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THE BETA-ALPHA AND ALPHA-BETA TRANSFORMATIONS
IN PLUTONIUM UNDER APPLIED STRESSES

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THE BETA-ALPHA AND ALPHA-BETA TRANSFORMATIONS
IN PLUTONIUM UNDER APPLIED STRESSES

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THE BETA-ALPHA AND ALPHA-BETA TRANSFORMATIONS IN PLUTONIUM UNDER APPLIED STRESSES

INTRODUCTION

Research was performed on the effect of concurrent applied stress on the $\beta \rightarrow \alpha$ and $\alpha \rightarrow \beta$ transformations in plutonium. The primary objective of this activity was to obtain indirect information on the mechanism of these transformations and to compare the transformations in plutonium to transformations in more conventional metals and alloys. A secondary objective was to study the effect of applied stress on the phase transformations of a metal that is unique because: (1) the $\beta \rightarrow \alpha$ and $\alpha \rightarrow \beta$ transformations are between monoclinic crystal structures that differ in volume by 9%; (2) the creep and ductility of the beta phase at a given stress and temperature can be varied over a very large range depending on whether it was formed from alpha or from gamma; and (3) the $\alpha \rightarrow \beta$ transformation takes place at temperatures where diffusion processes should be operative, whereas the $\beta \rightarrow \alpha$ transformation takes place at temperatures where diffusion processes are not likely operative.

A number of investigators have found a marked influence of applied stress on the kinetics of phase transformations. (1-17) The most extensive work has been done with iron alloys, although some information is available on other systems such as tin-antimony, (13) gold-cadmium (9) copper-zinc, (18) copper-aluminum, (19) indium-thallium, (7) and lithium-magnesium. (20) An applied stress may affect the kinetics of both diffusional and martensitic transformations, and it may introduce extra creep deformation in a metal during transformations.

The austenite-pearlite and austenite-bainite transformations in steel can be accelerated by an applied stress. (14, 15) The austenite-martensite transformation can be induced isothermally above the M_s temperature by elastic and residual plastic strains. In iron-nickel-carbon alloys (16-17) and indium-thallium alloys (7) the M_s temperature

temperature is raised by either uniaxial compressive or uniaxial tensile stresses, but it is lowered by hydrostatic pressures. The M_s temperature of the $\beta' \rightarrow \beta$ transformation of Au-47.5 at.% Cd alloy was raised during cooling under applied stress but was lowered during heating under similar conditions;⁽⁶⁾ that is, the reversible martensite transformation was always favored by applied uniaxial stress.

The most extensive work on the deformation of metals with concurrent phase transformation has been performed by Clinard and Sherby,^(9, 21) De Jong and Rathenau,^(22, 23) and Porter and Rosenthal⁽¹⁰⁾ on iron and iron alloys.

The three strain components proposed by Sherby and Clinard to make up the observed deformation during phase transformation are: (1) phase transformation strain [excess creep strain]; (2) normal creep strain; and (3) strain due to volumetric change. Clinard's and Sherby's conclusions concerning deformation during the $\gamma \rightarrow \alpha$ and $\alpha \rightarrow \gamma$ phase transformations in iron are:⁽⁹⁾

- "Transformation strain is responsible for a significant portion of the total strain, both on heating and on cooling, up to the highest stress studied (1085 psi)."
- "Transformation strain obtained on heating is significantly greater than that obtained on cooling."
- "Transformation strain is a nonlinear increasing function of applied stress."
- "The amount of transformation strain is a function of transformation rate."

They proposed that excess point defects formed during transformation contribute to dislocation climb, by which mechanism the deformation rate is accelerated. Vacancies form during the $\alpha \rightarrow \gamma$ transformation, a contraction, and interstitials form during the $\gamma \rightarrow \alpha$ transformation. They called the resulting excess creep strain, "Transformation strain."

De Jong and Rathenau⁽²²⁾ observed that in iron the $\alpha \rightarrow \gamma$ transformation strain on heating is essentially the same as the $\gamma \rightarrow \alpha$ strain on cooling. They also observed that excess creep strain is independent of

transformation rate and is a linear function of applied stress. These observations disagree with the more recent results of Clinard and Sherby⁽⁹⁾.

De Jong and Rathenau⁽²²⁾ concluded that the excess creep strain associated with phase transformation under applied stress is really not a kind of creep. They proposed that two processes might produce excess creep strain under applied stress:⁽²³⁾

- "Owing to the difference in density of the parent and the product phases, internal stresses are thought to occur during transformation. The material yields to the superimposed external stress at a lower value because of the preexisting internal stresses. Excess elongations are to be expected at even small external stresses if these stresses remove the equivalence of the slip systems which are activated by the change in volume."
- "During transformation, especially in the martensitic case (steel), out of the possible modes of transformation those are favored which allow the largest elongation in the direction of the external stress."

On the basis of previous work of other investigators on stress and concurrent phase transformations, similar studies on plutonium offered certain appeal. This report presents such information on plutonium transformed isothermally and also transformed by heating and cooling at fixed rates. The plutonium used was 99.95% pure. It contained 0.005% iron, 0.01% carbon, 0.005% uranium, 0.01% americium, and other impurities totaling less than 0.02%.

