°C where the twin-volume rose to 9-1/2% (Figure 1a). This difference is not statistically significant, however.

Figure 1a also shows the change observed in the volume fraction of twins in the prestrained tensile specimens. At temperatures between 375 and 500°C the volume fraction increased to a maximum of about 15%. At 550°C it dropped to 10% and at 650°C the volume fraction in the prestrained specimen was approximately 7%, the same as the blank.

Inspection of the unstrained and prestrained-and-annealed specimens revealed several alterations in the microstructure. The 0.8% prestrain caused an increase in the amount of twinning and probably a decrease in subgrain size (Figure 2). Because of the heterogeneity of the structures this latter feature is not certain, however. There was also a large increase in the area of internal boundary, but much of this increase must be attributed to twinning. Figure 3 shows that there were no obvious changes in grain structure in the prestrained-and-annealed specimens described in Figure 1a. The specimen annealed at 650°C did appear to have a slightly larger grain size and less substructure than the other annealed specimens.

In a second experiment, a single specimen was first annealed at 550°C for two hours. It was ground on one side, polished, and examined. The specimen was then prestrained and the volume fraction twin was measured again. Observations were made on the flat side that had been ground on the specimen prior to prestrain. The specimen was then heated 1.5 hours at 150°C along with a blank that had not been strained. A count was made of the twins after cooling to room temperature. The annealing and cooling operations were repeated, as indicated in Figure 1b, up to an annealing temperature of 650°C.

The twin volume in the blank showed a decrease from 7.6% to 4.1% during the tests, whereas the behavior of the prestrained sample was similar to the behavior of the other prestrained specimens described above: the twin volume in this sample also showed a "bump" in the curve at about 450°C.

The third experiment was the annealing of a prestrained sample in a hot-stage microscope to permit continuous observation of the structure during annealing. A sample was cut from a tensile specimen that had been extended 0.8%. The specimen was heated to 250°C, held for 90 minutes and cooled to room temperature; subsequently it was reheated to 360°C, held 90 minutes and finally cooled to room temperature. The quoted temperature is that of a thermocouple in contact with the back of the specimen.

Several new twins formed in the microstructure of this area of the surface during the annealing and upon cooling to room temperature, as shown in Figure 4. Oxidation of the surface is responsible for changes in contrast and brightness of the surface.

The practical temperature for use of the hot stage was limited by surface oxidation to about 400°C, except for very short times. One attempt to observe microstructural changes at higher temperatures was unsuccessful because of the oxidation of the specimen.

In the final experiment, a rod of unalloyed dingot uranium about 1/4-inch diameter was strained 1% and then annealed at 650°C to produce the structure shown in Figure 5a. This microstructure was relatively free of substructure but contained a few fine twin markings. After an additional strain of 1.2% (0.84% permanent set) at room temperature, new deformation twins were visible in the previously examined areas (compare Figures 5a and 5b). A stress of 51,000 psi was required. The Rockwell G hardness increased from 65 to 70 and the volume fraction twins increased from 0.066 to 0.208. Because the specimen was not reground before examination some surface relief was visible where the new twins intersected the surface. After the specimen was annealed for 3 hours at 400°C in an evacuated capsule, additional twins were visible (Figure 5c) that had not been observed in the as-strained specimen.

Interpretation of Results

Results of the experiments described above may be summarized as follows:

- During annealing at temperatures of 400-500°C, there is an apparent increase in the twin volume in prestrained specimens but not in unstrained blanks. This behavior is more pronounced for isochronal annealing of a series of specimens than for progressive annealing a single specimen at successively higher temperatures.
- New twins were observed in a prestrained specimen after annealing at 400°C.
- New twins were also seen upon examination of a specimen during annealing at 250 and 360°C in a hot-stage microscope.

