JOURNAL OF MATERIALS SCIENCE 9 (1974) 644–650

A technique for the measurement of diffusion creep from marker line displacements

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A procedure is presented which allows the determination of the relative displacement of grains across a common grain boundary during diffusional creep deformation. This relative displacement comprises the strain produced by accretion of material at the grain boundary and by grain-boundary sliding. The only measurements necessary are of marker line displacements across the grain boundary.

1. Introduction

To date, the observation of structural changes which accompany diffusion creep in polycrystalline materials (excluding wires having a bamboo structure) is limited to two-phase structures. Work employing second-phase particles [1-5] has given structural evidence of diffusion creep; however, the quantitative correlation between measurements based on these materials and the macroscopic creep behaviour is limited [2-5].

In the case of single-phase materials mechanical behaviour has been used exclusively to monitor diffusion creep with the exception of some work which indicates that the angular distribution of grain-boundary sliding is different during diffusion creep than during creep at higher stresses [6]. The lack of quantitative structural observation is, at least in part, a result of the fact that no simple technique is available to monitor changes on a microscopic scale. Lee [7] proposed a method which can be used (if his equation for total grain-boundary strain is modified [8]) to determine the average amount of "grain-boundary strain" or strain produced by diffusional accretion at the grain boundaries. In principle, his method can be used to determine the relative movement of grains with respect to individual grain boundaries; however, his experimental procedure is considerably more difficult than that proposed here as it is necessary to construct a grid of precisely known line spacing on the sample surface. In contrast, the method proposed here requires only that the marker lines be straight, they can be randomly spaced, as it is only necessary to measure marker line offsets across the grain boundary.

The procedure presented here allows the determination of the relative displacement of adjacent grains which accompany diffusion creep. The displacements produced by accretion (or removal) of material and by grain-boundary sliding can be established from the measured marker line offsets.

2. Determination of the diffusional and sliding components

The method requires the use of two sets of surface marker lines, ideally at 90° to each other and at 45° to the stress axis; however, these angles are arbitrary and it is only necessary that they lie at a reasonably large angle to each other (greater than about 70°) and to the stress axis (greater than about 35°). A photomicrograph of a copper sample after creep (35 h at 805°C at a stress of 5 MPa, 3.4% strain) with two sets of marker lines is shown in Fig. 1a. The sets of marker lines both make an angle of 38 ± 1° with the stress axis and 76 ± 1° with each other (the stress axis here bisects the angle between the sets of marker lines; however, this is not necessary). Fig. 1b shows a magnesium alloy after 6.5% strain. There are three sets of marker lines on this sample, two at an angle to the stress axis and one parallel to the stress axis (vertical direction in Fig. 1b). The diffusional and sliding components across individual grain boundaries are determined as follows.

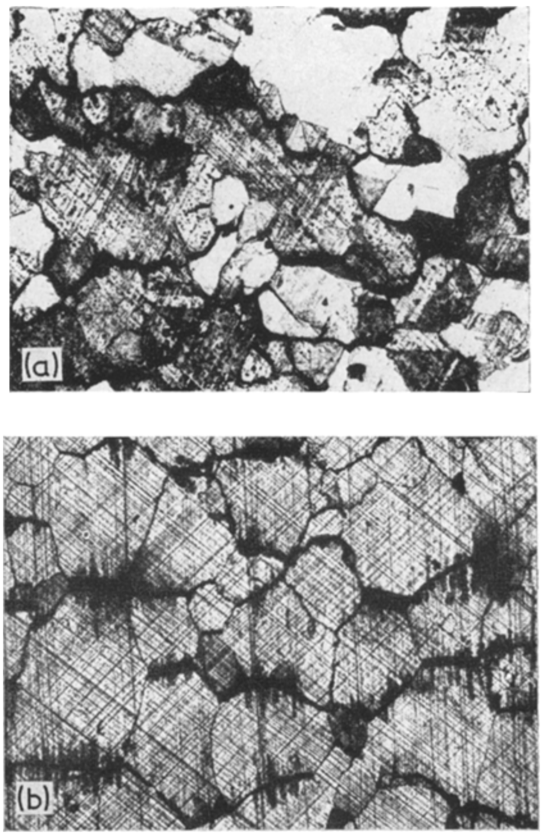


Figure 1 (a) Copper after creep for 35 h at 805° C under a stress of 5 MPa, 3.4% strain (×80). The stress axis is vertical and the marker lines are inclined at 38° to the stress axis. (The stained and spotty appearance is due to the process used to obtain the marker lines.) (b) Magnesium alloy ZR55 (Mg-0.6 wt% Zr) extruded bar annealed for 12 h at 550° C in hydrogen. Tested at 450° C for 75 h under a stress of 4.6 MPa, 6.5% strain (×65).

