Development Report

SC-DR-69-185
June 1969

A METHOD OF PRODUCING FOILS OF MULBERRY FOR TRANSMISSION ELECTRON MICROSCOPY

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TOTAL PAGES: 18

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ABSTRACT

This report presents the results of a study to determine methods of producing foils of U - 7-1/2% Nb - 2-1/2% Zr alloy to be used for transmission electron microscopy. Suitable foils were produced by spark cutting and spark planing followed by final thinning in a dual jet thinner using a solution of 6% $HClO_4$ in methanol. A chemical etch-polishing solution consisting of 35 ml HCl, 25 ml H_2O, 5 ml HNO_3 and 1 ml (25 drops) HF has also been developed for intermediate thinning to replace spark planing for faster preparation of gamma phase material.

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A METHOD OF PRODUCING FOILS OF MULBERRY FOR TRANSMISSION ELECTRON MICROSCOPY

Introduction

In order to study internal structure by means of transmission electron microscopy, a foil must be prepared thin enough to be penetrated by the beam of electrons. Uranium poses a greater problem than most metals because of its high absorbtion coefficient.

Because an alloy of U-7-1/2% Nb - 2-1/2% Zr (Mulberry) is also segragated, it contains some areas which are more chemically reactive than others. Uneven thinning can occur during preparation of foils, and this variation in thickness produces holes with adjacent areas too thick for electron transmission.

It is desirable that transmission specimens have as large an area as possible which is transparent to the electron beam. A complete foil thin enough for study would be ideal.

This goal can be attained by the polishing procedure described below.

Procedure

Table I describes the material and heat treatments of bulk material from which foils were prepared in this study.

Initial Sizing

Fifteen-mil-thick slices were spark machined from the charpy bars perpendicular to their length, using a Servamet Spark Machine* (Figure 1). Fifteen-mil thicknesses were chosen to permit removal of the damaged structure. The ten-mil sheets were cut with a pair of metal shears to a sample size of 3/16 inch x 3/8 inch x 10 mils. The bend specimens were spark sliced to yield a sample 3/16 inch x 3/8 inch x 25 mils (Figure 2).

*Metals Research Ltd., Cambridge, England.

TABLE I

Material Form	Heat Treatment
0.187 x 0.375 x 2 inch Charpy impact specimens	A. Gamma quenched in H_2O, aged at 150°C for 1 hour in air.
	B. Gamma quenched in helium, aged at 150°C for 1 hour in air.
10-mil sheet	A. Cold rolled 85%
	B. Cold rolled and gamma quenched in helium.
25-mil sheet (bend specimens)	A. Gamma quenched in helium, oxidized for 2 hours at 400°C.

