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Electromigration of polygonization substructures in aluminium

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Normal electromigration phenomenon, that is matter transport in metals subjected to high d.c. current densities, is accompanied by a quite distinct migration of polygonization substructures. This latter phenomenon has been studied in pure aluminium in the 194 to 494°C temperature range using the Berg-Barrett photographic X-ray technique, and interpretation discussed in terms of Seeger and Schottky's theory. Quantitative measurements give an apparent activation energy of 4 kcal mol^{-1} for the elementary process responsible for substructure displacement.

1. Introduction

Electromigration, i.e. matter displacement along metallic specimens carrying high d.c. current densities, is now quite well known, theoretically and experimentally, in most common metals and alloys. One can also observe in polygonized crystals, simultaneously with matter transport, an independent migration of the whole set of polygonization sub-boundaries. In fact, contrary to our previously published interpretation of single-temperature measurements, this bulk migration of a sub-boundary network is totally distinct and independent of crystal lattice migration [1].

The present study reports a series of measurements, in a substantial temperature interval 194 to 494°C, by X-ray diffraction technique, of subgrain boundary movement, in pure aluminum (99.995%) carrying high d.c. current.

2. Experiments

The method used is basically the same as that already described by us for aluminium [1] and 3.1% Si transformer grade steel [2]. Briefly, the Berg-Barrett diffraction technique was applied to polygonized, ribbon-shaped aluminium monocrystals. These were obtained by strain-anneal standard methods, electropolished, and ultimately fixed on the side of an insulating block, on a goniometer head, in the X-ray beam from a copper target Hilger microfocus unit. The specimen was oriented with the help of preliminary Laüe patterns, in order to satisfy Bragg's condition for a low-index reflecting set of crystal planes, while making a small angle, typically less than 4°, with the incident beam. X-ray penetration in the specimen is then quite shallow, avoiding on the nuclear type photographic plate undue superposition of boundaries situated at different distances from the surface; the penetration is still sufficient to allow precise detection of superficial boundaries through the overlying oxide layer. Experiments were conducted in air; oxidation, which has proved quite detrimental, impeding surface marker movements in bulk electrodiffusion measurements, has, however, no obvious influence on the phenomena described here [3].

During previous experiments [1], sub-boundary displacements under electrical current could be detected and measured up to 290°C, despite drastic loss of resolution caused, not only by widening of Bragg reflexion peak but also by buckling of the specimen, clamped at both ends, under axial compressive strain induced by dilatation.

Improvement of goniometer head design was necessary to permit nearly free axial dilatation of the specimen, thus making measurements possible in a larger temperature interval. Specimens, shaped to 10 × 55 mm ribbons, were thinned by electropolishing to 0.25 mm thickness.

The specimen was nearly horizontal, and its plane vertical, alongside a ceramic block which supported current carrying end-jaws (Fig. 1). Thermal expansion of the specimen was

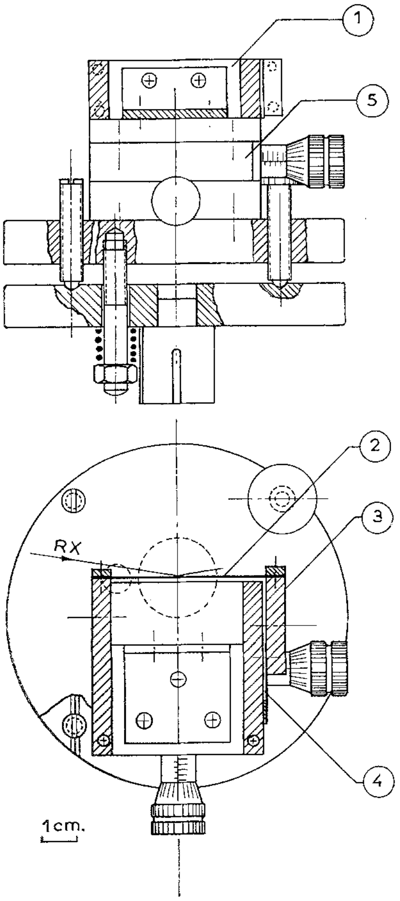

Figure 1 Goniometer head assembly. 1, Ceramic insulating block; 2, specimen; 3, mobile electrode; 4, steel spring blade; 5, ball slide.

accommodated at one end, the corresponding electrode being indirectly supported by a thin transverse steel blade spring. Specimen buckling, and associated restriction of usable diffracting field, were avoided. The whole current-carrying assembly was mounted on a specially designed, strong and stable goniometer head. Necessary adjustments of diffracting conditions can be made firstly by translating and rotating (vertical axis) the whole system (photographic plate included), using normal adjusting fixtures of X-ray camera (Lang type); and, secondly by translating the specimen above the goniometer head, with the help of two crossed micrometric slides.