The observed difference in volume fraction of twins (.071 vs .116) between the unstrained blanks and prestrained specimens (Figure 1a) is statistically significant. The observed difference in twin volume between the annealed and the prestrained samples after successive anneals up to a temperature of 450°C (Figure 1b) is also significant. Variations in sample variances and means in the series of observations on the as-machined and annealed specimen are not large enough to be significant. On the other hand, the observed variations in variance and mean for the prestrained samples are too large to have been reasonably due to chance causes and the increase in f_V for intermediate temperatures is statistically significant.

The observations indicate that some of the twins formed by the prestrain are removed by annealing at low temperatures since the twinned fraction decreases for annealing temperatures below 300°C. The larger twinned fractions observed at temperatures of 400-500°C would be due then to formation of new twins during annealing at the higher temperatures. There are two possible sources of stresses at elevated temperatures that could account for the formation of twins. One is the anisotropy of thermal expansion and the other is residual stresses. Examination of both of these sources in detail indicates that neither could explain the experimental observations.

One source of stress is the anisotropy of thermal expansion of the uranium. If this were the only source of stress, then, contrary to the experimental observations, there should be no difference in behavior between the as-machined and prestrained specimens, unless some other microstructural difference existed to concentrate the stress and make it more effective in the prestrained samples. One possibility is "structural debris" left behind by twins annealed out at lower temperatures, either during prior anneals or during heating to temperature. "Ghost" structures left after absorption of twins have been observed in some materials, but it is not known if restressing would generate a twin at the same location again.

A second source of stress is residual elastic stresses of a magnitude equal to or less than the elastic limit at room temperature. This elastic stress would be present only in the prestrained specimens and would vary from grain to grain within the specimen. At elevated temperature the stress required for twinning may be less than at room temperature and thus the residual elastic stresses could cause further twinning at elevated temperature. Generally, the stress for twinning is presumed to be insensitive to temperature variations, thus, arguing against this explanation; however, twinning in uranium has not been studied at temperatures between 350 and 600°C and the operative twin modes and twinning stresses are not known.

If deformation twinning is relatively easily induced at temperatures of 400-500°C, as suggested by these data, the variations in cavitational swelling that are observed among various uranium-base alloys could be related to the influence of alloying on the twinning process. Deformation twinning might function either as an alternative to crack formation and reduce cavitation, or as a means of inducing cracks because of the stress concentration at the end of a twin. In the first case twinning would be desirable, whereas in the second case it would be undesirable.

What little evidence there is associates twins with the fracture process and indicates that unalloyed uranium, which is highly susceptible to cavitation, twins readily during the recovery anneal. During mechanical cycling tests at 300°C, more twinning was observed in those alloys that swell the most during irradiation.[4] Additional data are needed to define the influence, if any, of alloying on deformation twinning at temperatures of 350-500°C and on the relation between twinning and fracture in uranium alloys.

Further experiments would also be required to reach an unequivocable conclusion as to whether deformation twinning may serve as a stress relief mechanism during annealing of cold-worked uranium.

REFERENCES

1. Angerman, C. L. and G. R. Caskey, Jr. "Swelling of Uranium by Mechanical Cavitation." To be published in Jnl. Nucl. Matls.

2. Caskey, G. R., Jr. "Effect of Silicon on Recovery of Tensile Flow Stress of Uranium." DPST-65-196, March 4, 1965.

3. Caskey, G. R., Jr. "Swelling of Mark VB Fuel." DPST-64-323. (SECRET) Description of point count technique given in Appendix.