The total strain accompanying the various transformations in pure plutonium is most conveniently analyzed by considering three contributions to the total strain:

- Normal creep strain. This has been discussed previously.⁽²⁶⁾ Normal creep can occur significantly during transformation under stress.
- Excess creep strain. This is the creep at small applied stresses where normal creep is not expected. Excess creep has been postulated to be caused by an excess concentration of point defects.
- Transformation strain. Transformation strain is defined as the macroscopic shape change resulting from the summation of the shape change accompanying the formation of each grain of the product phase. In a perfectly random transformation, this strain would be isotropic and equal to one-third the fractional volume change accompanying transformation. If the transformation occurs anisotropically, the resulting transformation strain will be similarly anisotropic.

SUMMARY AND CONCLUSIONS

Application of uniaxial compressive and tensile stresses markedly affects the total strain during the $\beta \rightarrow \alpha$ and $\alpha \rightarrow \beta$ transformations in plutonium. A number of conclusions can be drawn from the results:

- The total strain during $\beta \rightarrow \alpha$ transformation under compression is almost completely independent of the transformation rate.
- Applied compressive stresses above 9000 psi cause essentially all of the $\beta \rightarrow \alpha$ transformation strain to occur as a contraction parallel to the applied stress, i. e. $\Delta L/L_0 = \Delta V/V_0$.
- Applied tensile stresses of about 4000 psi cause all of the strain during the $\beta \rightarrow \alpha$ transformation to occur as a contraction perpendicular to the applied stress. No contraction is observed parallel to the applied tensile stress.
- Applied stresses during $\beta \rightarrow \alpha$ transformation produce a microstructural shape texture. The alpha grains are elongated in a direction parallel to the applied compressive stresses and perpendicular to the applied tensile stresses. The elongated grains are aligned in disks which are perpendicular to the direction of the compressive stress and parallel to the direction of the applied tensile stresses.
- Applied stresses during $\beta \rightarrow \alpha$ transformation produce a crystallographic texture. The (010) planes are aligned parallel to the applied tensile stresses and perpendicular to the applied compressive stresses.
- The anisotropic strains accompanying the $\alpha \rightarrow \beta$ transformation under uniaxial stress indicate that a texture also develops.
- The $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations in textured plutonium result in anisotropic transformation strain in the absence of applied stress.
- Both applied uniaxial tensile stresses and uniaxial compressive stresses raise the temperature for the beginning of the $\beta \rightarrow \alpha$ transformation at a constant cooling rate and for stresses up to at least 7000 psi, the maximum stress studied.
- Applied tensile stresses above 2500 psi cause all of the $\alpha \rightarrow \beta$ transformation strain to occur as an expansion parallel to the applied stress, i. e., $\Delta L/L_0 > \Delta V/V_0$.
- Some phase transformation excess creep occurs during the transformations in specimens subjected to an external stress.

- Phase transformation excess creep is greater under tensile than under compressive stresses. Little or no $\beta \rightarrow \alpha$ transformation excess creep occurs under compressive stresses.
- The $\beta \rightarrow \alpha$ transformation fulfills many of the requirements for a martensitic type transformation.

TOTAL STRAIN ACCOMPANYING THE $\beta \rightarrow \alpha$ TRANSFORMATION UNDER APPLIED COMPRESSIVE STRESSES

The strain during the $\beta \rightarrow \alpha$ transformation was evaluated as a function of applied compressive stress from 200 to 20,000 psi. The parent beta phase was formed by transformation from either the alpha or the gamma phase. The beta phase was significantly strengthened by first transforming the metal to gamma and then transforming back to beta.^(24, 25) This procedure for strengthening the beta phase was advantageous in the investigation because the much lower creep rate of beta formed from gamma permitted tests at stresses up to 20,000 psi. When beta was formed from alpha, tests could be performed at stresses no higher than 5000 psi without appreciable creep of the beta phase. The experimental procedure and steady-state creep data are reported elsewhere.⁽²⁶⁾

Specimens were heated to 260 C and held at temperature for 30 min to ensure complete transformation to gamma. To transform the gamma phase to the beta phase, they were then cooled to 120 C. The $\gamma \rightarrow \beta$ transformation was presumed complete after holding the specimens 1 hr at 120 C. Specimens in which the beta was formed directly from alpha were heated to 180 C and maintained at that temperature for 1 hr to ensure complete $\alpha \rightarrow \beta$ transformation. All specimens were cooled to 110 to 115 C for start of the tests.

Representative creep curves of the single phase alpha and beta phases as well as curves showing the strain associated with the $\beta \rightarrow \alpha$ transformation with no applied stress are shown in Figure 1.

The total, normal creep strain during continuous heating at a given stress was obtained from steady-state creep data using graphical integration according to the relationship:

$$\epsilon = \int_0^t \dot{\epsilon} dt$$

FIGURE 1

Representative Data of the Strain Accompanying the Plutonium $\beta \rightarrow \alpha$ Transformation and Single Phase Creep of Alpha and Beta Plutonium Under Applied Compressive Stresses at 110 C.

The normal creep strain was small compared to the volumetric change and generally could be neglected. Because the heating and cooling rates were generally very slow, graphical integration was required over only a few degrees. In most cases the transformation proceeded under isothermal conditions.