In general, there are displacements of grains across grain boundaries which lie at angles greater than about 45° to the stress axis produced both by accretion of material and by grain-boundary sliding. The magnitude and direction of the total displacement can be determined from the offsets of two sets of marker lines constructed as described above. (For example the total displacement of the displaced grain in Fig. 2b is the vector sum of D and S.)

The displacement vector can be broken down into two components, one produced by the accretion of material and the other by grain-boundary sliding. The sliding component is of course parallel to the grain boundary concerned, but two definitions of the accretion component have been put forward. The first puts the direction perpendicular to the grain boundary [9, 10] and the second puts it parallel to the stress axis [11]; it should be noted that the magnitude of the sliding component is different for these two cases.

The argument for taking the accretion component parallel to the stress axis as developed by Gifkins *et al* [11] is based on the necessity of maintaining coherency of the grains during creep (of a regular hexagonal array) and on the fact that the work done by diffusion is maximized. This direction is employed in the examples and calculations presented here because it appears to be more realistic in the case of a polycrystalline material [11]. However, as pointed out above,

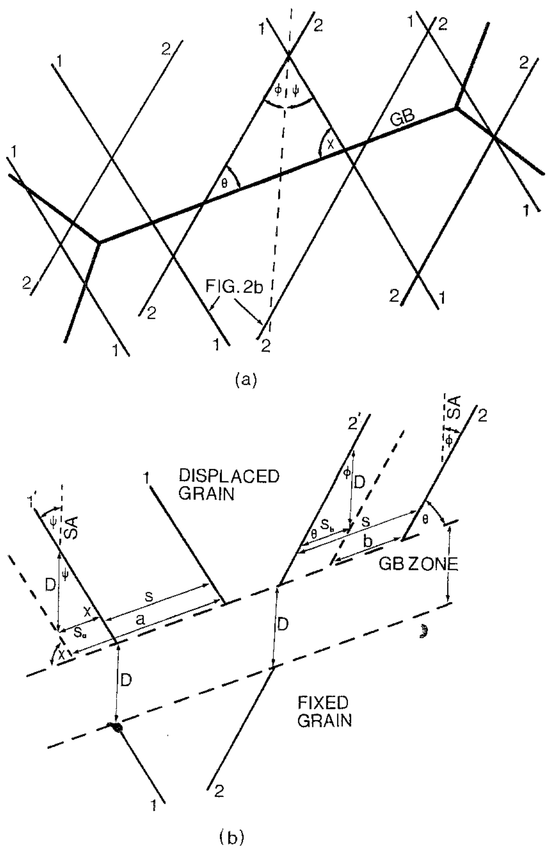


Figure 2 (a) Schematic illustration of grain-boundary marker line configuration before deformation by accretion of material and sliding. The grain boundary (GB) is represented by dark lines. The stress axis (SA) is vertical and the two sets of marker lines are labelled 1 and 2. (b) After deformation. Only the marker lines indicated by arrows in (a) are shown.

the analysis presented here determines the relative displacement of adjacent grains from marker offsets, and it is then possible to break this down into diffusional and sliding components according to a chosen definition.

Consider the schematic grain boundary and marker lines shown in Fig. 2a illustrating the geometry prior to creep. The stress axis is indicated by the vertical lines and the grain boundary lies at some arbitrary angle to the stress axis. The two sets of marker lines, labelled 1 and 2, lie at angles ψ and $-\phi$ to the stress axis, respectively. Positive angle is taken counterclockwise to the stress axis.

Fig. 2b illustrates the geometry after creep. The grain-boundary line has become a band lying between the dotted lines (G.B. zone) as a result of diffusional accretion of material. The accretion of material at the grain boundaries, which are roughly transverse to the stress axis, produces a groove where these grain boundaries intersect the sample surface (see Fig. 1). Unfortunately, the amount of diffusional accretion cannot be accurately determined from the width of these grooves because their edges are rounded by surface diffusion. The diffusional and grain-boundary sliding components can be determined from the displacement of marker lines in a direction parallel to the grain boundary.