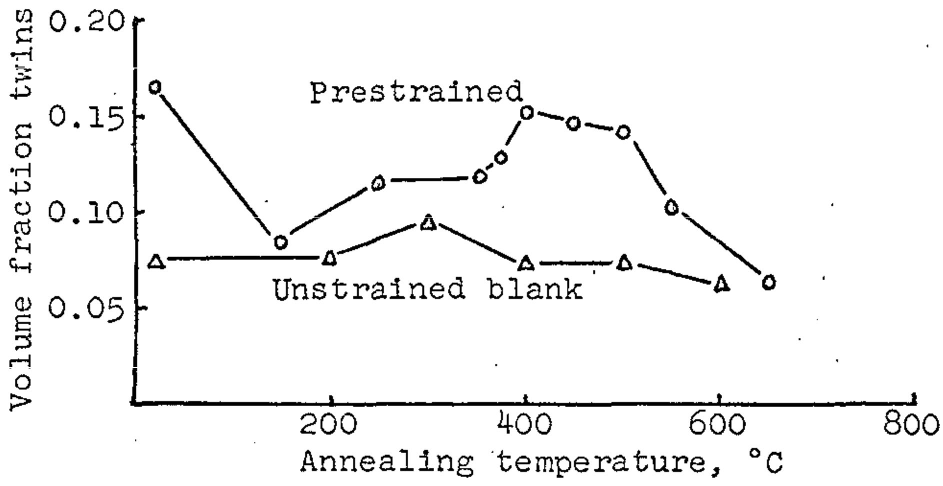
4. Angerman, C. L. and M. R. Louthan, Jr. "Mechanical Cavitation in Unirradiated Uranium." DPST-64-362 (Secret).

GRC:bdp

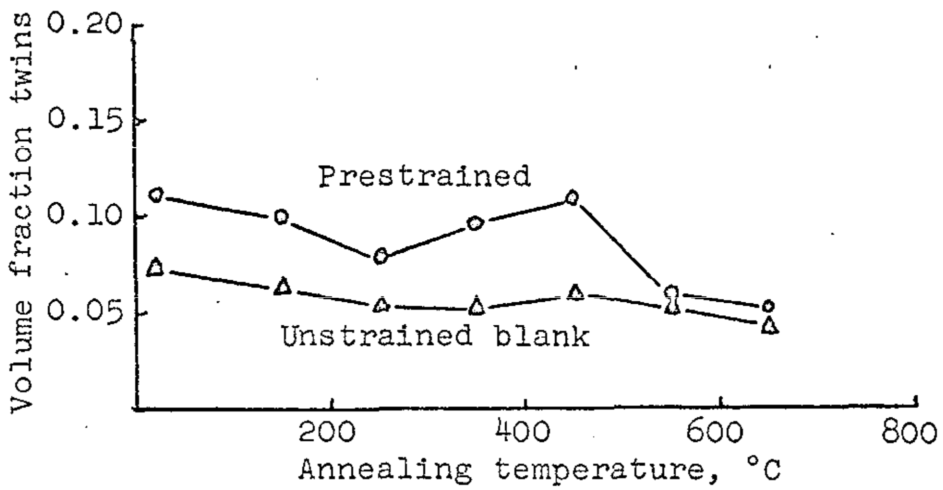
TABLE I

CHEMICAL COMPOSITION OF DINGOT 993

Element	Content, ppm
Carbon	28
Iron	57
Silicon	11
Aluminum	32
Chromium	11
Nickel	29