The total strain during transformation was investigated as a function of the transformation rate. The total strain, $\Delta L/L_0$, was always measured in the direction of the applied stress. Representative total strains of the $\beta \rightarrow \alpha$ transformation under an applied stress of 5000 psi for three cooling rates are shown in Figure 2. These curves show that the total strain during transformation is only slightly dependent on the $\beta \rightarrow \alpha$ transformation rate.

This was in contrast to the behavior of iron as reported by Clinard and Sherby.⁽⁹⁾ The strain during transformation was correlated with the approximated maximum transformation rate, because this was believed to be the best parameter to characterize the transformation rate. The

FIGURE 2

Total Strain Accompanying the Plutonium $\beta \rightarrow \alpha$ Transformation with 5000 psi Compressive Stress.

The increase of the maximum transformation rate from 0.15%/min to 50%/min by increasing the cooling rates resulted in only a 20% decrease in the total strain associated with the $\beta \rightarrow \alpha$ transformation (Figure 3). The transformation temperature was raised approximately 15 C when the maximum transformation rate was lowered from 50%/min to 0.15%/min. Note that the unit length change accompanying transformation was nearly the same regardless of whether the beta was formed from alpha or gamma.

The total strain during transformation increased rapidly with increasing applied stress and reached a constant value at about 9000 psi, Figure 4. This constant value of 0.09 was equal to $\Delta V/V_0$ and was approximately three times the linear change, 0.03, expected for the strain when transformation occurs with no applied stress. Applied stresses appear to cause all of the $\beta \rightarrow \alpha$ transformation to occur by contraction in the direction of the applied stress. If this were so, then the total strain accompanying transformation consisted only of the anisotropic volume change and a small amount of normal creep.

FIGURE 3

Effect of Transformation Rate on the $\beta \rightarrow \alpha$ Transformation Strain.

FIGURE 4

Effect of Applied Compressive Stress on the Total Strain Accompanying the Plutonium $\beta \rightarrow \alpha$ Transformation at a Cooling Rate of 0.5 - 1.0 C°/hr

TOTAL STRAIN ACCOMPANYING THE $\beta \rightarrow \alpha$ TRANSFORMATION UNDER APPLIED TENSILE STRESSES

The total strain during the $\beta \rightarrow \alpha$ transformation in plutonium was also investigated as a function of applied tensile stresses at different transformation rates. A tensile stress tends to oppose the contraction of the transformation in the direction of the applied stress. Representative total strains at stresses from 250 psi to 7000 psi at a cooling rate of 2 C°/hr are shown in Figure 5. These curves show that the effect of applied tensile stresses was more pronounced and somewhat different from the effect of applied compressive stresses on the $\beta \rightarrow \alpha$ transformation. The total strain accompanying the transformation with very low stresses was negative, but the contraction in the specimens was less than the strain with no applied stress.

At 1500 psi, the specimens elongated during the first stages of transformation. The positive rate of strain decreased as the transformation proceeded, became zero, and then became negative because of the contraction associated with the volumetric change. The effect was similar at 2900 psi, with the elongation being greater and the contraction less than

FIGURE 5

The Total Strain Accompanying the $\beta \rightarrow \alpha$ Transformation
Under Applied Tensile Stresses at a Cooling Rate of 2 C°/min

at 1500 psi. Some net elongation was observed in the direction of the applied tensile stress in specimens transformed with 7000 psi. It is reasonable to assume that the linear transformation strain is limited to values between zero and $\Delta V/V_0$, i.e. 0 to 0.09. Therefore, the elongation observed in specimens transformed in tension must be attributed to excess creep strain.

MICROSTRUCTURAL AND CRYSTALLOGRAPHIC RELATIONSHIPS

At high alpha temperatures the transformation of beta to alpha initially proceeds by the formation of thin disk shaped volumes, which consist of many individual columnar grains of the alpha phase. The long axes of the grains are all perpendicular to the plane of the disk. In a polished and etched cross-section the disks appear as bands, which are randomly oriented in the absence of applied stresses (Figure 6). When a compressive stress is applied to the metal, the disks form at right angles to the compressive stress, while grains within the disks are parallel to the applied stress (Figures 7-9). The alignment of the alpha disks is

FIGURE 6

Microstructure of a Plutonium Specimen That Had Been Beta Heat Treated 20 hr at 185 C, Isothermally Transformed to Yield 20% of the Alpha Phase at 79 C, and Then Completely Transformed by Quenching to -75 C. (This microstructure clearly shows the bands of columnar grains which form during transformation at 79 C. The smaller grains are the alpha which formed during the quench to -75 C.)

Neg. 8828

75X

250X

250X

(Upper: The applied stress was perpendicular to the polished surface.
Lower: The applied stress was parallel to the longitudinal axes of the grains.)

FIGURE 7

Microstructure of a Specimen Completely Transformed
from the Beta Phase to the Alpha Phase
Under a Compressive Stress of 15,000 psi

Neg. 8309, 8307

FIGURE 8

400X

Microstructure of a Specimen Completely Transformed from the Beta Phase to the Alpha Phase Under a Compressive Stress of 15,000 psi. (The longitudinal axis of the grains was in the direction of the applied stress.)

