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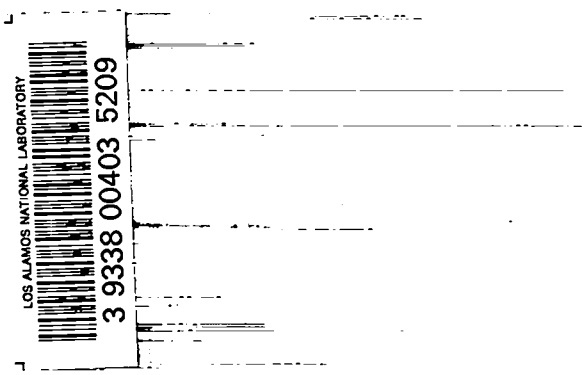
THE ALTERNATING CURRENT ELECTROETCHING OF PLUTONIUM



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LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA LOS ALAMOS NEW MEXICO

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THE ALTERNATING CURRENT ELECTROETCHING OF PLUTONIUM

by

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ABSTRACT

The equipment and procedures used in the alternating current electrolytic etching of plutonium metallographic samples are described. Examples are given which pertain to low-purity delta plutonium alloys subjected to various degrees of cold rolling and in different conditions of homogeneity, low-purity alpha plutonium in the cast and rolled conditions, and high-purity alpha plutonium that had transformed directly from the liquid state to the alpha phase under very high pressure.

ACKNOWLEDGMENT

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INTRODUCTION

The alternating current (AC) etching of high-purity alpha plutonium and delta and beta plutonium alloys made from high-purity materials has been described. (1) It is the purpose of this report to describe the application of the AC technique to low-purity* cast and rolled alpha and rolled delta plutonium, and to mention a modification of the earlier described technique in its application to high-purity alpha. This report is not intended to be comprehensive. Indeed, the almost limitless combinations and various degrees of specimen homogeneity, coring, impurity level, and mechanical treatment would seem to preclude a truly comprehensive treatment of the subject. However, the procedures described below may be helpful as guides to indicate profitable routes to follow to reveal consistently and satisfactorily the microstructural features in most plutonium samples.

EQUIPMENT

The equipment used in the AC electrolytic etching of plutonium is relatively simple (see Figure 1). It consists of a 110 V, 60 cycle, AC source; a variable transformer (10 amp capacity); voltage and current meters; and an electrolytic cell. One electrode of the cell is a stainless steel mesh placed in the bottom of a Pyrex crystallizing dish that contains the electrolyte. The polished surface of the plastic mounted specimen is the other electrode. A stainless steel probe inserted through a hole in the plastic mount provides electrical contact with the specimen. The specimen-to-electrode distance is nominally 0.5 cm, and the electrolytes are not stirred.

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* Greater than 200 ppm of impurities.

The electrolyte used in etching the delta-phase alloys contained, by volume proportions, 30 parts lactic acid, 5 parts water, and 2 parts sulfuric acid. That used in etching the alpha specimens contained 1 part nitric acid and 9 parts methanol. All specimens, after being etched, were quickly rinsed first with water and then with methanol and dried with a hot air blast.

PROCEDURES AND RESULTS

Delta Plutonium

Example 1. Plutonium-1 w/o gallium delta alloy of relatively low purity. The main impurities were iron, chromium, and cobalt. This alloy had been rolled to reduce its thickness by 27%. AC etching was accomplished at 50 V for 5 sec with the "delta" electrolyte. Following the AC etch, the specimen surface was cleaned by a DC flash treatment,* with the specimen as the anode, at 35 V for < 1 sec in an electrolyte containing, by volume proportion, 8 parts phosphoric acid, 5 parts ethanol, and 5 parts glycerine. The results are illustrated in Figure 2. Delta grains, the high impurity level, and the gallium deficiency in the regions surrounding the impurity eutectic (plutonium-iron, etc.) are revealed, but the fact that the alloy had been rolled is not obvious from the figure. The microsegregation of gallium has been described earlier. (2)

Example 2. Plutonium-1 w/o gallium delta alloy, rolled and recrystallized. This alloy has finer grains than the one in Example 1, but otherwise the microstructures are very similar. It was etched in the same manner as described under Example 1. See Figure 3.

