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MICROSTRUCTURES
OF
DEFORMED ALPHA PLUTONIUM

April 1969

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MICROSTRUCTURES OF DEFORMED ALPHA PLUTONIUM

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April 1969

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MICROSTRUCTURES OF DEFORMED ALPHA PLUTONIUM

R. D. Nelson and S. D. Dahlgren

ABSTRACT

The effect of temperature, strain rate, and total strain on the deformation mode of high purity cast alpha plutonium was determined. Deformation was predominantly by grain boundary sliding at 110 °C and at a strain rate of 10^{-4} per min, whereas slip was the major deformation mode at -10 °C and 10^{-1} per min. Deformation at intermediate temperatures and strain rates was by a mixture of grain boundary sliding and slip. Recrystallization occurred concurrently with deformation only after a critical strain was reached. The critical strain decreased with increasing temperature and was independent of strain rate in the range of 10^{-1} to 10^{-4} per min. Strains of 14, 6, and 3 pct were required at 105, 110, and 120 °C, respectively, before recrystallization started. Deformation by twinning was insignificant.

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MICROSTRUCTURES OF DEFORMED ALPHA PLUTONIUM

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INTRODUCTION

The temperature and strain-rate dependency of the modes of plastic deformation were evaluated for high purity as-cast alpha plutonium. The techniques used to study the deformation were similar to those employed previously for alpha plutonium by Bronisz and Gorum,⁽¹⁾ and Spriet.⁽²⁾ Deformation structures were produced on polished surfaces by deforming with compression loading at strain rates between 10^{-4} and 10^{-1} min^{-1} , and also by impact loading.

Bronisz and Gorum⁽¹⁾ found that slip occurred on more than one slip system at room temperature, and suggested that grain boundary sliding also contributed to the deformation. Evidence of deformation twinning was obtained by them from polished and etched samples. Later, Bronisz⁽³⁾ found that higher purity plutonium twinned more frequently and had a lower yield stress than lower purity plutonium. The study by Spriet⁽²⁾ confirmed much of the earlier deformation work on alpha plutonium. Spriet found that deformation was predominantly by slip, that up to three orientations of slip traces would appear in one grain, and that deformation bands and twinning were revealed by etching. In addition, he deformed polished samples at 100 °C, but concluded that the deformation character of alpha plutonium at 100 °C and at room temperature were not essentially different. Nelson⁽⁴⁾ recently found in creep tests that high purity alpha plutonium would recrystallize concurrently with compressive deformation at temperatures between 25 and 115 °C.

Alpha plutonium has a monoclinic crystal structure with 16 atoms per unit cell and the alpha phase is stable up to about 115 °C. The effective melting temperature of alpha

plutonium is 284 °C, so that 110 °C is .69 and -10 °C is .47 of the effective absolute melting temperature of the alpha phase. The effective melting point, obtained by considering the enthalpy and entropy changes associated with all the higher temperature phase transformations,^(5,6) is much lower than the 640 °C experimental melting point of unalloyed plutonium.

EXPERIMENTAL PROCEDURE

Electro-refined plutonium having less than 300 ppm total impurities was received from Los Alamos Scientific Laboratory in the form of 1/4 in. diam cast rods. Major impurities were americium (<100 ppm) and tungsten (<60 ppm). Less than 25 ppm each of other impurities were present. The rods were cut into right half-cylinders 0.35 in. long and 0.25 in. in diam, and the flat faces along the cylinder axes were metallographically polished. To avoid loss of the metallographic polish by oxidation, the polished samples were deformed in a silicone fluid heat treating medium contained in a chamber evacuated to a pressure of 10^{-2} torr.

Deadweight loading was used to compress the samples between 4.5 and 20% in length at strain rates between 10^{-4} and 10^{-1} min⁻¹. Strain rates were determined by dividing the total deformation by the elapsed time of the test, even though the strain rate was not constant during deformation. Curves of strain versus time for samples deformed with a constant compressive stress⁽⁴⁾ show changes in the strain rate to be less than a factor of four during the testing of a given sample, which is small compared to the three orders of magnitude change in strain rate investigated. Two samples were loaded into an ordinary metallographic mounting mold and impacted by striking the mold piston with one blow of a 2-lb hammer. Deformation temperatures for the two impact samples were 25 and 90 °C.

The samples, following deformation, were washed in carbon tetrachloride and their polished and deformed surfaces were immediately inspected metallographically. Subsequently, the deformation structures were replicated with acetate replicating tape for later electron microscope examination, and the samples were then repolished and etched to reveal the microstructures of the deformed metal. Second stage carbon replicas were prepared from decontaminated acetate replicas using the procedure developed by Miller, Bierlein, and Mastel.⁽⁷⁾

Additional samples were deformed under constant compression load to determine the strain required for recrystallization to start at 110 °C. The samples were 0.35 in. long and 0.25 in. in diam and were compressed with stresses of 55,000, 45,000, and 30,000 psi.