(a) Neg. N-107 100X

(b) Neg. N-96 100X

(c) Neg. N-103 100X

(d) Neg. N-98 100X

FIGURE 9

Photomicrographs Showing the Texturing that Occurs During the $\beta \rightarrow \alpha$ Transformation Under an Applied Compressive Stress
(The applied stress was from top to bottom.)

	(a)	(b)	(c)	(d)
Applied Stress (psi)	2000	7200	5000	5000
Cooling Rate ($^{\circ}\text{C}/\text{min}$)	0.005	0.005	0.005	0.5-1
Maximum Transformation Rate ($\%/ \text{min}$)	0.5	0.5	0.5	10

particularly evident from the appearance of the bands in specimens only partially transformed under 15,000 psi uniaxial compression (Figures 10 and 11.)

Neg. 8348

100X

FIGURE 10

Microstructure Showing the Bands in a Plutonium Specimen Partially Transformed Under a Compressive Stress of 15,000 psi at 70 to 80 C and Quenched to -75 C. (The columnar grains formed under the applied stress whereas the small grains formed during the quench. The band of columnar grains was perpendicular to the direction of the applied stress.)

Neg. 8448

150X

FIGURE 11

Microstructure of a Specimen 80 to 90% Transformed Under a Compressive Stress of 15,000 psi. (Note that the bands are parallel.)

During transformation the (010) alpha planes form with their alignment predominantly in a direction perpendicular to the applied compressive stress. X-ray diffraction patterns of the transverse and longitudinal sections of a specimen transformed with an applied compressive stress of 15,000 psi are compared to the random structure in Figures 12 - 14. Although the (020) and (211) reflections are superimposed, the presence or absence of the (010) planes parallel to the specimen surface can be determined by the relative intensities of the (040) reflections. The (040) reflection was very pronounced in the diffraction pattern from the transverse section, but it was not detected in the diffraction pattern obtained from the longitudinal section. Also reflections (201), (004), (20 $\bar{3}$), and others, where k of the (hkl) value is zero, were very weak in the section that was perpendicular to the applied stress. Those reflections belonging to the (010) zone axis were strong in the section parallel to the applied stress. This verifies that the (010) plane lies predominantly perpendicular to the direction of applied compressive stress.

FIGURE 12

X-ray Diffraction Pattern of an As-Cast, Random Structure

FIGURE 13

X-ray Diffraction Pattern of Plutonium Transformed from the Beta Phase to the Alpha Phase at 15,000 psi (The X-ray pattern was of a surface parallel to the direction of the applied compressive stress.)

FIGURE 14

X-ray Diffraction Pattern of Plutonium Transformed from the Beta Phase to the Alpha Phase at 15,000 psi (The X-ray pattern was of a surface perpendicular to the direction of the applied compressive stress.)

The formation of grains in bands that are at right angles to the applied stress instead of randomly oriented is another indication that the $\beta \rightarrow \alpha$ transformation is a shear type and not a nucleation and growth type transformation.

If a tensile stress is applied to the metal, the bands form in the direction of the applied stress, while the grains within the bands are parallel with the applied stress (Figures 15 and 16.) The (010) plane

Neg. 4384

100X

FIGURE 15

Microstructure of a Specimen Transformed from the Beta Phase to the Alpha Phase Under a Uniaxial Tensile Stress of 7000 psi. (The surface was parallel to the direction of the applied stress.)

Direction of Applied Stress

Neg. 357

100X

FIGURE 16

Microstructure of a Specimen Transformed from the Beta Phase to the Alpha Phase Under a Uniaxial Tensile Stress of 7000 psi. (The section was perpendicular to the direction of the applied stress.)

becomes aligned parallel with the direction of the applied stress. This is revealed by the very weak (040) reflection in a section perpendicular to the applied stress.

When plutonium was transformed under compression, the dimensions perpendicular to the plane of the disk depended only upon impingement, whereas, the disk extended to the boundaries of the specimen in the direction parallel to the plane of the disk. When the plutonium transformed under no applied stress or under tension, all dimensions of the disk were restricted by either impingement or the boundaries of the specimen.

ANISOTROPY OF THE $\beta \rightarrow \alpha$ AND $\alpha \rightarrow \beta$ TRANSFORMATIONS

A highly textured cylindrical rod specimen, having the $[010]$ direction parallel with the rod axis, was cycled once through the $\alpha \rightarrow \beta$ and back through the $\beta \rightarrow \alpha$ transformation. The specimen was heated to 115 C where the diameter and the length were measured in the alpha phase. The dimensions of a rod in the alpha phase can be readily compared to the corresponding dimensions in the beta phase at 115 C since both beta and alpha can exist at this temperature for long times. The specimen was heated to 160 C, held for 1 hr to transform the metal completely to beta, and then cooled to 115 C for measuring the length and diameter of the rod in the beta phase. Transformation of the beta phase to the alpha phase was accomplished by cooling to room temperature at 50C°/hr. The rod was then reheated to 115 C, and the dimensions of the alpha phase rod were measured. The dimensions of the rod in the alpha phase before cycling, in the beta phase, and in the alpha phase after cycling could therefore be compared.