Example 3. Plutonium-1 w/o gallium delta alloy, rolled to attain approximately 60% reduction in thickness. Figure 4 illustrates the results obtained when the specimen was etched as described under Example 1. This treatment reveals grain size and coring. The use of a lower AC voltage and a longer etching time (18 V for 10 sec) gave the results illustrated in Figure 5, where the impurities are more easily discerned.

Example 4. Plutonium-1 w/o gallium delta alloy, reduced 85% in thickness by rolling. This specimen also was etched as described

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* The equipment used is similar to that described above for the AC etching procedure, except that a DC power supply (0-50 V) and DC meters replaced the 110 V, 60 cycle, AC source and AC meters.

under Example 1. Use of a lower voltage and a longer etching time would have revealed the inclusions more clearly. The results are illustrated in Figure 6.

Alpha Plutonium

Before etching alpha plutonium specimens it is necessary to condition or "dope" the fresh "alpha" electrolyte. This is done by "etching" the stainless steel probe in the electrolyte for 30 sec at 1 amp, which causes the electrolyte to become slightly darkened. The conditioning is necessary to facilitate rinsing away the flocculent layer that forms on the specimen surface during etching. If the conditioning is not done, the layer is difficult to remove and stains the specimen.

Example 5. Low-purity, cast, alpha plutonium. This material was etched at 5 V (AC) for 10 sec in the "alpha" electrolyte and then was given the DC flash treatment as described under Delta Plutonium, Example 1. The results are illustrated in Figure 7. Though the grain size is not well delineated, the inclusions and certain "rough" areas or groups of pits are easily visible. These rough areas are indicative of coring and retention of higher temperature allotropes, as is shown by microhardness measurements. The rough areas had a hardness of 155 ± 5 DPH while that of the smooth areas was 245 ± 10 DPH.

Example 6. Low-purity alpha plutonium after being rolled in the beta phase to attain 37% reduction in thickness and then held at 110°C for 150 hr. This material, because of its many inclusions, was etched at 3 V (AC) for 20 sec. It did not require the DC flash treatment. As illustrated in Figure 8, the grain delineation is fair and the inclusions are clearly shown.

Example 7. High-purity alpha plutonium which had been transformed from its liquid state directly to the alpha phase at high temperature and very high pressure. This material was etched at 5 V (AC) for 25 sec. Notice that its grain boundaries, as illustrated in Figure 9, are different from those of alpha formed during normal cooling under vacuum or near atmospheric pressure. The grain boundaries of the alpha transformed at high pressure are much smoother and more regular than those of the more normally transformed high-purity material.

CONCLUSIONS

Fairly good reproducibility of the etched surfaces on metallographic samples of low-purity alpha plutonium and delta plutonium alloy was obtained by means of the AC electrolytic etching method described in this

report. Although the quality of some of these etched surfaces was not as good as the quality of those produced earlier on purer plutonium, under somewhat different etching conditions, the examples given here show that the AC method is applicable--by varying voltages, electrolytes, and etching times--to plutonium specimens having quite different purities and widely different mechanical and thermal treatment histories.

