JOURNAL OF MATERIALS SCIENCE 6 (1971) 1192–1196

Use of Berg-Barrett method in Electromigration Studies in 99.995% Aluminium

J. C. PIERI, J. BAGNOL, E. BERGER, R. JOUTY
Physics of Metals Laboratory, Université des Sciences et Techniques du Languedoc, Montpellier, France

When a metal carries DC current of high intensity, electromigration phenomena are produced. In the case of aluminium, the Berg-Barrett X-ray topographic technique is employed as a means of detecting and measuring the displacement of polygonisation sub-boundaries. Interpretation of the results leads to the determination of the self-diffusion coefficient of aluminium at 290°C.

1. Introduction

Electromigration phenomena can be observed in metallic specimens carrying high DC current densities. These current densities, because of the Joule effect maintain high temperature in the specimen, with associated atomic self-diffusion. The electric field responsible for the current introduces some anisotropy in this self-diffusion, the final result being a measurable displacement of markers in the specimen. Other effects which are parasitic can simultaneously affect these marker displacements [1], e.g. longitudinal or transverse contraction under surface-tension stresses, vacancy diffusion through longitudinal thermal gradients (Soret effect), creep by stresses associated with thermal expansion (the specimen being clamped at one or both current terminals).

Electromigration phenomena were detected and observed many years ago, and an extensive compilation of available results on metals, both experimental and theoretical, was published by Verhoeven [2].

More specifically concerning aluminium, the most recent results are the following:

(1) Penney [3] by optical measurement techniques, followed the displacements of a series of fine equidistant scratches along the surface of single crystal specimens due to the effect of high DC currents.

(2) Blech and Meieran [4], by electron transmission microscopy of thin films, measured the speed of void-formation near the cathode, during DC current conduction.

The currently accepted theory of electromigration phenomena was formulated by Friedel and Bosvieux [5] in 1962.

The aim of our studies was to follow the phenomenon by means of an X-ray diffraction technique: polygonisation sub-boundary displacements were measured by the Berg-Barrett technique on ribbon-shaped specimens carrying DC current along their length. These observations necessitated special adaptation of the currently used Berg-Barrett topographic technique. In this way, sub-boundary displacements during electromigration were found to be substantially the same as those of the usual inert lattice markers.

2. Specimen Preparation and Description of Apparatus

Aluminium specimens (of 99.995% nominal purity, obtained as 0.6 mm thick sheet from Aluminium Français) were prepared according to Fujiwara's technique [6]; this method, which is a combination of the critical strain-anneal and temperature gradient anneal methods, is applicable to high purity aluminium. It yields without difficulty single crystals of controllable lattice orientation; however, the dislocation content of such crystals is rather high, as much as 10^9 cm^{-2} as evaluated by electron microscopy [7] by Ham's method [8]. Single crystals thus obtained are polygonised, similarly to those obtained by the ordinary strain-anneal method without special conditions [9, 10], for the same purity. This

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polygonisation as first studied by Lacombe and Beaujard [11] using etch-pit patterns, and more recently, by Le Lann and others [12-14] by means of the Berg-Barrett technique.

Single crystal specimens are first spark-machined as 8 × 40 mm ribbons, then electrolytically thinned to a thickness of about 0.3 mm. They are mounted flat on the lateral surface of a stumatite* block, attached to the camera's goniometer head. The ends of the insulating block are fitted with electrical terminals, on which the specimen is secured by two screws. Heating technique, current control and measurement, are similar to those previously used when observing electromigration by optical microscopy [15].

The Berg-Barrett technique, as used by previously mentioned workers, raised some problems which we shall briefly review: in this large-area reflection-diffraction technique, in order to improve resolution in the localisation of sub-boundaries the thickness of the diffracting layer has to be kept small. It is necessary to use a low-energy incident X-ray beam intersecting the surface at a small angle ($< 4°$) to satisfy the Bragg condition for some set of intensely diffracting lattice-planes and the diffracted beam should be almost normal to the surface. These simultaneous conditions, together with the limited set of practically available characteristic X-ray wavelengths, severely restrict the choice of diffracting planes. When the conditions are properly satisfied, the penetration of the incident beam in the specimen may be limited to about 10 μm, avoiding undue superposition on the film of effects of sub-boundaries located at different depths.