The length increased 7.9% and the diameter increased 0.8% during the $\alpha \rightarrow \beta$ transformation. This indicated that nearly all of the volume change during transformation occurred by expansion in a direction parallel to the length of the specimen. The length and the diameter decreased 1.2 and 4.3%, respectively, during the $\beta \rightarrow \alpha$ transformation. This indicated that most of the volume change during the $\beta \rightarrow \alpha$ transformation occurred by contraction in radial directions. The sample after transformation cycling had increased in diameter by 3.4%. From this it can be concluded that both the $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformation occurred anisotropically.

The anisotropic nature of the $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations of the textured rod indicated that a texture should still be present in the alpha phase after an $\alpha \rightarrow \beta \rightarrow \alpha$ cycle with no applied stress. A texture after cycling was confirmed by X-ray diffraction and by metallography, but it was different from the texture that had been observed before cycling. After cycling, the disks were aligned essentially parallel to the direction of the original applied compressive stress (Figure 17), rather than

Neg. 394

100X

FIGURE 17

Plutonium That Was Textured by $\beta \rightarrow \alpha$ Transformation Under 20,000 psi Compressive Stress and Then Cycled into Beta by Heating and into Alpha by Cooling. (Polarized light illumination.)

Neg. 276

100X

(Upper: Surface that was perpendicular to the direction of the applied stress. Lower: Surface that was parallel to the direction of the applied stress.)

Direction of Applied Stress

A double-headed arrow pointing diagonally upwards and to the left, indicating the direction of applied stress.

90 degrees to the applied stress as observed before cycling (Figure 7.) The alignment of the elongated alpha grains in the radial direction is in accordance with the observation that strain accompanying $\beta \rightarrow \alpha$ transformation occurred primarily radially.

X-ray diffraction revealed that the (020) and (040) reflections were very weak from the transverse section and moderately strong from the longitudinal section. Before cycling, the (020) and (040) reflections were most pronounced in the transverse section. The X-ray diffraction pattern of the transverse section after cycling was similar to the X-ray diffraction pattern (Figure 13) of the longitudinal section before cycling. All of the reflections of planes belonging to the $[010]$ zone axis were stronger than in the pattern from randomly oriented specimens and much stronger than reflections from these planes in either the longitudinal section after cycling or the transverse section before cycling. The degree of texture was less after cycling than it was before cycling. It should be noted that the degree of texture prior to cycling was not a perfect $[010]$ texture.

The anisotropic $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformation strains and the 90 degree rotation of the crystallographic and microstructural textures imply that there is a specific crystallographic orientation relationship between the parent and the product phases of the $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations. From the 90 degree rotation of the texture it can be concluded that there is more than one crystallographic variant of the orientation relationship in spite of the low symmetry of the two phases. These are characteristics to be expected of martensitic transformations.

The longitudinal section of a textured cylindrical rod specimen with the (010) plane essentially perpendicular to the surface and rod axis was polished metallographically and transformed through the $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations. The predominant planes that intersected the surface and that were in the surface of the specimen belonged to the $[010]$ zone axis. Very little surface distortion was observed after the $\alpha \rightarrow \beta$ transformation, but considerable surface distortion had occurred during

the $\beta \rightarrow \alpha$ transformation (Figure 18.) The surface rumpling may have been partly the result of martensitic shear; however, the distortion appeared to be so irregular that it could have been attributed to plastic flow caused by transformation volumetric change. Quantitative observation of martensitic tilting was impossible.

An as-cast specimen having a somewhat random structure was polished in the beta phase and quenched to -80°C . The surface was extensively rumpled but had no linear surface upheavals (Figure 19) characteristic of many martensitic transformations.

Neg. 303

250X

FIGURE 18

Surface of a Textured Specimen That Was Polished and Then $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ Cycled (The specimen was slowly cooled during the $\beta \rightarrow \alpha$ transformation. The initial texturing was accomplished by $\beta \rightarrow \alpha$ transformation under 20,000 psi compressive stress. The polished surface was parallel to the applied stress.)

Neg. 125

250X

FIGURE 19

Surface of a Specimen That Was Polished in the Beta Phase and Quenched to -80°C .

EFFECT OF APPLIED STRESS ON THE $\beta \rightarrow \alpha$ TRANSFORMATION TEMPERATURE

The temperature for the beginning of the $\beta \rightarrow \alpha$ transformation was determined as a function of applied uniaxial tensile and uniaxial compressive stresses. These temperatures were compared with those for the start of the transformation as a function of applied hydrostatic pressure (Figure 20.)

FIGURE 20

Effect of Applied Uniaxial Tensile and Compressive Stresses on the Temperature of the Start of the $\beta \rightarrow \alpha$ Transformation at a Cooling Rate of 2 C°/hr

The three methods of stressing the specimens produce virtually linear changes in the transformation temperature up to 7000 psi. The slopes of the curves are +3 C/1000 psi for tension and compression and +0.4 C/1000 psi for hydrostatic pressure. These data clearly show that there is an appreciable difference between the three types of loading. All three types of loading increase the transformation temperature. The very large temperature increases of the start of the transformation of specimens transformed in compression and the positive temperature increase of the specimens transformed in tension are difficult to explain on the basis of a diffusional transformation. The temperature increases can be explained on the basis of a martensitic type transformation.