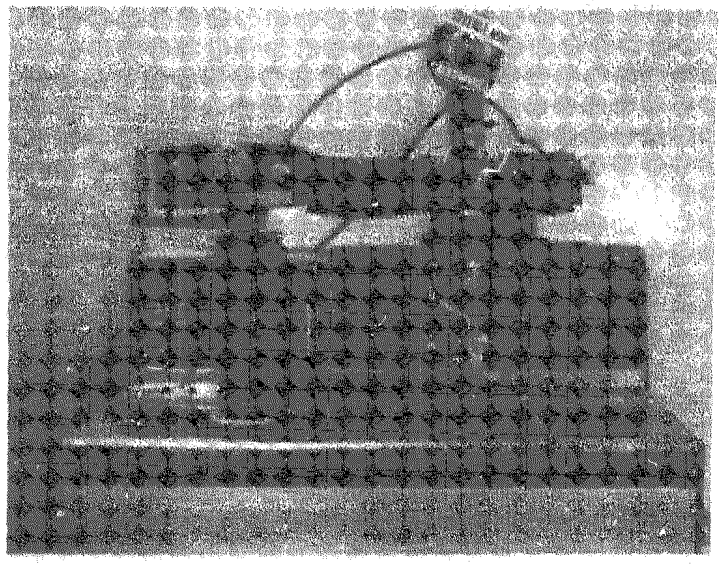
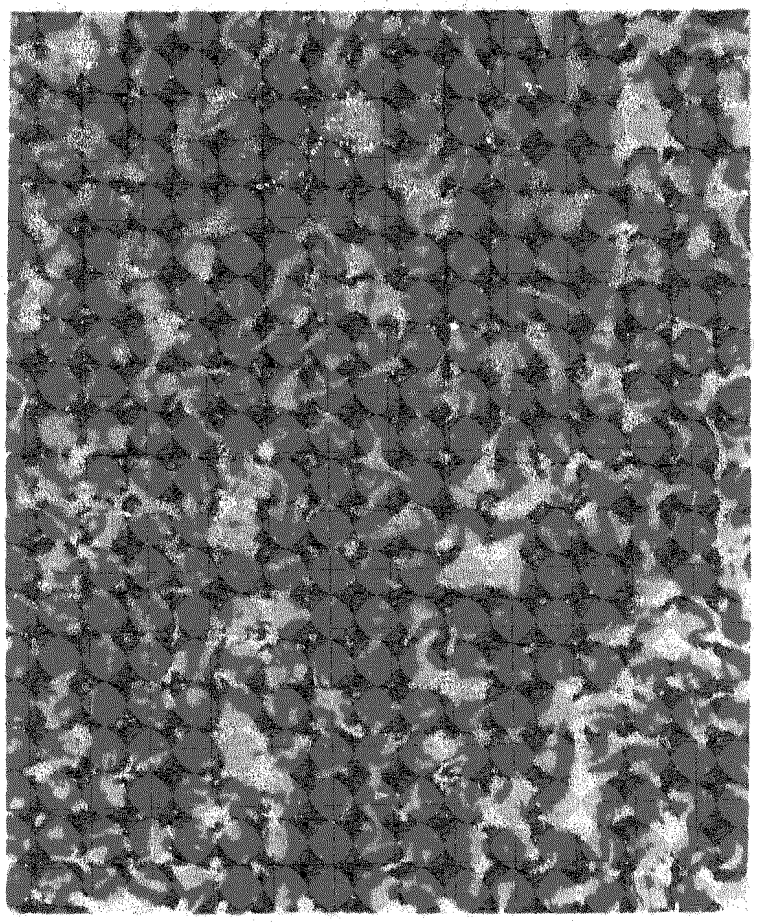
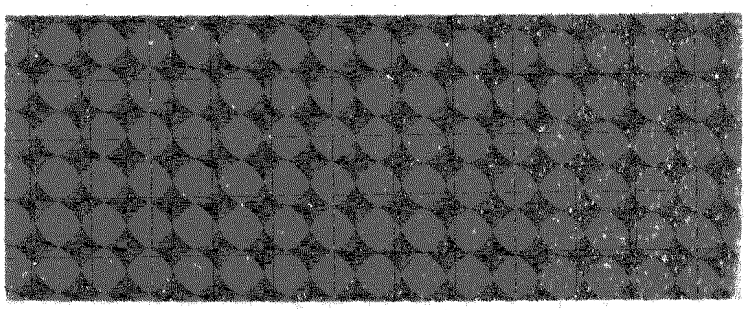


Figure 1. Servomet Spark Machine



100X Magnification



7X Magnification

Figure 2. Surface of Spark-Sliced Bend Specimen

Intermediate Thinning

In order to reduce the preparation time of the foils, a rapid chemical etch-polish solution was sought. Development of the etch-polish solution began by observing the reaction of various etching reagents with the alloy. Aqua Regia (5-ml HNO_3, 25-ml HCl, 30-ml H_2O) attacked the material, but at too slow a rate. HF was then added to help in attacking the surface layer which was presumably UO_2. It not only increased the reaction with the oxide but also speeded the general attack. By adjusting the content of HCl in the solution, an optimum surface polish condition was obtained with a solution consisting of 35-ml HCl, 25-ml H_2O, 5-ml HNO_3, and 25 drops of HF. The temperature of the reagent was increased from ambient to 60°C in steps of 10°C. As the temperature increased, the length of time for removal of the oxide decreased up to 40°C. Therefore, the solution is used at 40-50°C.

