10/10/52
MASTER

REF-807
CONF-660809-1

TRANSMISSION ELECTRON MICROSCOPY OF PU-1 W/O GA

CFSTI PRICES

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H.C. \$ 1.00; MN .50

INTRODUCTION

Because of its high atomic number, severe chemical reactivity and extreme toxicity, the metal plutonium cannot be easily examined by routine methods of transmission electron microscopy. Therefore, even though Douglass has demonstrated the feasibility of handling and examining thin foils of plutonium,¹ previous investigators have mainly relied on replication techniques.^{2,3} However, the amount of information which can be extracted from replica photomicrographs on essential dislocation behavior is necessarily limited.

In order to better correlate microstructure, particularly dislocation configuration and interaction, with the bulk properties of plutonium and plutonium alloys, an attempt has been made to directly observe dislocations in plutonium. This work has revealed distinct dislocation images and these are reported in this note.

EXPERIMENTAL TECHNIQUES

The starting material for the foils was delta-stabilized (Pu-1 w/o Ga) plutonium foil, 1.5-2.0 mils thick. A typical analysis is shown in Table I. The as-rolled foil was vacuum encapsulated and held at 350°C for two hours. The heated capsule was subsequently removed from the furnace and immediately crushed under a carbon tetrachloride dry ice bath at -50°C. Another series of specimens were also prepared from Pu-1 w/o Ga foil which had been furnace cooled in a vacuum from 450°C.

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The thinning of plutonium sheet was performed in a Glenn Dual Jet Thinning Unit.[†] The use of the immersed jets provides a continuous stream of electrolyte impinging on both sides of the specimen and has proved very successful in producing uniform, thin foils of a variety of metal alloys.⁴

The electrolyte used in this thinning unit consisted of 1 part anhydrous sodium chromate to 5 parts glacial acetic acid and the bath was maintained at 25°C during the thinning operation. In order to prevent preferential attack, the edges of the specimen were coated with Microstop lacquer. Preliminary efforts at thinning were made at approximately 300 ma at 40 volts, however further work is needed to determine the optimum thinning conditions.

It was decided to initially perform the entire thinning operation in a glovebox rather than an open faced "B" box. Only the power supply to the thinning unit remained outside of the box.

After thinning, the foil was quickly immersed in clean acetic acid and thoroughly rinsed with methanol. The foil was cut on a plastic plate under methanol and transferred in a closed container to a Plexiglas box similar to that devised by Douglass.¹ This transparent enclosure is provided with open glove ports to facilitate the handling and mounting of the thin foils. Air samples taken in the vicinity of this box during the loading and unloading of foils on the specimen rotating-tilting holder have on occasion revealed high contamination rates. Therefore, respirators are worn as a precautionary measure by all personnel in the microscope room until the foil is in the microscope specimen chamber or discarded in a sealed container. Generally, however, very little contamination

[†](Commercially available from G.E.M.S., Inc., Jeannette, Pennsylvania)

has been found in the microscope column and the room itself has remained "cold".

The thinned foils were mounted between 75 mesh copper grids in a Phillips EM-200 rotating-tilting specimen holder and examined at 100 kv.

RESULTS AND DISCUSSION

The greatest obstacle to obtaining good thin films of plutonium is the problem of rapid oxidation which occurs even under vacuum.⁵ Although the foil appears clean and bright, examination of thin areas under the electron beam reveal a dark, ragged oxide surface which obscures the metallic structure (Figure 1). However, some areas relatively free of oxide were found, particularly near the freshly-cut specimen edge.

Because of the low thermal conductivity of the plutonium foils, beam heating occurred and a considerable amount of rapid dislocation rearrangement and sub-boundary movement occurred immediately upon exposure to the electron beam. After this initial movement, no further effect of thermal stresses was observed, even at exposure periods of 30 minutes.

Figure 2 shows the sub-boundaries in such an area. The boundaries at the regions marked (B) are seen to be composed of spot contrasts of dislocations normal to the foil. These appear similar to image contrasts due to surface stress relaxation of dislocations, particularly screw orientated ones, analyzed by Tunstall, et. al.,⁶ in other metals. The dislocations form low-angle boundaries approximately 500-1000 Å in diameter, but it should be emphasized that this microstructure is probably not representative of bulk plutonium

since image forces on the dislocations in these thin sections may alter the dislocation configurations produced during beam heating. Other dislocations within the sub-boundaries are shown at regions marked (A) in Figure 2 along with a number of large voids, (V). An array of dislocations is shown in Figure 3 at higher magnification. These parallel dislocations may have been introduced during the cutting and handling of the foil and subsequently arranged in this metastable configuration during the electron beam heating of the specimen. Individual dislocations and dislocation arrays were quite stable and did not move during examination as is often the case in stainless steel, aluminum, etc. There was no evidence of stacking fault fringes produced by splitting into partial dislocations, but the limited number of dislocations observed in these initial specimens does not necessarily preclude a low value of stacking-fault energy for the Pu-1 w/o Ga alloy.

An interesting feature in the photomicrographs are the long, parallel striations across many of the sub-boundaries. These appear to be perturbations on some contamination or oxide surface layer, analogous to slip traces which persist in stainless steel. However these strata do not emanate from any particular dislocation as in the case of slip traces, but instead pass across dislocation images as seen in Figure 3. Specimen contamination was minimized by utilizing "cold fingers"⁷ kept at liquid nitrogen temperatures around the specimen throughout the observation period and dislocation movement with time due to stresses from specimen contamination as noted by Pashley and Presland⁸ was not observed in the plutonium foils. It is likely then, that these strata were produced during the severe buckling of these foil areas during the initial beam heating. Attempts will be made to examine this formation by means of cine photography.

Finally, no features in the microstructure which could be attributed to radiation damage were observed either during direct observation in the microscope or in the resulting photomicrographs.

SUMMARY AND CONCLUSIONS

As might be expected from the high degree of beam heating of the foils, the transmission photomicrographs reveal a microstructure which is probably not indicative of the heat treatment of the bulk material prior to thinning. Nonetheless, these first photomicrographs of dislocations in Pu-1 w/o Ga are an encouraging beginning in the study of dislocation behavior in plutonium and its alloys. Future work will be directed at minimizing the formidable obstacle of oxidation and obtaining larger and more representative thin areas.

ACKNOWLEDGEMENTS

The authors wish to thank S. E. Bronisz and D. Douglass for very helpful discussions and to R. R. McDonald for advice and encouragement. We also wish to thank J. Ferarese for his valuable aid in monitoring radioactive contamination.

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TABLE I

TYPICAL ANALYSIS OF PRINCIPAL IMPURITIES
IN DELTA PLUTONIUM SHEET

<u>Element</u>	<u>Impurity Content (ppm)</u>
Al	52
Cr	26
Fe	206
Ni	87
Si	101
U	80
C	92
Am	109
Pu ²⁴⁰	5.86 wt %
Ga	1.02 wt %

Figure 1. TRANSMISSION PHOTOMICROGRAPH SHOWING OXIDATION AROUND AN ETCH PIT IN A PU-1 W/O GA FOIL.

Figure 2. DISLOCATIONS WHICH HAVE REARRANGED DUE TO ELECTRON BEAM HEATING TO FORM LOW-ANGLE SUB-BOUNDARIES. FOIL HAD BEEN FURNACE COOLED FROM 450°C.

Figure 3. DISLOCATION ARRAY WITHIN SUB-BOUNDARY IN FOIL QUENCHED FROM 350°C. NOTE STRIAE ACROSS THE DISLOCATION IMAGES.