TOTAL STRAIN ACCOMPANYING THE $\alpha \rightarrow \beta$ TRANSFORMATION UNDER APPLIED COMPRESSIVE STRESSES

The maximum compressive stress that could be utilized in the study of the $\alpha \rightarrow \beta$ transformation was 2500 psi. The creep rate of beta formed from alpha is so high that little strain above 2500 psi (other than normal creep) was observed during transformation. The magnitude of the normal creep component is apparent in Figure 21, where representative creep of the alpha and beta phases at 125 C and 2500 psi are depicted along with the total strain accompanying the $\alpha \rightarrow \beta$ transformation when it is not subjected to an applied stress.

FIGURE 21

Representative Data of the Strain Accompanying the Plutonium $\alpha \rightarrow \beta$ Transformation and Single Phase Creep of Alpha and Beta Plutonium Under Applied Compressive Stresses at 125 C

The time dependency of the strain during $\alpha \rightarrow \beta$ transformation was investigated as a function of stress and as a function of transformation rate. Typical curves are shown in Figure 22. A given applied stress influenced the $\alpha \rightarrow \beta$ transformation more than it did the $\beta \rightarrow \alpha$ transformation.

FIGURE 22

Typical Curves of the Total Strain Accompanying the Plutonium $\alpha \rightarrow \beta$ Transformation with Applied Compressive Stresses

The total strain during transformation was approximately zero for stresses between 1500 psi and 2000 psi; a variation in stress at this stress level causes little change in the total strain. This implies that most of the transformation volume change occurs in a direction perpendicular to the applied compressive stress. Accordingly, the specimens should have been highly textured, but this was impossible to establish since metallographic techniques for polishing and etching the beta phase in unalloyed plutonium have not been developed. Even so, there appeared to be a small strain opposing the volumetric change that could not be attributed either to the microstructural shape texture or to the single phase beta creep. In addition, the amount of total strain depends some on the $\alpha \rightarrow \beta$ transformation rate (Figure 23). Thus the data indicate that there may have been some creep deformation with concurrent $\alpha \rightarrow \beta$ transformation, and that the extra creep strain of the stressed specimen was only slightly dependent on the transformation rate.

FIGURE 23

Effect of Stress and Transformation Rate
on the Total Strain Accompanying the $\alpha \rightarrow \beta$ Transformation

STRAIN OF THE $\alpha \rightarrow \beta$ TRANSFORMATION CONDUCTED UNDER APPLIED TENSILE STRESSES

Extra creep deformation during $\alpha \rightarrow \beta$ transformation was more evident under applied uniaxial tensile stresses than it was under applied uniaxial compressive stresses. An applied tensile stress favors expansion in the direction of the applied stress during $\alpha \rightarrow \beta$ transformation. If all of the transformation were to occur in the direction of the applied stress without normal creep and without excess strain accompanying the transformation, then $\Delta L/L_0$ would equal $\Delta V/V_0 \approx 0.09$; this is equivalent to the $\beta \rightarrow \alpha$ transformation conducted under an applied uniaxial compressive stress. The total strain accompanying the $\alpha \rightarrow \beta$ transformation increased with applied stress and increased with decreasing transformation rate. Experiments showed that $\Delta L/L_0$ during $\alpha \rightarrow \beta$ transformation under an applied tensile stress exceeded 0.09 as evident in Figure 24.

FIGURE 24

The Total Strain Accompanying the $\alpha \rightarrow \beta$ Transformation
with Applied Tensile Stresses

During the $\alpha \rightarrow \beta$ transformation, the presence of disks of beta similar to the alpha disks of $\beta \rightarrow \alpha$ transformation has not been verified and therefore it is impossible to place limits on the range of transformation strain.

DISCUSSION OF RESULTS

Excess Transformation Creep

Some excess transformation creep was observed during $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations under applied stress, but it was much less than the anisotropic transformation strain. Excess creep, during the $\beta \rightarrow \alpha$ transformation under compressive stresses, was very small.

The model proposed by De Jong and Rathenau⁽²²⁾ to explain deformation with concurrent phase transformation is based on (1) the generation

of internal stresses during transformation because of the difference in specific volume of the parent and product phases and on (2) the average strength of the parent and product phases. According to this model, the metal deforms at a lower applied stress during transformation because the internal stresses formed during transformation are superimposed on the applied stress.

De Jong and Rathenau⁽²³⁾ correlated strain during transformation with the strength of the metal. They showed that the net strain after one complete cycle through the transformation was directly proportional to the change in volume, directly proportional to the applied stress, and inversely proportional to the average hardness of the two phases. They made the correlation for four iron alloys, cobalt, and a Cu-39.2 wt/% Zn alloy.

The data from the present investigation of $\beta \rightarrow \alpha$ and $\alpha \rightarrow \beta$ transformations conducted under stress can be interpreted in a similar manner. However, the deformation in plutonium when cycled through the $\alpha \rightleftharpoons \beta$ transition was much higher than would be predicted from De Jong's and Rathenau's correlation. For example, the predicted strain upon transformation cycling at 2500 psi is between 0.02 and 0.04. The experimental value was 0.12 when the specimens were heated and cooled at rates of 2 to 5 C°/hr.