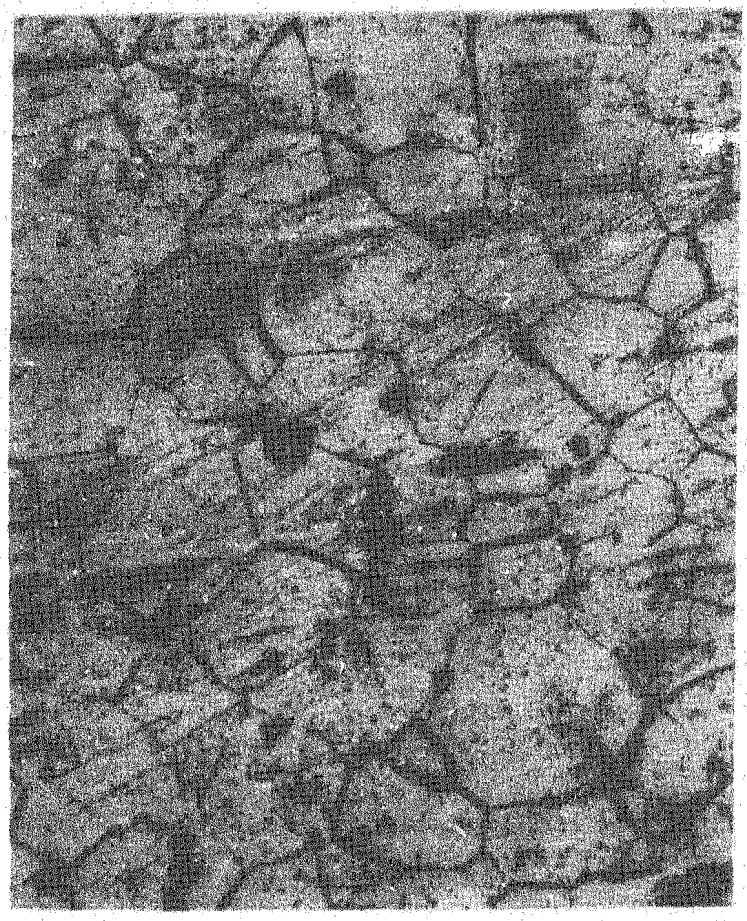
Gamma Phase Material -- The gamma phase specimens, after being spark-sliced and degreased in methyl ethyl ketone were held at one corner with a pair of gold-plated tweezers. They were alternately dipped into the solution and held in air for approximately 5 seconds until the black surface layer, produced during the spark machining, was removed.

It should be noted here that during this dipping, the reaction will proceed with some violence and care should be taken to protect the operator from splatter.

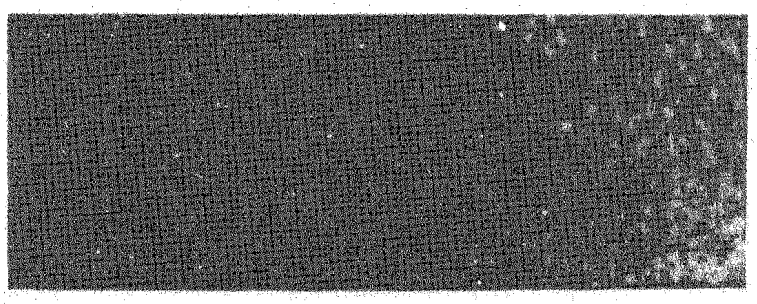
After the removal of this layer, the sample was kept under the solution, and the reaction continued.

