Structure of the "Structureless" Replica Film for Electron Microscopy

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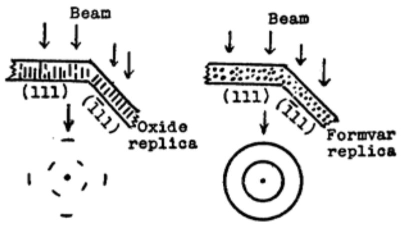
Introduction

The thin films, which reprint faithfully the elevations and depressions of the surfaces of substances, are called "structureless" replica in electron microscopy. It is known that the oxide replica(1) and electrodeposited films(2) formed on metal surfaces give more resolved images than Formvar replica. It has been demonstrated in the present study that the structureless oxide replica has a crystallographic structure distinguishable from that of Formvar or collodium replica. The reason why the oxide, sulphide and electrodeposited replica films give the resolved images in electron microscopy are to be explained in the present study.

Experimental

The specimen of iron single crystal used here was a piece of wire (diameter, 2 mm.; length, 10 mm.). This was etched with C_2H_5OH-Br (10:1 by vol.) solution for 10-30 seconds. The oxide replica was formed on this etched surface by the author's method(3) and it was stripped from the substrate by Mahla-Nielsen's method(4). The micrograph of this replica film is shown in Fig. 2. In Fig. 2 there are octahedral crystals oriented parallel to each other and the (001) planes of these crystals are inclined by 25-30° to the replica film. The electron diffraction pattern from the etched surface of the specimen showed the same inclination of (001) plane to the macroscopic surface as that in microscopic observation. This coincidence verified the faithfulness of the oxide reprint performed here.

The electron beam running perpendicular to the replica film of Fig. 2 gave the diffraction pattern of Fig. 1, which verified the existence of regularly oriented Fe_3O_4 crystals. The oxide crystals formed on metal single crystals orientate themselves according to the orientation of the substrate(5). On the contrary, Formvar replica gives the diffraction pattern consisting of diffuse haloes. The orientation of the oxide particles formed on the boundary plane (*e. g.*, (111) plane in Fig. 2) of single crystal should be distinguishable from the orientation of oxide particles of another face (*e. g.*, ($\bar{1}$11) plane in Fig. 2). Therefore, the diffraction spots obtained from the oxide replica appear in the separated positions on a back focal plane of objective lens, which are determined by the orientation of the oxide particles to the incident beam (*vid.* Fig. 3).



Diffraction pattern on focal plane

Fig. 3.—The diffraction pattern of oriented oxide replica and that of Formvar replica.

Discussion

The diffraction pattern obtained on a focal plane of a microscope behaves as a new diffraction lattice for the observable image (Abbe's principle); as a matter of fact, this was demonstrated with the electron microscope in three stages(6). It is, therefore, reasonable that the sharp diffraction spots obtained from oxide replica on a focal plane lead to more resolved images than Formvar replica giving diffuse haloes.

According to the diffraction experiment, the oxide particles formed on the polycrystalline

(1) "Metallurgical Application of Electron Microscope" Monograph and Report Series No. 8, Institute of Metals, London, 1950.
(2) R. Weil and H. J. Read, *J. Appl. Phys.*, **21**, 1068 (1950); G. M. Corney, *J. Appl. Phys.*, **22**, 682 (1951).
(3) S. Yamaguchi, *J. Appl. Phys.*, **22**, 680 (1951).
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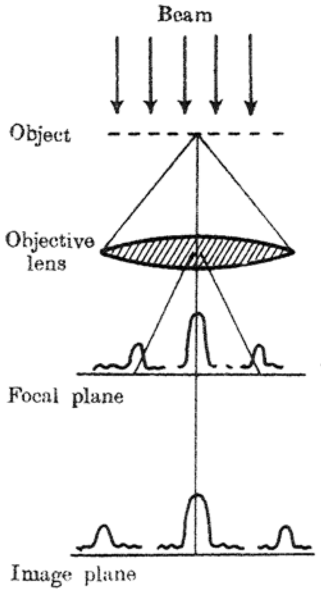
(5) H. R. Nelson, *J. Chem. Phys.*, **5**, 252 (1937).
(6) A. C. van Dorsten, H. Nieuwdorp and A. Verhoeff, *Philips Tech. Rev.*, **12**, 33 (1950); M. E. Haine, P. S. Page and R. G. Garfitt, *J. Appl. Phys.*, **21**, 173 (1950).



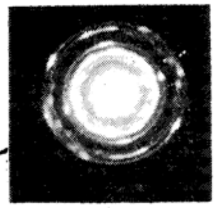


Fig. 1.

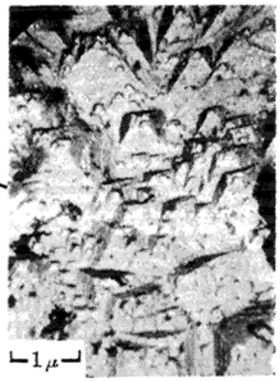


Fig. 2.

Fig. 1.—Diffraction pattern obtained by the electron beam running perpendicular to the replica film of Fig. 2.

Fig. 2.—Micrograph of the oxide replica formed on iron single crystal.

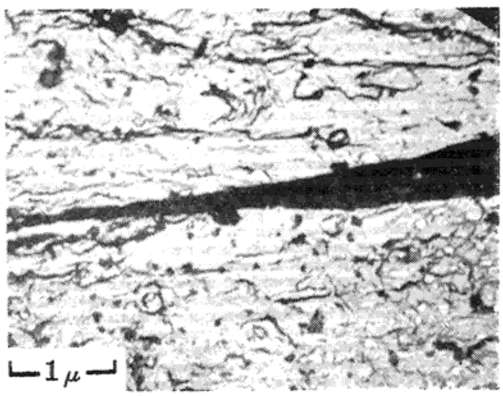


Fig. 4.—Micrograph of the sulphide replica formed on silver plate.

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metals are arranged at random. The orientation of the oxide particles to their substrate can be recognized with the oxidized surface of a single crystal, as is shown in Fig. 2. It is, therefore, plausible that the small oxide particles formed on the individual crystals of polycrystalline metal surface orientate themselves in a small area. This fact makes it possible that the oxide replica gives a resolved image and is suitable for dark-field illumination in microscopy. The same results as for the oxide replica should be applicable to the electrodeposited films on metal surfaces.

It is impossible for silver to obtain oxide replica film for microscopy. This metal gives the stable sulfide film on its surface, which is formed with sodium sulfide solution and can be stripped off the substrate with nitric acid. This sulfide film gave a more resolved image (Fig. 4) in the present study than Formvar replica. It is concluded here that the films on metals formed chemically give more resolved images than Formvar replica of metal surface mechanically prepared.

Summary

The reason the oxide replica gives a more resolved image in electron microscopy than Formvar or collodium replica is because the small oxide particles (50-100 Å.) composing replica film are oriented according to the orientation of the metal crystals of substrates. The electrodeposited replica and sulphide film formed on metals give the resolved images in microscopy. This is explained in the same way as for oxide replica.

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