The model presented by Sherby and Clinard⁽⁹⁾ to explain excess deformation is that excess point defects formed during transformation contribute to dislocation climb; this accelerates the deformation rate. Vacancies are formed during transformation from a less dense phase to the more dense phase; whereas, interstitials form during transformation from the more dense phase to the less dense phase. Accordingly, excess vacancies and excess interstitials should form during the $\beta \rightarrow \alpha$ and $\alpha \rightarrow \beta$ transformations of plutonium, respectively.

From this model it must be inferred that more excess creep should occur during the $\beta \rightarrow \alpha$ transformation than during the $\alpha \rightarrow \beta$ transformation. Experimentally, the opposite was observed. This can

be explained on the basis of a higher creep rate of the beta phase during the $\alpha \rightarrow \beta$ transformation which was conducted at a temperature 40 C higher than the $\beta \rightarrow \alpha$ transformation. The excess creep occurs during the latter stages of the $\alpha \rightarrow \beta$ transformation and during the first stages of the $\beta \rightarrow \alpha$ transformation. The significance of this observation is that excess creep occurs in the weaker beta phase during both the $\beta \rightarrow \alpha$ and the $\alpha \rightarrow \beta$ transformations.

Both models are compatible with the experimental evidence for excess creep deformation during the low temperature allotropic transformation under applied stress. In either of the two models, the anisotropic strain during $\alpha \rightarrow \beta$ and $\beta \rightarrow \alpha$ transformations is large compared to the sum of normal creep and extra transformation creep deformation.

Martensitic $\beta \rightarrow \alpha$ Transformation

Although the results of this investigation are not sufficient criteria for determining unequivocally whether the $\beta \rightarrow \alpha$ transformation is martensitic or diffusional, they do indicate that the transformation behaves in a manner consistent with known martensitic transformations. This conclusion is based on the following observations:

- Both uniaxial tensile stresses and uniaxial compressive stresses raise the temperature for the beginning of the $\beta \rightarrow \alpha$ transformation at a fixed cooling rate.
- Stresses applied during either the $\alpha \rightarrow \beta$ or the $\beta \rightarrow \alpha$ transformations produce pronounced crystallographic texturing and microstructural shape texturing.
- There is an apparent crystallographic relationship between the alpha and beta phases.

The alpha grains of plutonium are aligned during transformation under applied stress. The alpha grains of plutonium formed by transformation from beta under stress are also in the form of needles or rods. The grains transformed under compressive stresses are longer and thinner than those formed by transformation under no stress; the higher the stress the thinner are the alpha grains. More significantly, the elongated grains are aligned in disks which are random in the absence of applied stresses, perpendicular to applied compressive stresses, and parallel to applied tensile stresses.

Both applied uniaxial tensile and compressive stresses raise the temperature for the beginning of the $\beta \rightarrow \alpha$ transformation for a given cooling rate. That is, all applied stresses favor the alpha nucleation process. This is completely in keeping with the hypothesis that the $\beta \rightarrow \alpha$ transformation is a martensitic transformation. Similar effects have been observed in the austenite $\rightarrow$ martensite transformation in steel and in martensitic transformations in gold-cadmium⁽⁶⁾ and indium-thallium alloys.⁽⁷⁾ The shear and normal compressive components favor the plutonium $\beta \rightarrow \alpha$ transformation; the shear component of tensile stresses also favors the $\beta \rightarrow \alpha$ transformation in plutonium. There are no shear components in hydrostatic compression.

Kaufman and Cohen⁽³⁰⁾ and Patel and Cohen⁽¹⁷⁾ explain this effect in steel on the basis that the resolved shear stress aids the austenite $\rightarrow$ martensite transformation more than the normal compressive component opposes the transformation. The austenite $\rightarrow$ martensite transformation occurs by shear plus expansion. Accordingly, the shear stress in the plutonium $\beta \rightarrow \alpha$ transformation can be considered to aid the transformation more than the normal tensile stress opposes it; the metal contracts during transformation. For this explanation to be valid, the shear displacement would have to be much larger than dilational strains. These strains are not known for the plutonium $\beta \rightarrow \alpha$ transformation, but evidently the shear strain is much larger than the dilational strain.

Mechanical work done on the matrix undergoing transformation adds to the chemical free energy, but mechanical work done by the transforming region subtracts from the chemical driving force. Kaufman and Cohen⁽³⁰⁾ derived an expression relating this mechanical energy to the free energy for martensite transformations:

$$\Delta G = 1/2 V_m S \left[\gamma_o \sin 2\beta \pm \epsilon_o (1 + \cos 2\beta) \right], \text{ where:}$$

V_m is the molar volume; S is the applied stress; β is the angle between the stress axis and the normal to the habit plane; γ_o is the transformation shear strain; and ϵ_o is the dilational transformation strain. The plus sign

is used for uniaxial tension and the minus sign is used for uniaxial compression.

An applied compressive stress should raise the temperature for the beginning of the transformation more than an applied tensile stress equal in magnitude. Since this is not observed experimentally, it can be concluded that the normal components are negligible compared to the shear stresses.