By moving the tweezers to the diagonal corner of the specimen every 2 to 3 minutes the attack could be controlled to produce a specimen of even thickness. However, each time the specimen was removed from the solution, while the tweezers' position was changed, an oxide would form. The dipping technique was used again until the oxide was removed (Figure 3 shows a chemically etch-polished surface.)

Alpha-Gamma Material -- The two-phase bend specimens could not be thinned in the chemical etch-polish solution. One of the two phases (presumably α) was attacked much more rapidly than the other, leaving the specimen surface darkly stained and very rough. Because the etch-polish could not be used, specimens were thinned by spark planing. This was time consuming because a low-intensity spark was needed in order to assure a low depth of cutting damage. The one advantage of spark planing was the increase in the number of specimens that could be machined at one time.



100X Magnification



7X Magnification

Figure 3. Chemically Etch-Polished Surface

In all cases the samples were reduced in thickness to 7 to 8 mils. This thickness was chosen to reduce the tendency of forming a large-angle wedge at the edge of a hole upon final thinning. In addition, any surface roughness that occurred during spark machining was eliminated before final thinning was complete.

Final Thinning

The final step was the same for all specimens involved. A dual-jet thinning device, produced by G. E. M. S. Inc.,* was employed (Figure 4). A polishing solution of 6-percent $HClO_4$ in methanol was cooled to -10°C by immersion in a denatured ethyl-alcohol bath which was cooled by liquid nitrogen.

The specimen was gripped at one corner and its edges were protected from the electrolyte by coating them with an electrically insulating material called "Miccrostop".** The specimen was then placed midway between the stainless steel jets, set 1-1/2 inches apart, and perpendicular to the flow of solution with one vertical edge located in the center of the stream (Figure 5). The polishing was performed at 10 volts and with a jet speed that produced 180 milliamps. At the end of 1 minute the sample was moved until the other vertical edge was in the center of the stream. This procedure was repeated every minute until a hole formed at both ends (Figure 6). Then the sample was centered in the stream of electrolyte and polishing was continued until the holes converged. At that moment, the circuit was opened and the sample was immediately removed, washed in methanol, and dried between sheets of lens tissue. Finally, the peninsulas between the holes were cut by pressing a razor blade through the specimen (Figure 7). The cutting of thin foils often introduces damage in the sample; however, no evidence of foil bending was observed in the electron micrographs obtained.

Results and Conclusions

Results

1. Precipitates and grain boundaries can be readily observed. Figures 8-10 show typical results.
2. The polishing solution used for final thinning was stable at room temperatures and could be used for at least 30 specimens.

*G. E. M. S., Inc., Box 331-C - R. D. 2, Jeannette, Pennsylvania, 15644
**Michigan Chrome and Chemical Co., 8615 Grinnel Ave., Detroit, Michigan

Conclusions

1. The methods described above will consistently produce thin foils suitable for transmission electron microscopy studies of Mulberry.
2. The chemical etch-polish solution developed for use with gamma phase material facilitates preparation by reducing the time factor.