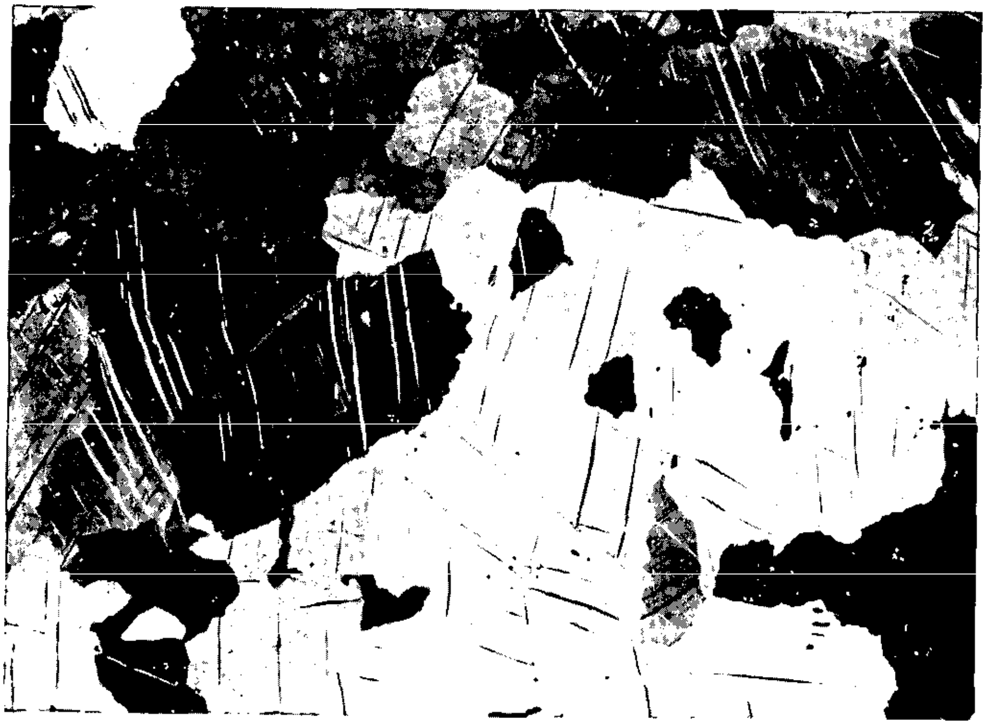


a. Separate specimens annealed at each temperature.



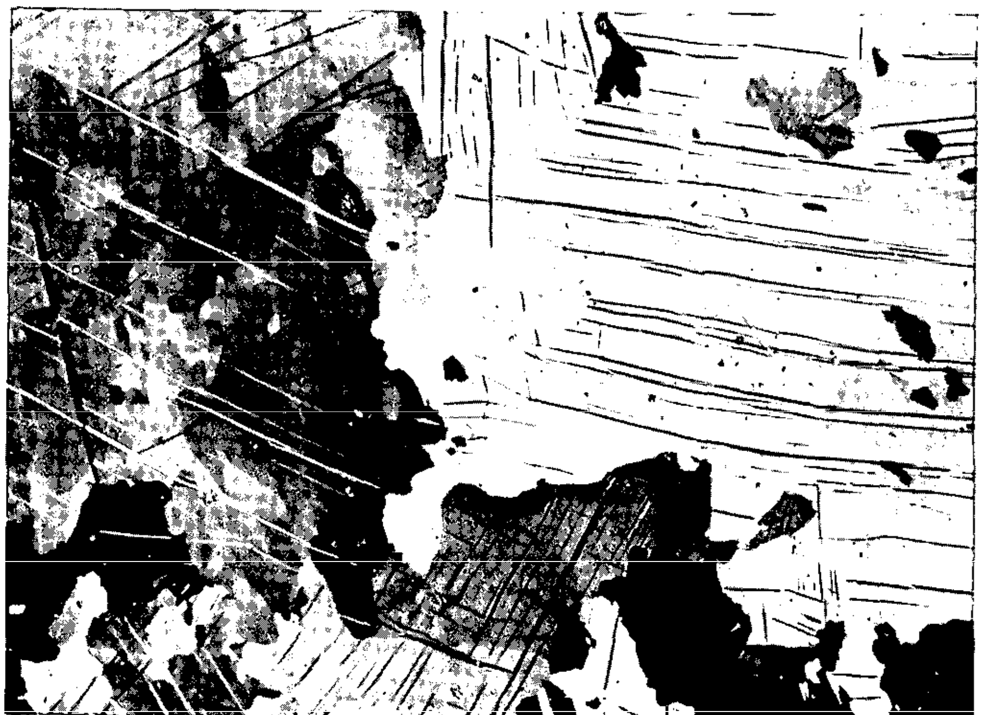
b. Two specimens annealed at successively higher temperatures. Twin count made after each anneal.

FIGURE 1. VARIATIONS IN VOLUME FRACTION TWINS DURING ALPHA ANNEALING



Neg. 55123

a. Unstrained blank. Volume fraction of twins (f_V) = 0.07.