RESULTS AND DISCUSSION

The experiments showed that alpha plutonium deforms by grain boundary sliding, slip, and deformation with concurrent recrystallization. Deformation by twinning was insignificant. Metallographic examination of polished and deformed surfaces revealed grain boundary sliding and slip. The start of recrystallization could not be determined from the deformed surface microstructures, and was ascertained from constant load deformation versus time curves. Metallography confirmed the occurrence of recrystallization. Repolished and etched samples were examined with polarized light to establish whether twinning was present.

Deformed surface structures of samples compressed 4.5% under four different conditions of temperature and strain rate (Figure 1) show how the deformation mode changes from grain boundary sliding to slip with increasing strain rate and decreasing temperature. Micrographs of samples compressed at the lowest temperature, -10 °C, are at the bottom (Figures 1c

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FIGURE 1. Structures Formed on Polished Surfaces of High Purity As-Cast Alpha Plutonium by 4.5% Deformation at Four Different Temperature-Strain-Rate Conditions

and d), and those tested at the lowest strain rate, 10^{-4} min^{-1} , are to the left (Figures 1a and c). Grain boundary sliding was predominant during deformation at the high temperature, 110°C , and at the low strain rate, 10^{-4} min^{-1} (Figure 1a), while slip traces appeared on deforming at the low temperature, -10°C , and the high strain rate, 10^{-1} min^{-1} (Figure 1d). The structure obtained on deforming at 110°C and 10^{-1} min^{-1} (Figure 1b) showed a mixture of grain boundary sliding and slip as did the structure obtained on deforming at -10°C and 10^{-4} min^{-1} (Figure 1c). The micrographs in Figures 1b and c show about the same amounts of both straight and wavy slip traces, grain boundary sliding, and general surface upheaval. Structures obtained after deforming at temperatures and strain rates intermediate to those shown in Figure 1 confirmed the change in deformation mode from grain boundary sliding at high temperatures and low strain rates to slip at low temperatures and high strain rates.

It was not certain that the 4.5% deformation given the polished samples was sufficient for recrystallization to have started. Consequently, compressive deformation versus time curves (Figure 2) were obtained for the three strain rates 3.3×10^{-1} , 3.2×10^{-3} , and $1.6 \times 10^{-4} \text{ min}^{-1}$ at 110°C for the purpose of determining the strain required for recrystallization to start.⁽⁴⁾ The start of recrystallization is indicated by the arrows and letters RS at the sharp increase in slope of the deformation versus time curves in Figure 2. Approximately 6% deformation is needed at 110°C before recrystallization can start, regardless of the strain rate. Other data show that approximately 3% and 14% strain are needed for recrystallization to start at 120 and 105°C , respectively, thus indicating an increase in the critical strain required to start recrystallization at lower temperatures. Therefore, 4.5%

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FIGURE 2. Strain Versus Time Curves Showing the Strain Required for Recrystallization to Start at 110 °C

strain was considered insufficient to cause observable recrystallization in any of the polished samples deformed at or below 110 °C.

A summary of the deformation behavior of alpha plutonium as a function of strain rate, temperature, and strain is presented in Figure 3. This schematic shows that grain boundary sliding occurs at higher temperatures and lower strain rates; that a mixture of grain boundary sliding and slip occurs during low temperature-low strain rate as well as during intermediate temperature-intermediate strain rate and high temperature-high strain rate deformation; and that slip occurs at low temperatures and high strain rates. Recrystallization does not start until a critical strain, ϵ_{RS} , is given a sample, and ϵ_{RS} becomes lower as the temperature of deformation becomes higher.

GRAIN BOUNDARY SLIDING

Grain boundary sliding was evident after deformation at high temperatures and low strain rates (Figure 1a). Slip is

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FIGURE 3. Summary of Deformation Behavior of Alpha Plutonium as a Function of Temperature, Strain Rate, and Strain, ϵ_{RS} , Required for Recrystallization to Start

not at all prominent in Figure 1a, and nearly all of the observable surface relief is due to grain boundary sliding. Grain boundary sliding was the most clearly evident of all the features disclosed by the electron microscope (Figure 4). First of all, grain elevation variation is apparent in this figure. Secondly, the grains have sheared at their boundaries. Several scratch marks that were continuous before deformation are displaced across grain boundaries. The arrow points to one scratch broken into three parts at two separate grain boundaries. This line clearly shows the direction of shear strain.