REFERENCES

1. K. A. Johnson, in preparation.

2. K. A. Johnson, "Homogenization of Gallium-Stabilized Delta-Phase Plutonium," LASL Report LA-2989, November 1963.

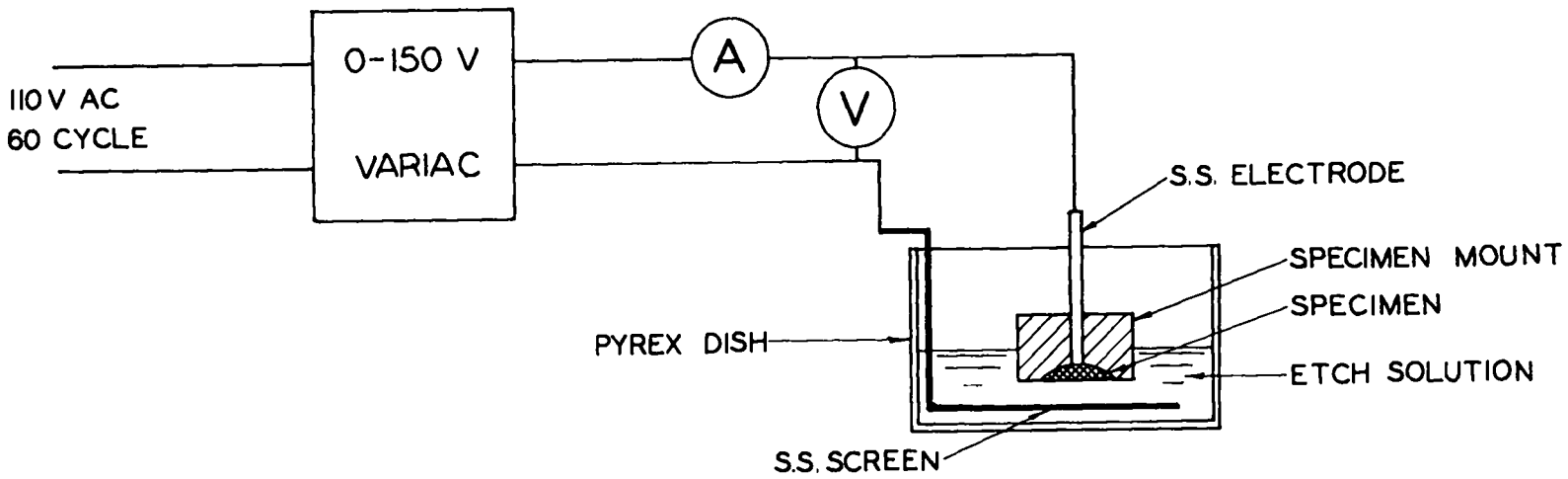

Figure 1. Schematic of equipment used in AC etching. The specimen probe and the mesh screen electrode are made of stainless steel.

Figure 2. Example 1. Plutonium-1 w/o gallium delta alloy, rolled to reduce its thickness by 27%. Note the impurities in the grain boundaries and the gallium deficiency in the boundaries and in the areas adjacent to the impurities, as shown by the relief. 500 X.

Figure 3. Example 2. Plutonium-1 w/o gallium delta alloy, rolled and then recrystallized. Similar to Figure 2 except for the finer grain size. 500 X.

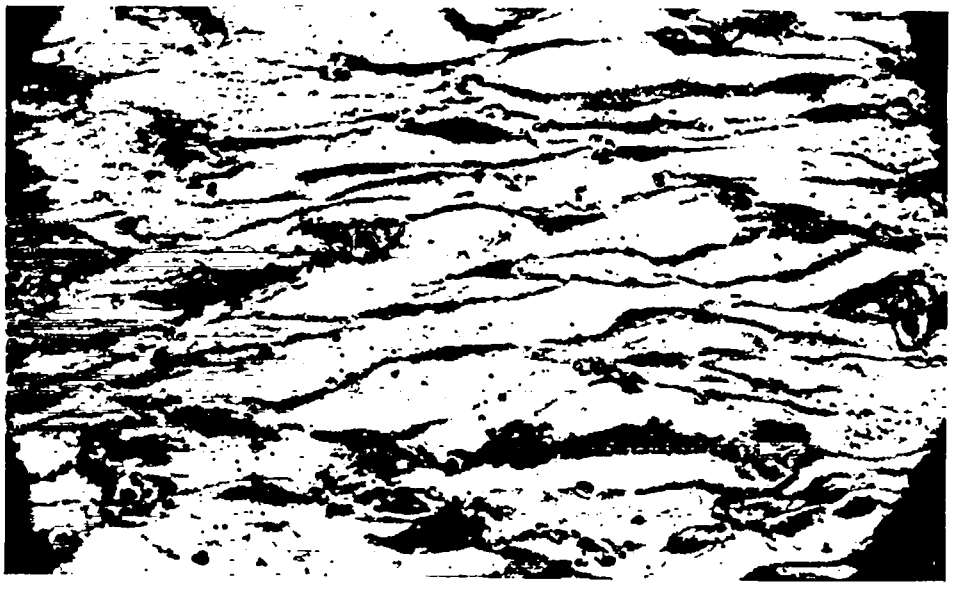

Figure 4. Example 3. Plutonium-1 w/o gallium delta alloy, rolled to attain 60% reduction in thickness. Note the residual coring as indicated by the grain boundary relief. 500 X.