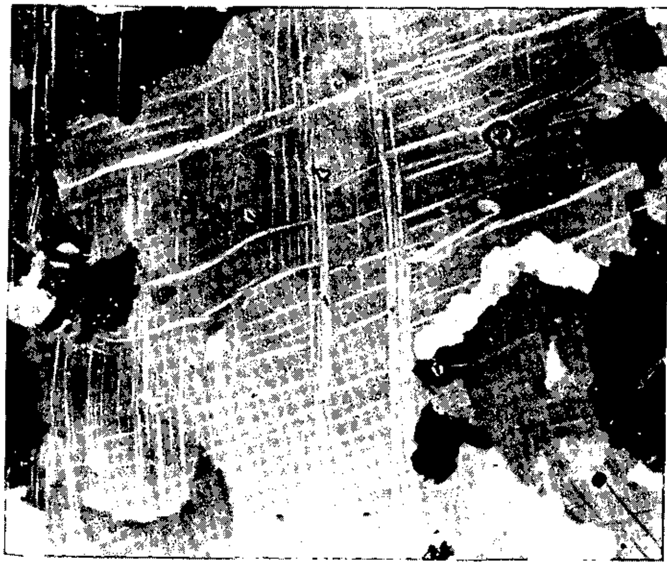
Neg. 58081

b. Specimen prestrained 0.8% in tension, f_V = 0.165.

FIGURE 2. EFFECT OF TENSILE PRESTRAIN AT ROOM TEMPERATURE ON THE MICROSTRUCTURE OF UNALLOYED URANIUM (150X)

Neg. 58098

a. Annealed at 150°C, f_V = 0.86.



Neg. 58091

b. Annealed at 250°C, f_V = 0.116.

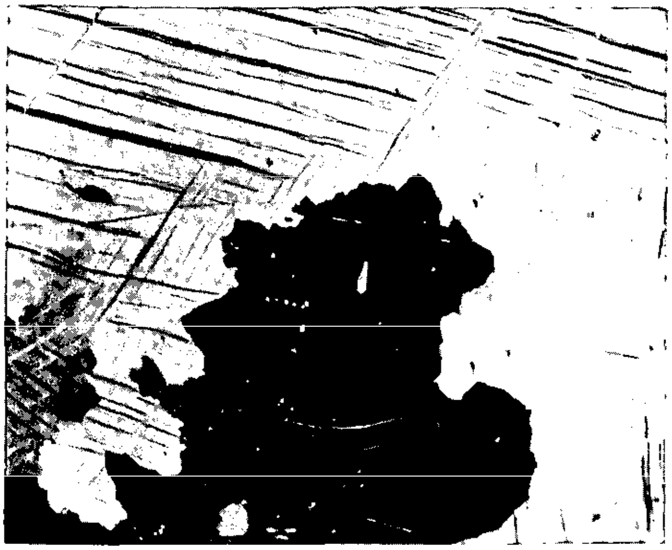
Neg. 58090

c. Annealed at 350°C, f_V = 0.118.

Neg. 58097

d. Annealed at 375°C, f_V = 0.128.

FIGURE 3. EFFECT OF ANNEALING ON VOLUME FRACTION OF TWINS IN MICROSTRUCTURE OF SPECIMENS STRAINED 0.8% IN TENSION AT ROOM TEMPERATURE (150X)