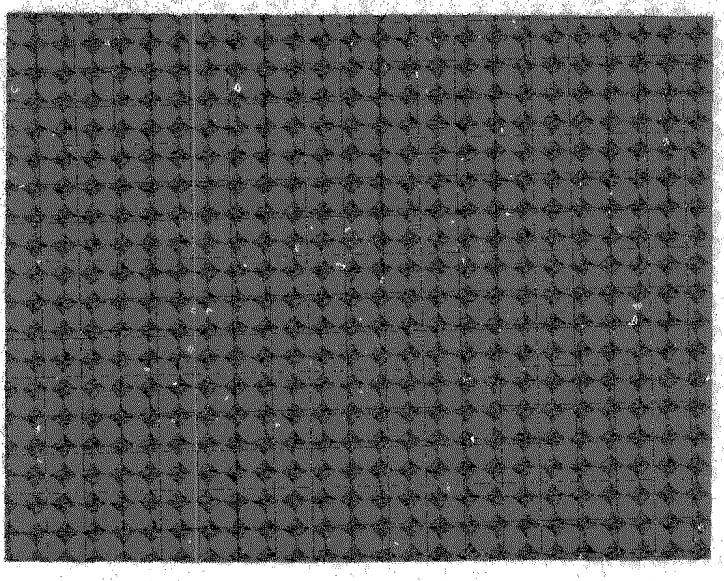


Figure 4. Dual Jet Thinning Unit

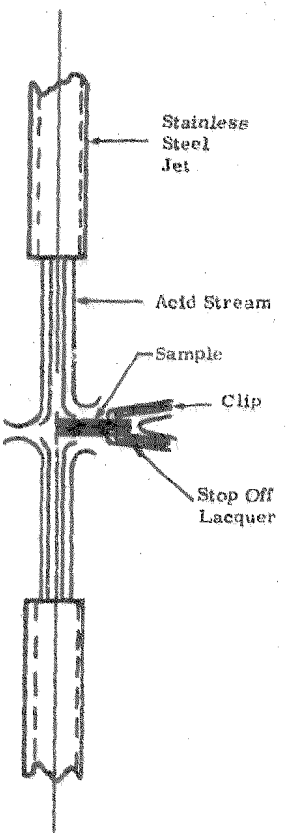


Figure 5. Position of Sample with Respect to Acid Stream During Initial Stage of Final Polishing in a Dual Jet Thinner.

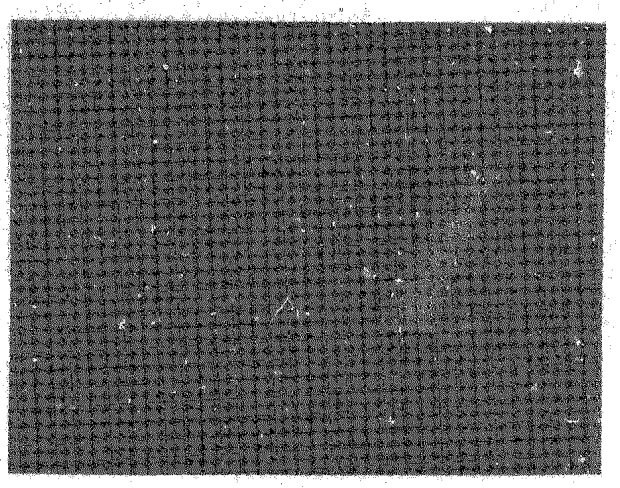


Figure 6. Sample During Initial Stage of Final Thinning. (7X Magnification)

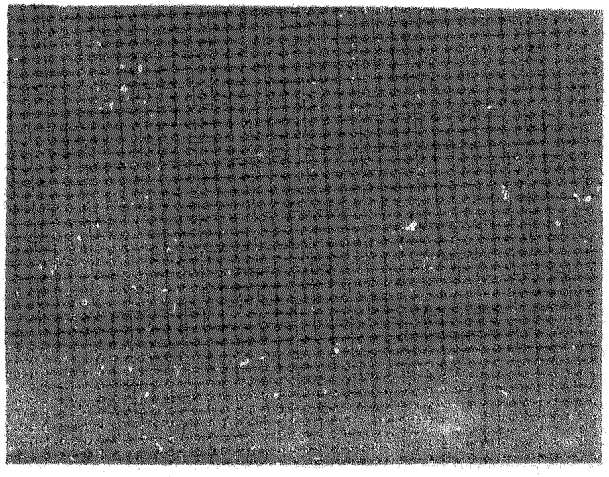


Figure 7. Sample After Final Thinning. Dashed Lines Show Cutting Planes. (7X Magnification)