Figure 5. Example 3. Same sample as shown in Figure 4 but with a lighter etch. Although the coring is not as clear as in Figure 4, the impurities are more clearly seen. 500 X.

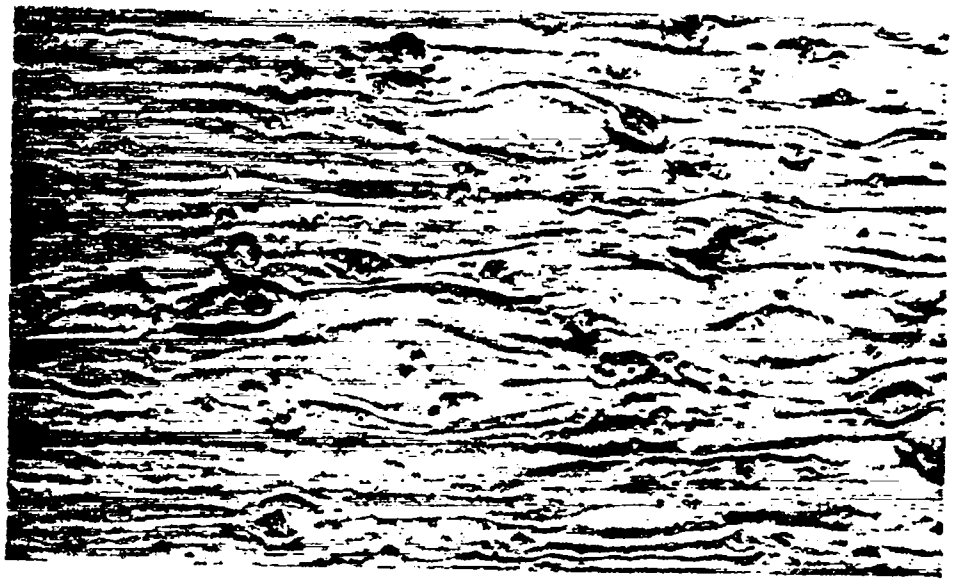

Figure 6. Example 4. Plutonium-1 w/o gallium delta alloy rolled to reduce its thickness by 85%. Note the several large, oval, unstriated areas. These areas are probably deficient in gallium and thus did not deform as much as the other areas. 500 X.

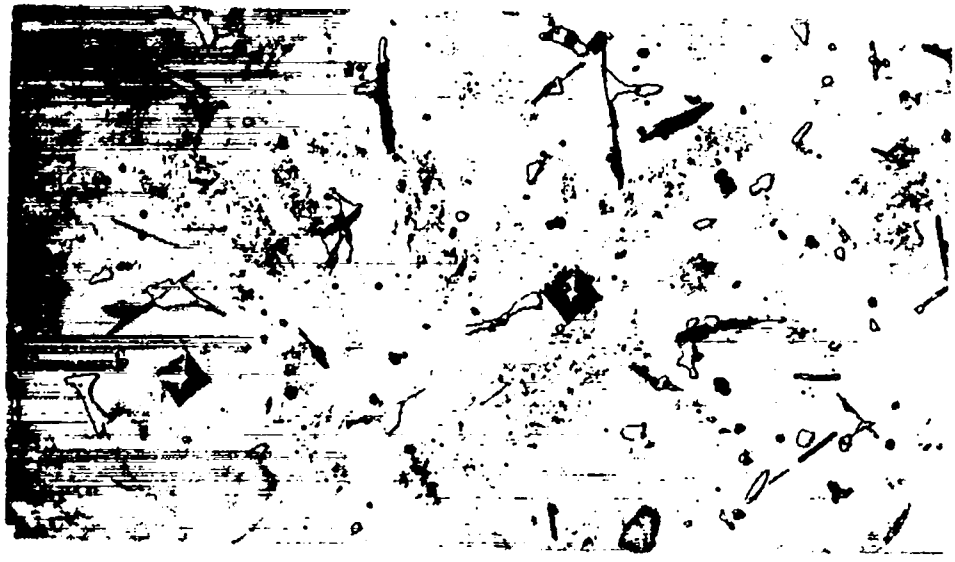

Figure 7. Example 5. Low-purity, cast, alpha plutonium. Note the pitted areas, which indicate the retention of other plutonium allotropes at room temperature. The major impurity appears to be the Pu_6Fe type compound. 500 X.