SLIP

As indicated by Figure 3, rapid deformation favors slip, and results from the impacted samples were consistent with this trend. The structure of the sample impacted at 90 °C (Figure 5) shows several grains favorably oriented for slip, with

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FIGURE 4. *Replica Electron Micrograph of a Polished Alpha Plutonium Surface After 4.6% Deformation at 110 °C and at a Strain Rate of 3×10^{-2} Min Showing Grain Boundary Sliding*

5 Microns

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FIGURE 5. *Structure Produced on a Polished Alpha Plutonium Surface by Impact Loading at 90 °C*

100 Microns

their extremities visible only because the slip traces stop or change direction there. Although only one slip trace direction is seen in any one grain in Figure 5, other areas of the same sample showed that two slip systems can be activated in a single grain. Many shorter slip traces were located parallel

to one another on the sample impacted at 25 °C instead of the straight slip traces characteristic of Figure 5. Two sets of slip traces were also observed in the sample impacted at 25 °C. The amount of strain and strain rate for the impacted samples could not be determined because the total deformation was nil and nearly all the deformation was localized in the sample extremity near the loading piston.

That slip occurred on a minimum number of slip systems can be seen by the many parallel slip traces in Figure 6. A ragged boundary was formed around the large grain in Figure 6 because only one slip system operated. Occasionally, two slip trace orientations appeared and, on rare occasions, a third trace orientation was observed (Figure 7). The sample used for Figure 7 was deformed 8% at 24 °C and at a strain rate of $3.2 \times 10^{-1} \text{ min}^{-1}$. No consistent changes in the appearance of

FIGURE 6. *Slip Lines and Irregular Grain Boundaries Produced by 2.4% Deformation at -10 °C and at a Strain Rate of 10^{-3} Min^{-1}*

Neg 0681223-6

50 Microns

FIGURE 7. *Structure Showing Three Slip Traces on a Single Grain of Alpha Plutonium*

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┌──────────┐
100 Microns

the slip traces with changes in temperature and strain rate were apparent, and narrow slip traces, broad slip bands, and wavy slip traces were visible after a variety of deformation conditions. One of the more unusual slip features observed is shown in Figure 8. A large amount of slip occurred locally because of the limited number of slip systems operative in alpha plutonium. Electron micrographs of slip traces were obtained but did not show any new details of the slip process.

STRUCTURES AFTER REPOLISHING AND ETCHING

Repolished and etched surfaces of the deformed samples showed the same features as previously reported by Bronisz and Gorum,⁽¹⁾ Bronisz,⁽³⁾ and Spriet.⁽²⁾ The abundance of slip bands appeared to be proportional to the quantity of slip traces for given deformation conditions. Twinning was extremely rare. Figure 9 shows a good example of two orientations of twins in the same grain and also twin intersection. The twin intersection is very similar to that reported by Spriet.⁽²⁾

Neg 0681223-8

50 Microns

FIGURE 8. Unusual Structure Showing 4 Slip Steps in a Sample Deformed 4.5% at 24 °C and at a Strain Rate of $4.2 \times 10^{-3} \text{ Min}^{-1}$

Neg 0681223-9

100 Microns

FIGURE 9. Twinning Revealed by Polarized Light in a Sample Deformed 4.5% at -10 °C and at a Strain Rate of $3 \times 10^{-4} \text{ Min}^{-1}$

CONCLUSIONS

- Deformation of alpha plutonium was predominantly by grain boundary sliding at high temperatures and low strain rates ($110\text{ }^{\circ}\text{C}$ and 10^{-4} min^{-1}), whereas slip was the major deformation mechanism at low temperatures and high strain rates, ($-10\text{ }^{\circ}\text{C}$ and 10^{-1} min^{-1}).
- Deformation was by a mixture of grain boundary sliding and slip at intermediate temperatures and strain rates.
- Recrystallization occurred concurrently with deformation only after a critical strain was reached. The critical strain was dependent upon temperature but independent of strain rate.
- Observation of up to three orientations of slip traces, grain boundary sliding, slip bands, twins, and recrystallization concurrent with deformation is consistent with earlier published work on deformation of alpha plutonium.

ACKNOWLEDGMENTS

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REFERENCES

1. S. E. Bronisz and A. E. Gorum. J. Nucl. Mater., vol. 2, p. 353. 1960.
2. B. Spriet. J. Nucl. Mater., vol. 12, p. 159. 1964.
3. S. E. Bronisz. J. Nucl. Mater., vol. 9, p. 101. 1963.
4. R. D. Nelson. Trans. TMS-AIME, vol. 242, p. 331. 1968.
5. A. J. Ardell. Acta Met., vol. 11, p. 591. 1963.
6. R. D. Nelson, T. K. Bierlein, and F. E. Bowman. The Steady-State Creep of High Purity Plutonium, BNWL-32. Battelle-Northwest, Richland, Washington, January 1965.
7. G. E. Miller, T. K. Bierlein, and B. Mastel. Microfractography of Alpha Plutonium - Replication Techniques, HW-79846. Available from Clearinghouse for Federal Scientific and Technical Information, Springfield, Virginia, December 1963.

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