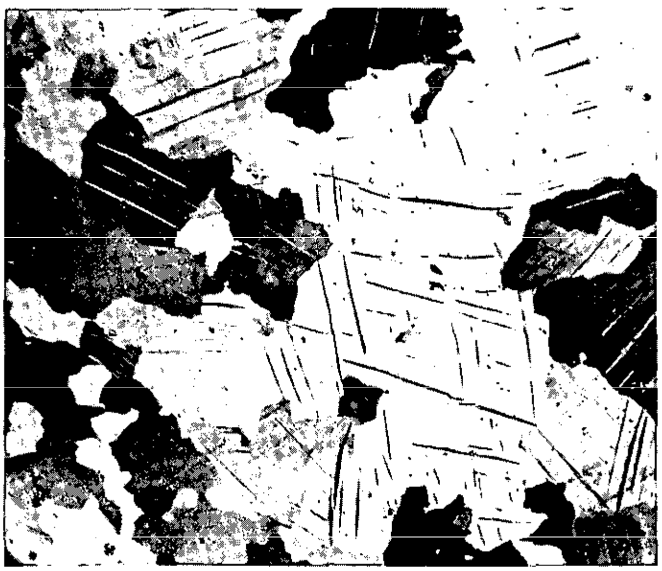


Neg. 58088

e. Annealed at 400°C, $f_v = 0.154$.

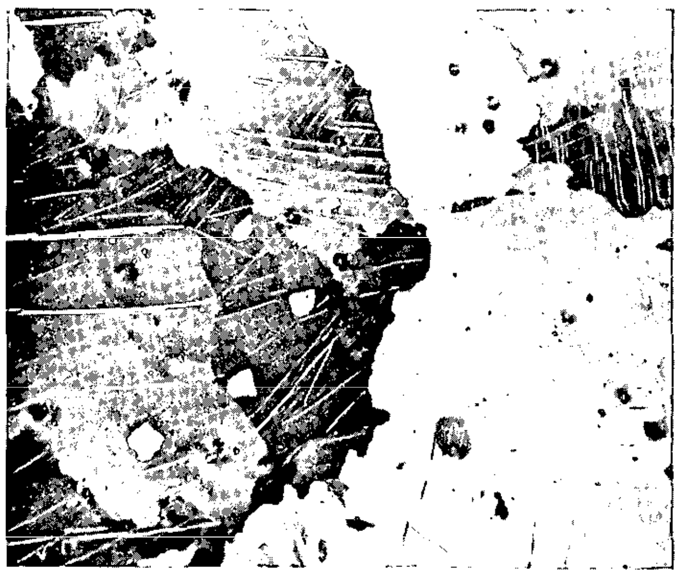
Neg. 58085

f. Annealed at 450°C, $f_v = 0.146$.



Neg. 58102

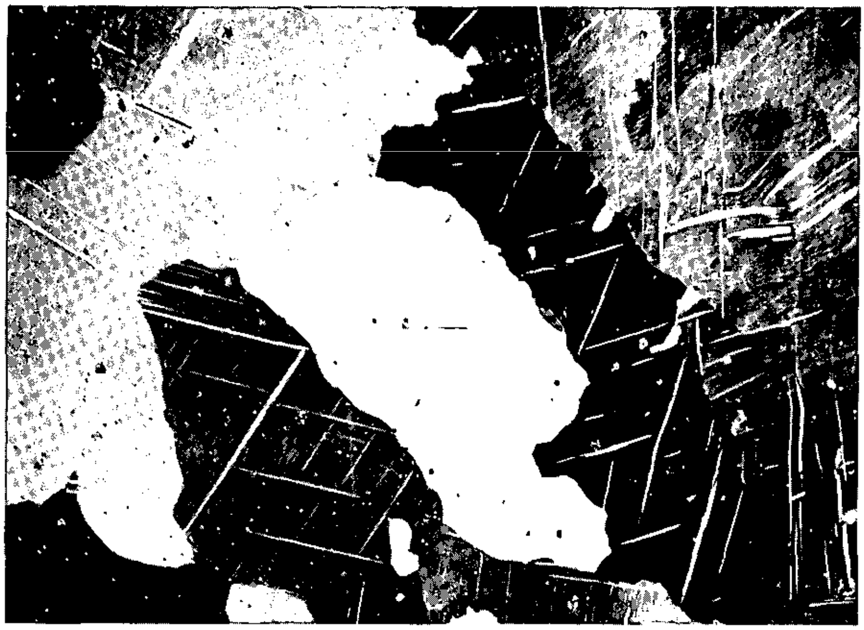
g. Annealed at 500°C, $f_v = 0.144$



Neg. 58104

h. Annealed at 550°C, $f_v = 0.102$.