The remaining three necessary adjustable degrees of freedom were provided, in the goniometer head itself, by displacement of the upper disc: this upper disc is positioned on the lower one by three ball-ended adjusting screws, according to the classical Lord Rayleigh (hole, plane and slot) scheme. A suitable coil spring maintains the screw ends in permanent contact with their receptacles.

Temperature distribution along the axis was checked, using thermocouples welded along a dummy specimen. Temperature gradients were found to be less than 20°C cm^{-1} along most of the specimen's length for a 494°C temperature in the centre. Temperature gradients of such low value were considered to be of negligible consequence for subgrain boundary migration.

Specimen polygonization being of the microscopic [4] type, a stabilization anneal was necessary before starting electromigration measurements. This annealing treatment was made by prolonged a.c. heating of the mounted specimen. Its effect is to stabilize the size and structure of subgrains and to eliminate residual internal stresses, thus avoiding subsequent parasitic displacements of sub-boundaries.

The steel blade spring which accommodates the specimen's thermal expansion exerts on the specimen an extension stress inducing unidirectional specimen creep for any d.c. current polarity. This creep increases with temperature and is a source of systematic error in sub-boundary displacement measurements.

When necessary, this error can be eliminated in one of the following ways: either (1) by separately measuring parasitic displacements during independent a.c.-heated runs, or (2) by duplicating d.c. measurements, for each specimen and each temperature, using both current polarities in succession; two different values of displacement are obtained, and, calculating their mean, the influence of parasitic unidirectional effects can be eliminated. Typically, in a rather extreme case, for $T = 420°\mathrm{C}$ (specimen illustrated in Fig. 3), a displacement of 140 μm per day is observed in one direction, and 50 μm per day in the other direction for the same part of the specimen. 95 μm per day is taken as true electromigration effect.

3. Experimental results

Sub-boundary displacements were measured by recording two successive Berg-Barrett patterns on the same photographic plate, at several days' interval, without change in electrical current or any orientation or translation parameter of the specimen, X-ray source, or photographic plate. Initial and final patterns of sub-boundaries were unambiguously identified on the plate by using widely different exposure times.

Displacements were measured relatively to electrodes, that is in the fixed laboratory reference system [5]. In experiments covering the 194 to 494° C temperature interval, sub-boundary speeds ranging from 20 to 140 µm per day were measured. These displacements were always directed towards the cathode (−), and current reversal was always accompanied by a reversed displacement. Observed sub-boundary speeds were considerably greater than speeds observed for superficial markers in bulk electrodiffusion experiments [6], a fact which made the phenomenon measurable in a much larger temperature interval. In our experiments, current intensities were in the 60 to 100 A range; specimen cross-section (measured for each specimen) was about 2.5 mm^2; current densities were thus in the 2.4×10^7 to 4×10^7 A m^{-2} range. Using different specimens with similar geometry but several different reflections – [(1 1 1) and (2 1 0)] – it was shown that measured speeds do not depend upon reflecting planes.

4. Interpretation

Sub-boundary displacement results from the combined action of the thermal agitation, and a pressure, P, related to the electrical current. Thus, the speed of displacement, at absolute temperature T, can be expressed as

$$V = A\,P \exp\left(-\frac{Q}{RT}\right) \qquad (1)$$

where Q = activation energy for elementary displacement, A and R are constants. Pressure, P, has the same sign as the corresponding displacement. The electronic structure of grain boundaries has been the subject of a theoretical model, proposed by Seeger and Schottky [7] and studied subsequently by Lormand [8]. If the same model is tentatively used for sub-boundaries, it is possible to evaluate the electrical pressure due to the electrical current, acting on any sub-boundary. Seeger and Schottky consider intergranular surface energy γ to be of essentially electronic origin, assimilating boundaries to potential barriers; the width of these barriers is equal to the boundary thickness and their height is related to γ and to bulk electronic properties of the metal.

Starting from these hypotheses and taking into consideration the fact that subgrain-boundary motion is contrary to the ionic motion of its constituents in the chosen reference system, the pressure, P, is found to be the result of two components [9] of opposite signs.

The first component, P_f, is an electrostatic pressure originating in the direct action of the electric field on the excess electronic density in the boundary. It is directed towards the anode for a positive excess density. It can be calculated from Ohm's law, giving

$$P_f = K_1\,\gamma\,\rho\,j$$

where $K_1 = 0.332$ MKS.A for aluminium, γ = surface energy of the boundary, ρ = resistivity, and j = current density.

The second component, P_c, is an electrokinetic pressure, originating in diffusion processes of electrons crossing the boundary (electron wind); it is cathode-directed. Its theoretical calculation, with the help of an electronic computer, yields numerical results which can be represented by the following approximate interpolation formula

$$P_c = -K_2\,j\,\gamma^n\,b^m$$

with $K_2 = 3.68 \times 10^{-24}$ MKS.A for aluminium, b = boundary thickness, n and m dimensionless exponents with values $n = 2.10$ and $m = 1.68$.