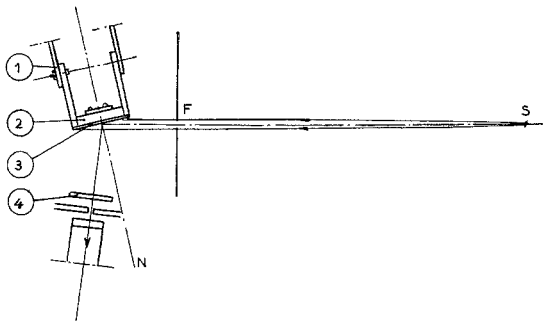

Figure 1 (1) DC current terminals; these are connected to a DC supply (80 A, about 2 V) by extra flexible wiring, the weight of which is supported by compensating springs.
(2) Baked stumatite block fixed to the camera's goniometer head.
(3) Specimen.
(4) Photographic plate, backed by slit and counter used to orient the crystal correctly before the exposure.

As X-ray source we used a Hilger microfocus unit with a focus of about 4 μm apparent width and 40 μm height. Unfiltered $K\alpha_1$ radiation from a copper target was used (focus-specimen distance being about 250 mm; a collimator slit about 1 mm wide prevents stray scattering by areas other than those for which Bragg condition is almost fulfilled).

Ilford nuclear type plates, with very fine grain and 100 μm emulsion thickness, were used. The plate was perpendicular to the diffracted beam, at a distance of 5 to 10 mm from the specimen. At such distances, there is practically no image-magnifying effect, and sub-boundary displacements may be directly measured on the plate (fig 1).

Our Berg-Barrett diffraction patterns were obtained with specimen and plate in fixed position. With this type of exposure, not every sub-boundary is detected [14]. The sub-boundaries which are visible are: (a) horizontal sub-boundaries of any angle and (b) sub-boundaries of low misorientation (about 5') of any orientation except pure twist.

Resolution obtained with fixed specimens is very good, and with about 1 h exposure time, polygonisation substructure can be easily observed. When the specimen is heated by a DC current, there is an appreciable loss of resolution owing to the rapid widening of Bragg reflections with increasing temperature. Resolution is still sufficient for detection of sub-boundaries if the exposure time is increased to about 3 h, and the temperature of the specimen is not too high. Some compromise must accordingly be accepted between high current densities giving high electric fields with associated high temperatures, promoting fast electromigration phenomena, and the moderate temperatures necessary for precise location of boundary positions. Fig. 2 is an example of a Berg-Barrett topograph prepared at room temperature showing sub-boundaries. Experimental details are listed in the Appendix. Specimens of low cross-sectional areas and high surface area – i.e. thin ribbons – are most suitable because, for a fixed temperature, they permit

*Stumatite: registered trade mark for insulating ceramic produced by "laboratoire d'Electroporcelaine" – 26 St-Vallier. This refractory in its raw state can be machined before final firing.

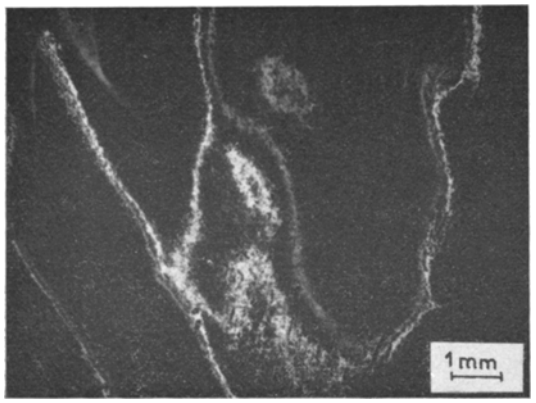

Figure 2 Single-exposure topograph. (For details see Appendix.)

higher current densities owing to the more efficient cooling through their external surface.

Even with such flat thin specimens, transverse and longitudinal temperature gradients in their central region remain quite small. Consequently, the contribution of the Soret effect to sub-boundary migration may be considered as a small correction.

Specimen temperature was measured at the end of each experiment by means of a thin-wire thermocouple attached to the specimen. The effect of temperature gradient on sub-boundary migration in the central part, where the temperature distribution show a very flat maximum, may be neglected to a first approximation.

3. Experiments

With the specimen mounted on a goniometer head, a transmission Laue pattern was first taken, and a projection plotted on the stereographic plane by conventional techniques. A suitable set of lattice planes was chosen, properly satisfying conditions specified in section 2. The specimen, now mounted as previously described on a stumatite block, was accurately positioned for maximum Bragg reflection from the chosen set of planes, the reflected beam being detected by a scintillation counter positioned as shown in fig. 1.