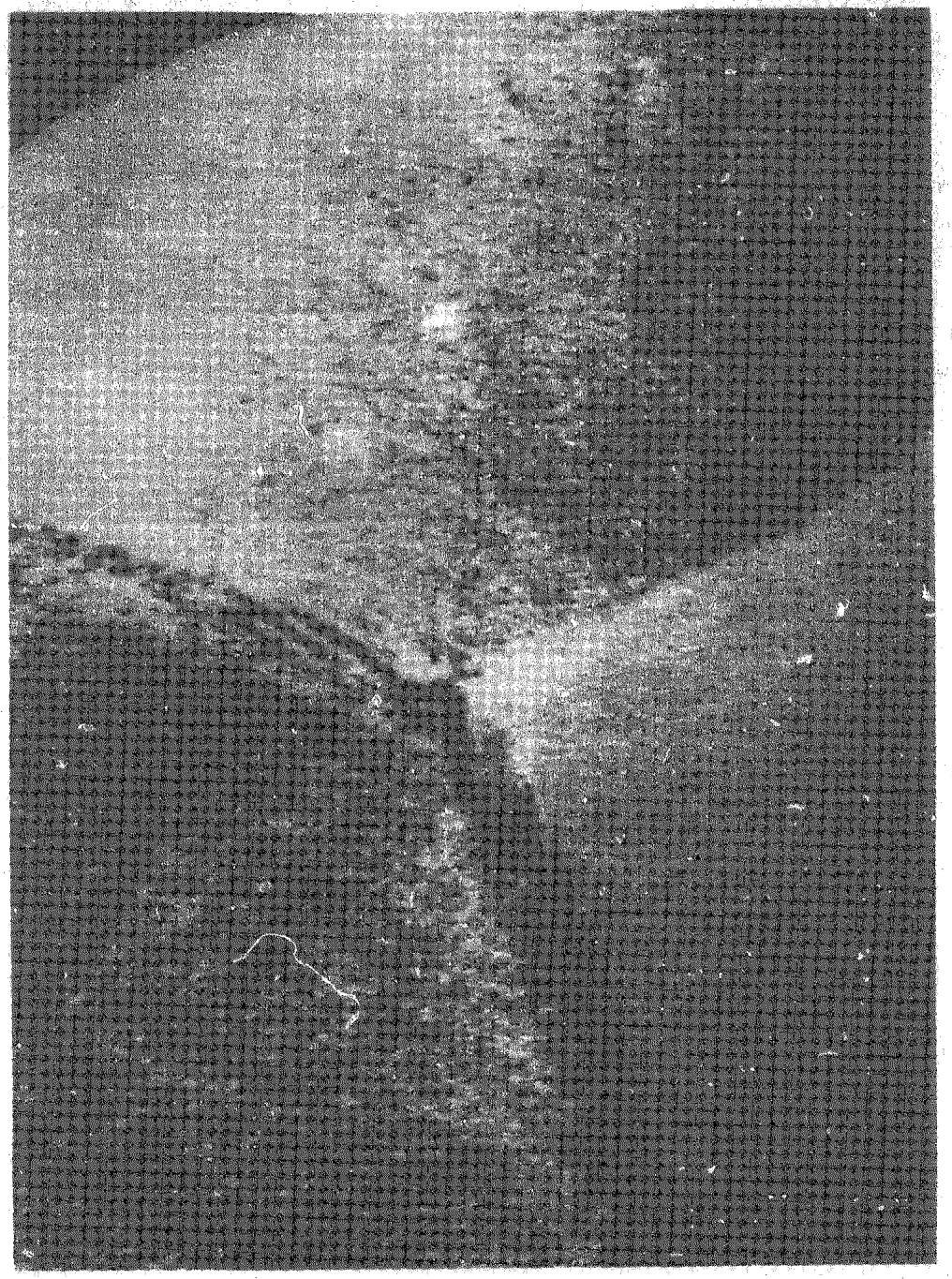


Figure 8. Triple Point Showing Precipitates in Boundaries. (40,000X Original Magnification)