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REFERENCES

1. G. V. Kurdjumov. "The Nature of Martensitic Transformations," J. Metals, vol. 11, p. 449. 1959.
2. B. L. Averbach. "The Effect of Plastic Deformation on Solid Reactions," Cold Working of Metals, American Society for Metals, Cleveland, 1949. p. 262.
3. J. F. Watson and J. L. Christian. "A Study of Austenite Decomposition at Cryogenic Temperatures," Trans. Met. Soc., AIME, vol. 224, p. 998. 1962.
4. G. B. Kurdymov, et. al. "The Effect of Prior Plastic Deformation on the Martensitic Transformation in an Fe-Cr-Ni Alloy," The Physics of Metals and Metallography, vol. 6, p. 85. 1958.
5. A. H. Cottrell. "Tensile Properties of Unstable Austenite and Its Low-Temperature Decomposition Products," J. Iron Steel Inst., vol. 151, p. 93. 1945.
6. L. C. Chang and T. A. Read. "Phase Changes in Metals - The Gold-Cadmium Beta Phase," Trans. Met. Soc. AIME, vol. 189, p. 47. 1951.
7. M. W. Burkart and T. A. Read. "Diffusionless Phase Changes in the Indium-Thallium System," Trans. Met. Soc. AIME, vol. 197, p. 1516. 1953.
8. A. W. McReynolds. "Effect of Stress and Deformation on the Martensite Transformation," J. Appl. Phys., vol. 20, p. 896. 1949.
9. F. W. Clinard and O. D. Sherby. "Strength of Iron During Allotropic Transformation," Acta Met., vol. 12, p. 911. 1964.
10. L. F. Porter and P. G. Rosenthal. "Effect of Applied Tensile Stress on Phase Transformations in Steel," Acta Met., vol. 7, p. 504. 1959.
11. M. G. Lozinsky and I. S. Simeonova. "Superhigh Plasticity of Commercial Iron Under Cyclic Fluctuations of Temperature," Acta Met., vol. 7, p. 709. 1959.
12. M. G. Lozinsky. "Grain Boundary Adsorption and Superhigh Plasticity," Acta Met., vol. 9, p. 689. 1961.
13. A. G. Guy and J. E. Paylick. "Acceleration of Deformation by Concurrent Phase Change," Trans. Met. Soc. AIME, vol. 221, p. 802. 1961.
14. M. D. Jepson and F. C. Thomson. "Acceleration of the Rate of Isothermal Transformation of Austenite," J. Iron Steel Inst., vol. 162, p. 49. 1949.
15. S. Bhattacharyya and G. L. Kehl. "Isothermal Transformation of Austenite Under Externally Applied Tensile Stress," Trans. Amer. Soc. Metals, vol. 47, p. 351. 1955.
16. S. A. Kulin, M. Cohen, and B. L. Averbach. "Effect of Applied Stress on the Martensite Transformation," Trans. Met. Soc. AIME, vol. 194, p. 661. 1952.

REFERENCES

17. J. R. Patel and M. Cohen. "Criterion for the Action of Applied Stress in the Martensite Transformation," Acta Met., vol. 1, p. 531. 1953.
18. A. B. Greninger and V. G. Mooradian. "Strain Transformation in Metastable Copper-Zinc and Beta Copper-Tin Alloys," Trans. Met. Soc. AIME, vol. 128, p. 337. 1938.
19. A. B. Greninger. "The Martensite Transformation in Beta Copper-Aluminum Alloys," Trans. Met. Soc. AIME, vol. 133, p. 204. 1939.
20. C. S. Barrett and O. R. Trautz. "Low Temperature Transformation in Lithium and Lithium-Magnesium Alloys," Trans. Met. Soc. AIME, vol. 175, p. 579. 1948.
21. Frank W. Clinard, Jr. "Behavior of Iron During Allotropic Transformation," Stanford University, November 15, 1964. (Ph. D. Dissertation.)
22. M. De Jong and G. W. Rathenau. "Mechanical Properties of Iron and Some Iron Alloys While Undergoing Allotropic Transformation," Acta. Met., vol. 7, p. 246. 1959.
23. M. De Jong and G. W. Rathenau, "Mechanical Properties of an Iron-Carbon Alloy During Allotropic Transformation," Acta. Met., vol. 9, p. 714. 1961.
24. Quarterly Progress Report, Metallurgy Research Operation, April, May, June 1964, edited by J. J. Cadwell, HW-82651, General Electric Company, Richland, Washington. July 15, 1964.
25. Quarterly Progress Report, Metallurgy Research Operation, July, August, September, 1964, edited by J. J. Cadwell, HW-84281, General Electric Company, Richland, Washington. September 15, 1964.
26. R. D. Nelson, T. K. Bierlein, and F. E. Bowman, Steady-State Creep of Plutonium, BNWL-32. January 1965.
27. R. O. Elliot, C. E. Olsen and S. E. Bronisz. "Effect of Preferred Orientation on the Electrical Resistivity of Alpha Plutonium at Low Temperatures," Phys. Rev. Letters, vol. 12, p. 276. 1964.
28. D. R. Stephens. "The Phase Diagram of Plutonium," J. Phys. Chem. Chem. Solids, vol. 24, p. 1197. 1963.
29. B. A. Bilby and J. W. Christian. "The Crystallography of Martensitic Transformations," J. Iron Steel Inst., vol. 197, p. 122. 1961.
30. L. Kaufman and M. Cohen. "Thermodynamics of Martensitic Transformations," Progress in Metal Physics, edited by B. Chalmers, Pergamon Press, vol. 7, p. 165. 1958.

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