FIGURE 3. (Continued)



Neg. 58129

i. Annealed at 650°C, $f_v = 0.064$.

FIGURE 3. (Continued)

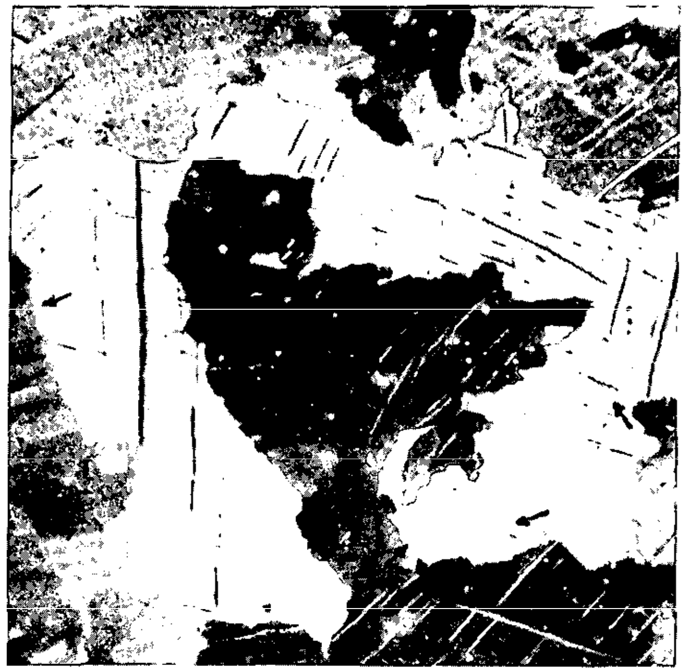
a. Initial microstructure of specimen deformed 0.8% in tension.

b. Microstructure at 250°C following 90-minute anneal at 250°C.

c. Microstructure at 20°C after cooling to 20°C following anneal at 250°C.

FIGURE 4. FORMATION OF NEW TWINS DURING ANNEALING IN HOT-STAGE MICROSCOPE
(All photomicrographs made in hot stage at 150X)

d. Microstructure at 360°C following 90-minute anneal at 360°C subsequent to anneal at 250°C.



e. Microstructure at 20°C following both annealing treatments.

The following new twins were observed.

(Figure b) During anneal at 250°C twin formed in subgrain (A) to connect with preexisting twins. New, long twin (B) formed crossing several subgrains.

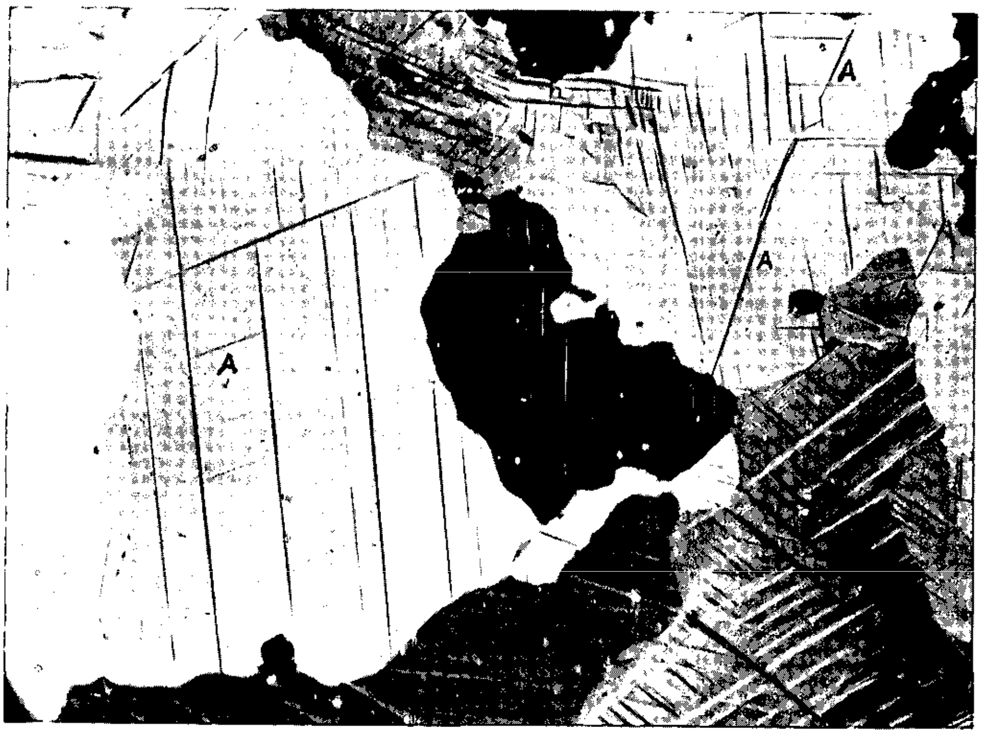
(Figure c and d) No additional changes.