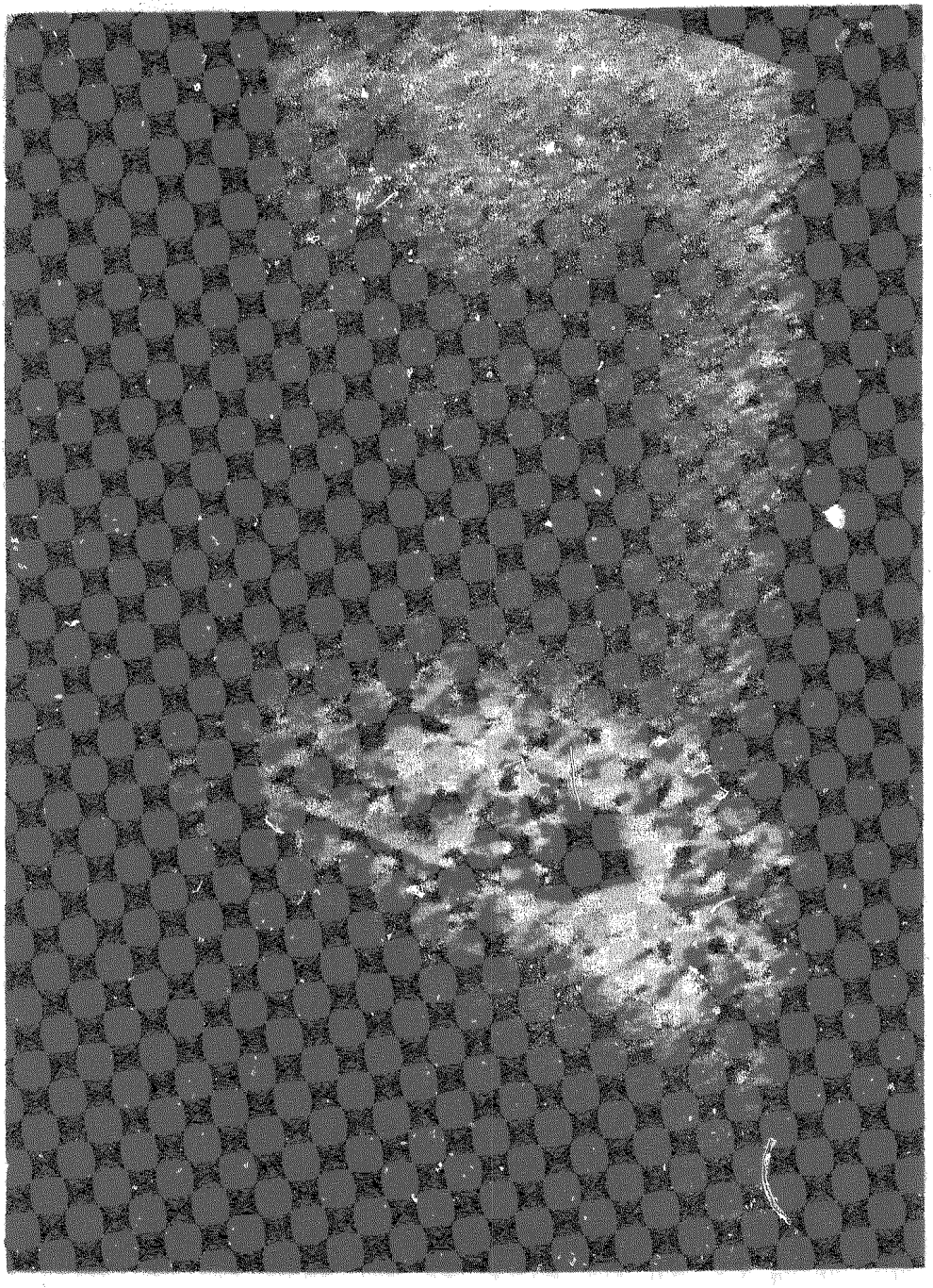


Figure 9. Precipitates within a Grain.
(78, 000X Original Magnification)