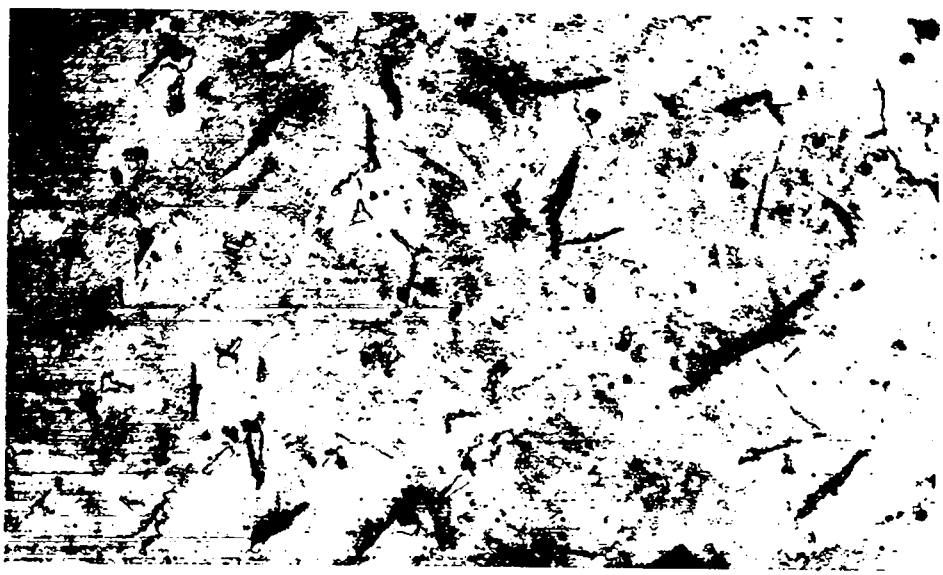

Figure 8. Example 6. Low-purity alpha plutonium, rolled to 37% reduction at beta temperatures and then held 150 hr at 110°C. 500 X.

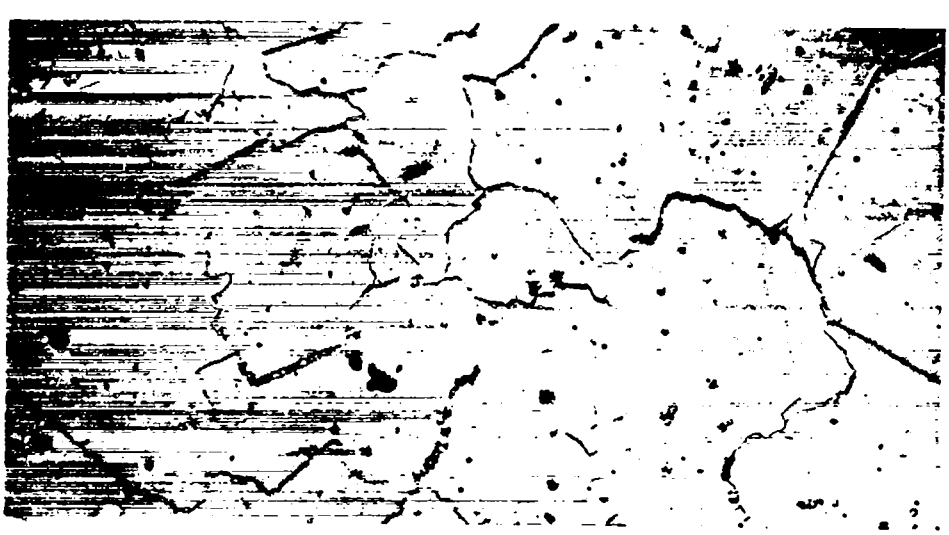

Figure 9. Example 7. High-purity alpha plutonium that had been transformed from the liquid directly to the alpha phase at high pressure and temperature. These intergranular boundaries are much smoother and more regular than those of "normal" high-purity alpha plutonium. 500 X.