Next, the specimen was heated by the Joule effect for 24 h. Being clamped at both ends, it buckles slightly along its length and because of thermal stresses induced by dilatation, begins to lose contact with the stumatite support. Creep and relaxation of thermal stresses progressively stabilise the shape of the specimen during this first heating, after which measurements can be made without being perturbed by transient effects.

The curvature adopted by the specimen restricts the region where Bragg reflection can occur; nevertheless this field remains wide enough in our experiments to cover the maximum displacement of markers or sub-boundaries.

Next, and without switching off the DC current, an exposure was taken; at the end of this first exposure, the X-ray beam was switched off, a metallic shield put between specimen and photographic plate (to avoid any risk of fogging by thermal radiation), and, without moving either plate or specimen, the heating current was maintained for ten days' duration.

A second exposure was then taken on the same plate. This second exposure was overexposed with respect to the first, so that the initial and final position of subgrain boundaries could be distinguished, and the direction and speed of electromigration displacements relative to the electrodes unambiguously measured. Finally, electrode polarity was marked on the edge of plate before development. Fig. 3 shows a composite topograph prepared in this manner, and the two sets of sub-boundary positions are clearly discernible.

4. Results

To understand sub-boundary migration, the following points should be kept in mind: let us consider, in the specimen carrying DC current along its length, some reference cross-section plane cutting the specimen into two halves A and B, whose trace on specimen surface coincides with some fiducial mark. Diffusion displaces

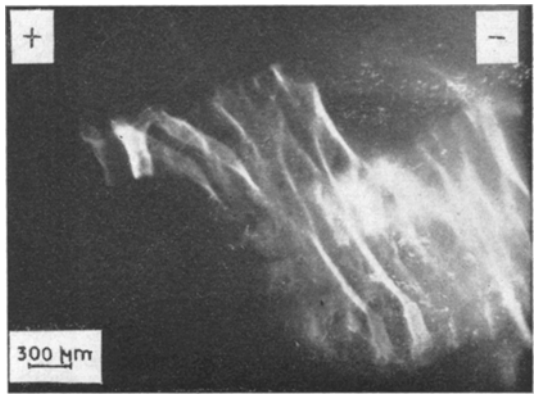

Figure 3 Double-exposure topograph. (For details see Appendix.)

atoms, and two opposite fluxes of atoms cross the reference plane in both directions. With a DC electric field, diffusion in one direction is promoted at the expense of the other; atom fluxes in both directions are no longer equal, and we have a net flux of atoms crossing the reference plane, let us say, from A to B, or a net flux of vacancies from B to A. We thus have a resultant flux of matter across our reference plane. As the ends of the specimen are kept fixed, and as the duration of the experiment is not long enough for noticeable variations of cross-section to occur in the intermediate regions, the reference plane and associated surface markers have to move, in respect to the electrodes, in a direction opposite to that of the atoms. The flux of matter across a plane fixed with respect to the electrodes is thus almost nullified. Briefly, we may use two different co-ordinate systems [16]:

(a) One (laboratory system) is fixed, rigid and rigidly connected both to specimen clamps and to photographic plate.

(b) another (local system) is connected to the crystalline lattice. It is deformable and mobile relative to the preceding one.

Fig. 3 was taken with a double exposure as previously described. It can be inferred from this that those polygonisation sub-boundaries which are detected by the Berg-Barrett technique are displaced as a whole, together with the crystal lattice towards the cathode (−) relative to the laboratory system, in a way exactly similar to that of the inert markers previously used in electromigration experiments. This confirms the well-known stability of polygonisation sub-structures in aluminium. Cathode-directed migration of sub-boundaries and crystal lattice implies anode-directed migration of atoms relative to the lattice. It is already known that in aluminium, the friction effects of electrons on crystal ions are larger than the direct electrostatic force due to the electric field (see below). As for many similar phenomena in aluminium, atom migration mechanism is achieved through a vacancy migration process.