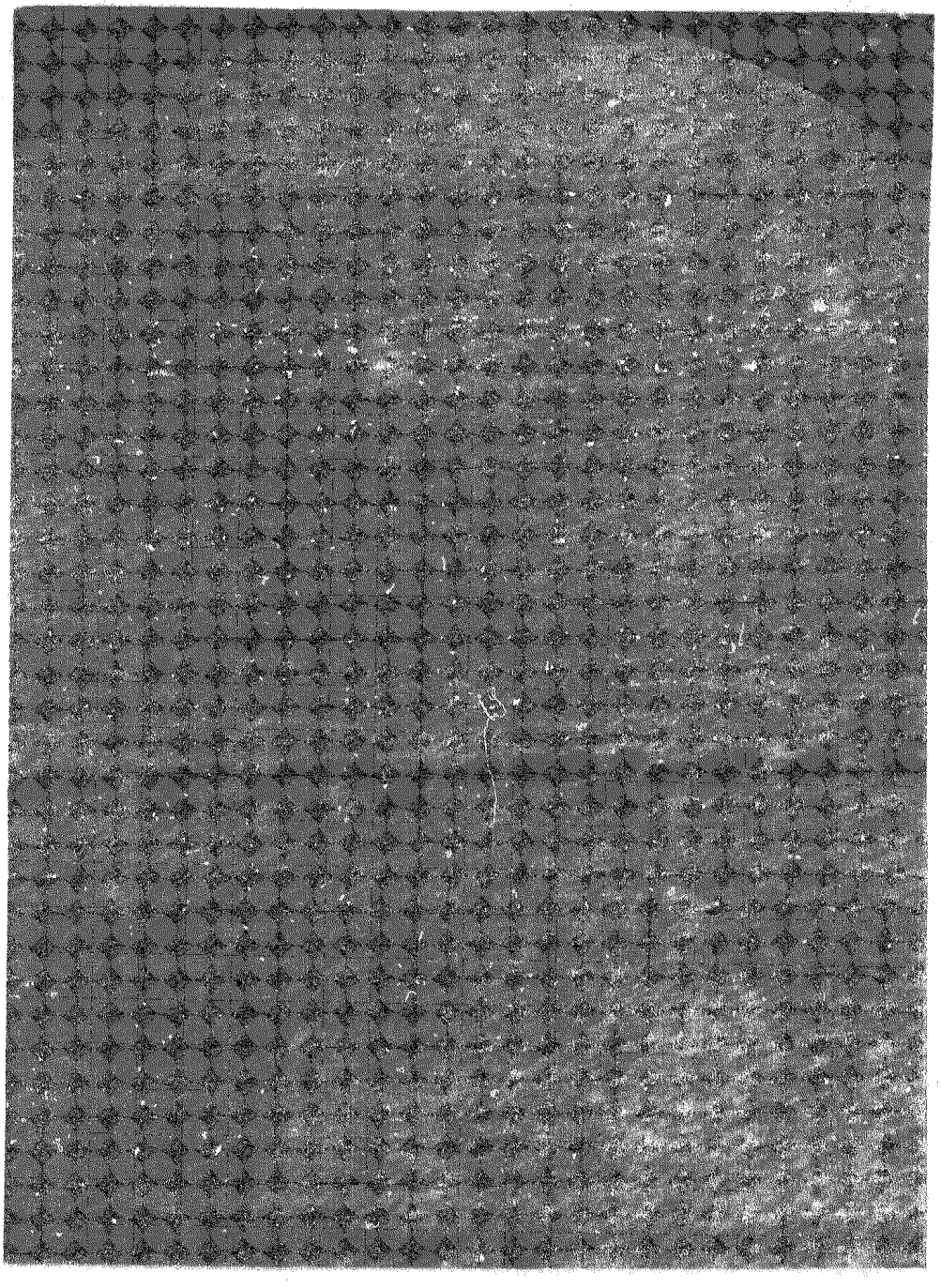


Figure 10. Line of Precipitates; may be Low-Angle Boundary. (60,000X Original Magnification)

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