Depending upon the relative values of both components, the resultant pressure and sub-boundary motion may be anode or cathode-directed. Experimentally, in aluminium, a cathode-directed motion was always found, indicating a dominating electrokinetic effect.

Theoretical evaluation of the resultant pressure, P, depends on values of boundary surface energy and boundary thickness. Intergranular energy may be evaluated if the angle of misfit between adjacent subgrains is known. Accurate measurements of angular disorientation were made by electron diffraction of suitably thinned specimens of the same material, observing Kikuchi line displacements [10]. Typical misfit angles are about 50′ of arc. Following Read and Shockley's method [11], and using known values of intergranular energy for large disorientation, [12], we found for the subgrain boundaries in our experiments

$$\gamma \simeq 25 \times 10^{-8}\,\text{J}\,\text{m}^{-2} \quad (10^{-8}\,\text{J}\,\text{m}^{-2} = 1\,\text{erg}\,\text{cm}^{-2})$$

One can thus calculate, with reasonable accuracy, the value of the electrostatic component of pressure, which is typically of about 10^{-2} N m^{-2} in our experiments.

The legitimacy of using simultaneously Seeger's model, where intergranular energy is entirely of electronic origin, and Shockley's model which is based on the consideration of elastically interacting dislocations is open to discussion. In fact, a substantial part of the dislocation core energy in metals should be of electronic origin, and we feel that the evaluation of P_f given above should be at least of the right order of magnitude.

On the other hand, calculation of the electrokinetic pressure P_c critically depends upon boundary thickness b, for which no independent and sufficiently accurate basis of evaluation exists.

Nevertheless, the influence of temperature upon the resultant pressure P (coming principally from dependence of ρ upon T) may reasonably be expected to be quite small. Equation 1 may thus be simplified as follows:

$$V = A' j \exp\left(-\frac{Q}{RT}\right) \qquad (2)$$

where A' is a constant. The apparent activation energy of the global process is then identified with the activation energy of the subgrain boundary displacements.

Experimental results, plotted as $\log(v/j)$ against $1/T$ (Fig. 2), give a straight line, the slope of which gives a remarkably low value for the activation energy $Q = 4$ kcal mol^{-1} = 0.16 eV at^{-1}.

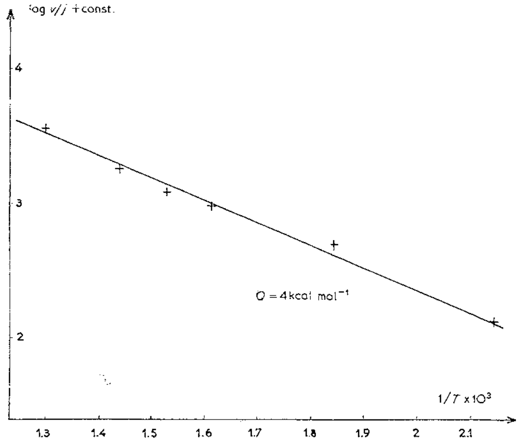

Figure 2 Log (v/j) as a function of ($1/T$) for the 194 to 494°C temperature range.

5. Conclusions

It has been shown that Seeger and Schottky's theory, applied to electromigration of polygonization sub-boundaries in aluminium, explains qualitatively the observed phenomena, and gives an evaluation of the electrostatic component of the electrically induced pressure. A value of the activation energy for the elementary process in the 194 to 494°C temperature interval was obtained. This value $Q = 4$ kcal mol^{-1} is much lower than the already known value, $Q_s = 25.9$ kcal mol^{-1} for electromigration of surface markers, as measured by Penney [6] and confirmed by the author's own measurements [13]. Comparison with activation energy for sub-boundary migration induced by other than electrical means should be quite interesting.

In the present state of study, electromigration of polygonization substructures in aluminium, although having the same direction, appears to differ fundamentally from electrodiffusion of bulk crystalline lattice.

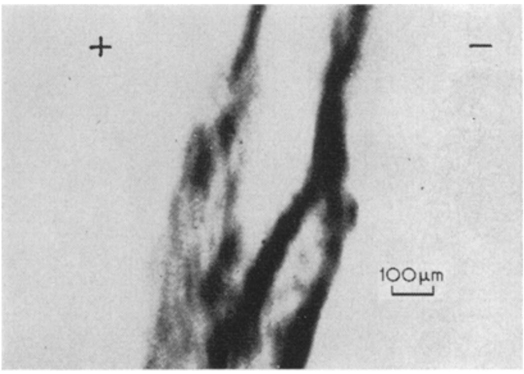

Figure 3 Double exposure Berg-Barrett photograph of aluminium specimen using (1 1 1) reflection – temperature 420°C; d.c. current 90 A; current density: 3.6 × 10^7 A m^{-2}. Time interval between exposures two days; apparent displacement results from creep and electromigration. Electromigration speed 95 μm per day.

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