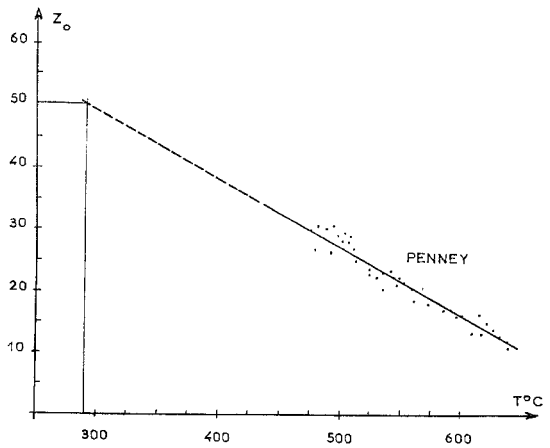

Figure 4 Extrapolation of Penney's values for the effective valency to 290° C.

From a knowledge of lattice migration speed, an "effective valence" concept has been developed, which will be briefly reviewed here [2].

Two forces act simultaneously on metal ions, producing their displacement:

(*a*) Electrostatic forces on ion cores due to the electric field E, given by $\mathbf{E} = \rho \,.\, \mathbf{i}$, where ρ is the resistivity.

(*b*) "Friction" forces due to interactions between moving conduction electron and ion cores.

The total force F is then given by

$$F = e \,.\, E\left(Z - \frac{\delta_e}{e}\right)$$

where δ_e = friction coefficient, e = electron charge, Z = valence).

Putting $F = eZ_0$ where $Z_0 = Z - (\delta_e)/e$ is defined as an "effective valence" related to the electron mobility $u = v/E$ (v = average electron velocity) and to the self-diffusion coefficient $\tilde{D}$.

Einstein's equation re ative to Brownian motion, as modified by Herring, can be written

$$\frac{v}{F} = \frac{\tilde{D}}{fkT}$$

where f is a correlation factor (= 0.83 for fcc crystals), k the Boltzmann constant and T the temperature. One obtains the following relation:

$$u = \frac{F}{E}\frac{\tilde{D}}{fkT} = \frac{eZ_0\tilde{D}}{fkT}$$

and thus the "effective valence" Z_0 is given by

$$Z_0 = \frac{ufkT}{\tilde{D}e}.$$

Assuming that the order-of-magnitude of the effective valence at 290°C can be obtained by linear extrapolation (fig. 4) of Penney's results, and taking the value of resistivity $\rho = 5.9 \times 10^{-8}$ Ω m at 290° C and $i = 26.3 \times 10^6$ A/m² (whence E can be obtained), a value of the self-diffusion coefficient $\tilde{D} = 1.3 \times 10^{-9}$ m² s^{-1} can be determined [17] from formula giving Z_0.

This value of $\tilde{D}$ pertains to the relatively low temperature of 290°C, and we could not apply the same method to substantially higher temperatures, because of excessive resolution losses. The present method is applicable only to a rather restricted temperature range, but its interest lies precisely in the fact that this range is much lower than that of classical electromigration experiments, which necessitate higher temperatures to obtain detectable displacements in a reasonable time.

Acknowledgement

We are grateful to Professor Falgueirettes, director of "Laboratoire de Minéralogie et Cristallographie II", for kindly lending us the diffraction camera and scintillation counter used in the present work.

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Received 5 April and accepted 21 April 1971.

Appendix

Particulars for fig. 2
Reflecting plane: $(\bar{1}13)$
θ: Bragg angle = 39° 13′
$\alpha_1 = 36° + \pi/2 - \theta = 86° 47'$
$\alpha_2 = 14°$
} (see fig. A1)
Time-exposure: 1 h
No heating

Particulars for fig. 3
Reflecting plane: (200)
θ: Bragg angle = 22° 26′
$\alpha_1 = 20° + \pi/2 - \theta = 87° 34'$
$\alpha_2 = 14°$
} (see fig. A1)
First exposure: 2 h 30′
Second exposure after ten days' DC current conduction: 3 h 30′
DC current: 80 A
Specimen cross-section: 0.38 × 8 mm
Central region temperature: 290°C
Substructure displacement direction with regard to the electrodes: to cathode (−)
Atomic displacement direction relative to lattice: to anode (+)
Displacement speed: 30 μm/day.

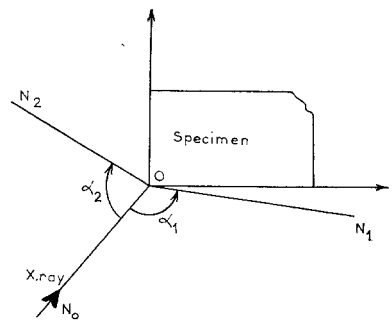

Figure A1. Geometry of Berg-Barrett exposure.