In Fig. 2b the upper grain has been displaced arbitrarily with respect to the lower grain (fixed grain) in directions parallel to the grain boundary and to the stress axis. The displacement parallel to the stress axis produced by diffusional accretion is referred to as the diffusional component, D, and sliding parallel to the grain boundary is referred to as the sliding component. It is shown that both sets of marker lines have been displaced a distance S parallel to the grain boundary as a result of grain-boundary sliding. This distance remains to be established, it is only known that it is equal for both sets of marker lines. If the marker lines in the displaced grain in Fig. 2b, 1 and 2, are imagined to be moved this distance in a direction opposite to the sliding direction the line positions 1′ and 2′ are obtained. This is the position the marker lines would have if only diffusional accretion had occurred and no sliding had taken place.* The distance between the sets of marker lines 1′ and 2′ and the extensions of the respective marker lines from the fixed grain, shown by dotted lines in the displaced grain, in a direction parallel to the stress axis, is D. The apparent sliding distances, S_a and S_b, are the displacements of the marker lines parallel to the grain boundary produced by diffusional accretion.

It is apparent from Fig. 2b that in order to determine the unknown values of D and S, two triangles must be considered, one for each set of marker lines. For each triangle two sides and two angles are of importance, D, S_a, ψ and χ and D, S_b, ϕ and θ for the triangles associated with the marker lines labelled 1 and 2, respectively.

In order to establish S_a and S_b the sliding direction must be determined from the marker line offsets as follows. Let $a_\perp$ and $b_\perp$ be the respective offsets in a direction perpendicular to the stress axis and let a and b, the offsets parallel to the grain boundary, be positive if the offset is toward the stress axis and negative if the offset is away from the stress axis. (The stress axis is assumed to lie along a line between the two marker lines under consideration.) There are two possibilities:

(1) If $a_\perp > b_\perp$ then sliding is to the right and:

$$S_a = a - S \qquad (1)$$
$$S_b = b + S$$

(2) If $b_\perp > a_\perp$ then sliding is to the left and:

$$S_a = a + S$$
$$S_b = b - S$$

where S is not yet known. According to the law of sines,

$$\frac{\sin\theta}{D} = \frac{\sin\phi}{S_b}$$

and

$$\frac{\sin\chi}{D} = \frac{\sin\psi}{S_a} \qquad (2)$$

where ϕ and χ are angles in the triangles mentioned previously (see Fig. 2) between the grain boundary and the respective marker lines.

Combining Equations 1 and 2 and solving for D

$$D = (b + S)\frac{\sin\theta}{\sin\phi} = (a - S)\frac{\sin\chi}{\sin\psi} \qquad (3)$$

Let

$$\frac{\sin\theta}{\sin\phi} = \alpha \qquad \text{and} \quad \frac{\sin\chi}{\sin\psi} = \beta\,.$$

*This neglects the influence on the marker line positions of grain-boundary sliding which produces an offset in a direction perpendicular to the sample surface. This factor is considered in detail in Section 4.

Solving Equation 3 for S

$$S = \frac{\alpha\beta - b\alpha}{\alpha + \beta}. \quad (4)$$

Combining this with Equation 3 gives for D

$$D = \frac{\alpha\beta\,(a + b)}{\alpha + \beta}. \quad (5)$$

Equations 4 and 5 give D and S in terms of measured quantities. The quantities which must be measured are the displacement of marker line 1 parallel to the grain boundary, a, the analogous displacement of marker line 2, b, and the angles ψ, χ, θ and ϕ which are the angles marker lines 1 and 2 make with the stress axis and grain boundary, respectively (see Fig. 2).

Under diffusion creep conditions, grain boundaries which make an angle less than about 45° with the stress axis lose material and the grains on either side of the boundary move together (no grain-boundary zone is formed in this case). Here also the diffusional component perpendicular to the stress axis and the sliding component parallel to the grain boundary can be established from offsets of the two sets of marker lines. This is illustrated in Fig. 3. The measurements and equations are the same as those used in the case of diffusional accretion illustrated in Fig. 2 (in this case a and b are parallel to the stress axis).

It should be noted that in cases where denuded zones can be etched up on the sample surface it may be possible to use the width of these zones and the offsets of one set of marker lines to establish relative grain displacements. However, this procedure does not allow the determination of the amount of material removed from grain boundaries which lie at angles less than about 45° to the stress axis as no denuded zones are formed at these grain boundaries.