(Figure e) Several new twins (arrows) formed. These twins appear white in the photomicrograph.

FIGURE 4 (Continued)

Neg. 58323

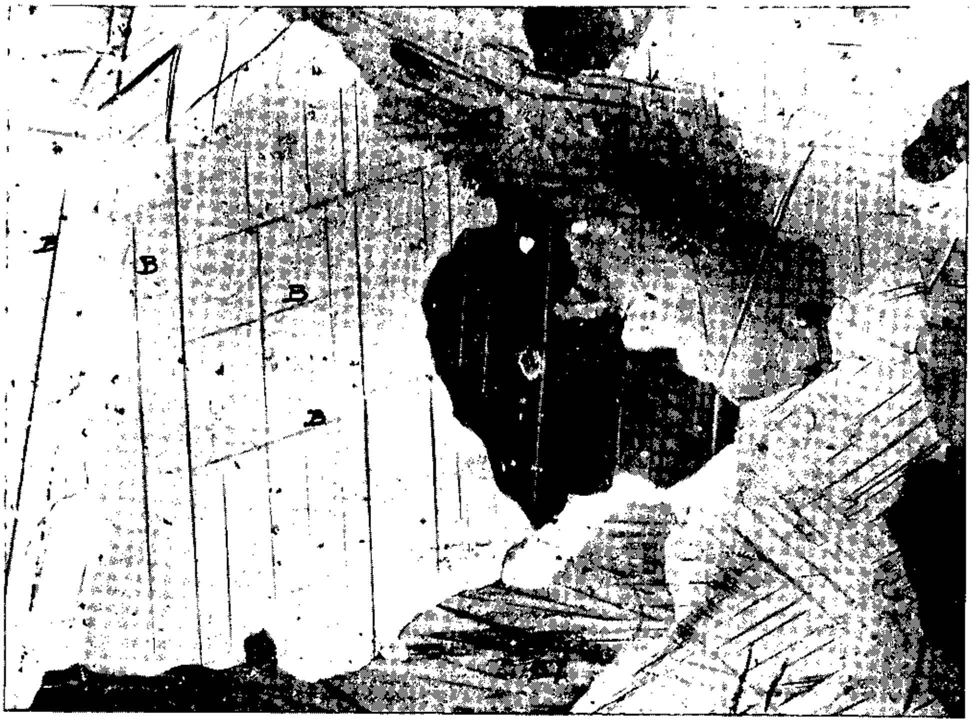
a. Annealed structure - annealed for 2 hours at 650°C after 1% prestrain.



Neg. 58328

b. Same area as Figure a, after an additional 1.2% strain. Note new deformation twins (A).

FIGURE 5. MICROSTRUCTURAL CHANGES PRODUCED BY DEFORMATION AND ANNEALING (150X)



Neg. 59476

c. Same area as Figure b after 3-hour anneal at 400°C. Note new twins (B).

FIGURE 5. (Continued)