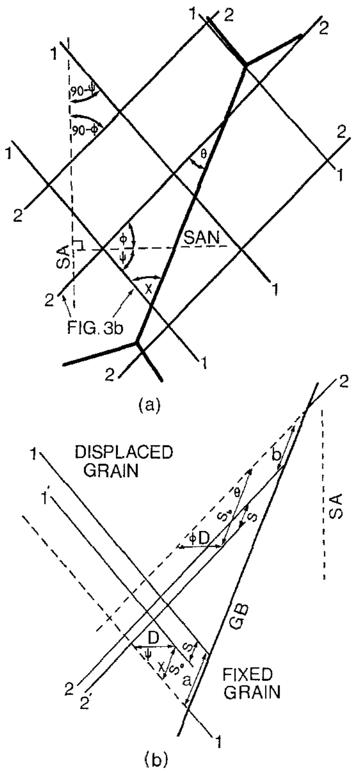


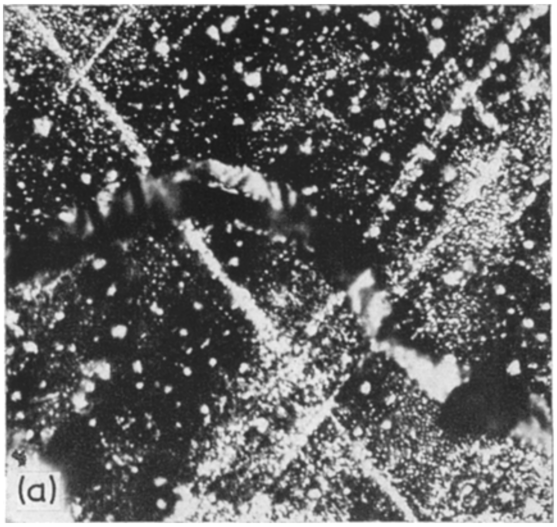
Figure 3 (a) Schematic illustration of grain-boundary marker line configuration before deformation by removal of material and sliding. Grain boundary (GB) is represented by dark lines. The stress axis (SA) is vertical (SAN is the normal to the stress axis) and the two sets of marker lines are labelled 1 and 2. (b) After deformation. Only the marker lines indicated by arrows in (a) are shown.

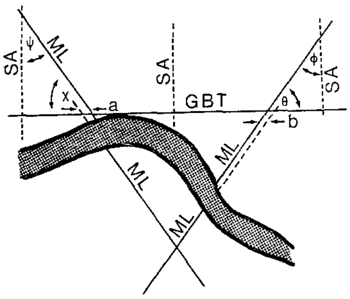
3. Experimental technique

Ideally a number of marker lines of both sets should intersect a given segment of grain boundary so the marker lines used must be fine and closely spaced. Diffusion creep generally occurs at conveniently measurable rates only in structures with a grain size of about 100 μm or less. Therefore, the marker lines should have a spacing of about 10 μm or less and be 1 to 2 μm in width. A satisfactory array of lines can be obtained by polishing with diamond powder (4 to 8 μm).

In order to obtain measurable marker line offsets a creep strain of a few per cent should be obtained. For example, for copper of 50 to 100 μm grain size this means a test of about 35 h duration at about 800°C. Marker lines of the necessary fineness, scratched on the copper surface, are destroyed by surface diffusion during a test under these conditions. This problem has been overcome in the case of copper by coating the polished surface with a thin layer of vapour-deposited carbon and placing the fine marker lines in this layer. These marker lines remain observable under the test conditions mentioned above. A photograph of a surface after such a test is shown in Fig. 1a. To illustrate the application of the measurement technique two examples from this sample will be considered.

A segment of a grain boundary is shown in Fig.





(b)

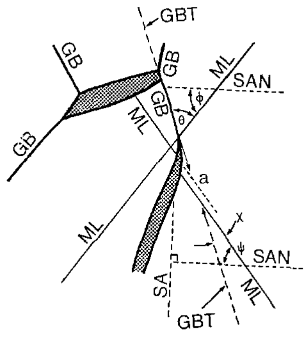
Figure 4 (a) Area from sample shown in Fig. 1 showing detail of marker lines and grain-boundary zone produced by accretion of material and sliding (×940). (b) Tracing of (a) showing the grain-boundary zone (shaded) and one marker line (ML) from both sets. The grain-boundary tangent (GBT) and stress axis (SA) are indicated. Measured angles and marker line displacements are: $\psi = 38°$, $\chi = 52°$, $a = 3.0$ μm, $\phi = 38°$, $\theta = 53°$ and $b = 2.1$ μm.

4a (the marker line offsets in Figs. 4a and 5a can best be seen by holding these figures at a low angle to the line of sight) and a tracing of this area is shown in Fig. 4b which includes the grain-boundary zone, one marker line from both sets and the data necessary to determine D and S. The values obtained are 3.3 and 0.4 μm, respectively. These values hold only for the point where the grain-boundary tangent is taken in Fig. 4b since the values of D and S vary continuously along the grain boundary as a function of the orientation of the grain-boundary tangent.

Another grain boundary which lies roughly parallel to the stress axis and has lost material is shown in Fig. 5a and a tracing of the area is shown in Fig. 5b. The measured quantities are indicated and S and D are found to be 1.3 and 1.5 μm, respectively. Again these values hold only for the point on the grain boundary where the grain-boundary tangent is taken in Fig. 5b since they vary as a function of the orientation of the tangent. In Fig. 5b the tangent is the same over a segment of the boundary, thus the values of S and D are constant over this segment.

For magnesium, marker lines of the necessary fineness remain observable after tests of four days or more at 450°C. No carbon coating is necessary.

It should be noted that the offset measure-



(b)

Figure 5 (a) Area from sample shown in Fig. 1 showing detail of marker lines and grain boundary deformed by removal of material and sliding (×570). (b) Tracing of (a) showing grain boundaries (GB), grain-boundary zones (shaded) and one marker line (ML) from both sets. The grain-boundary tangent (GBT), the stress axis (SA) and stress axis normal (SAN) are indicated. Measured angles and marker line displacements are: $\psi = 52°$, $\chi = 16°$, $a = 5.5$ μm, $\phi = 52°$, $\theta = 59°$ and $b = 0$ μm.

ments necessary for the determination of D and S sometimes can be conveniently made on the ground glass screen of a projection metallograph.

4. Discussion

The technique presented here can be used to separate the sliding and diffusional contributions to diffusion creep either under the assumption that diffusional accretion is parallel to the stress axis and sliding is parallel to the grain boundary or under the assumption that diffusional accretion is perpendicular to the grain boundary (these assumptions are the subject of current debate [11]). It should be noted that the diffusional component as defined in general produces a component parallel to the grain boundary which has previously been defined as a sliding component [9, 10].

Quite apart from matters of definition, the assumption that the diffusional component is parallel to the stress axis is not strictly true for a polycrystalline material because of stress concentration effects which do not appear in the uniform hexagonal or cubic arrays usually considered theoretically [9-11]. However, it is likely that deviations will be averaged out if the average value of a number of measurements is taken.

An error is introduced into the determination of D and S because of grain-boundary sliding in a direction perpendicular to the sample surface. This can be corrected for on an average basis as follows. The average angle at which grain boundaries intersect the sample surface is from 57° for a random surface through a grain structure to about 80° at a surface after annealing (or creep) [12]. For a given boundary the height difference across the boundary, v, produced by this sliding can be measured (by the difference in the "in focus" position of the grains on either side of the boundary at the same time offset measurements are being made). The relative displacement of the surface marker lines in a direction parallel to the sample surface and normal to the intersection of the grain boundary with the surface is given, on the average by $v \cot 80°$ where the average angle of intersection is taken to be 80°. The offset of the marker lines produced by this effect will generally be in a direction to increase the marker line displacements (except in cases of negative grain-boundary sliding) and, therefore, must be subtracted from the measured displacements, after correction for grain boundary and marker line angles.

For the grain boundaries shown in Figs. 4a and 5a, v was too small to be measured; however, an average value of v of about 0.5 μm is expected for this sample. In this case the average displacement of the marker lines in a direction parallel to the stress axis is about 0.09 μm. For a grain boundary at 90° to the stress axis and marker lines at 38° to the stress axis, as in Fig. 4, the error in the marker line offsets is about 0.07 μm. The measured marker line offsets in Fig. 5 are 3.0 and 2.1 μm. For offsets of this size the 0.07 μm error produced by sliding normal to the sample surface can safely be neglected as it is within the accuracy to which the offset measurements can be made; however, in the case of smaller offsets or larger values of v a correction may become necessary.

It should be noted that the procedure presented here allows determination of S and D even when grain-boundary migration has occurred. Migration does not alter the measured offsets. This can be visualized by imagining migration of the grain-boundary zone in Fig. 2b. It is apparent that the zone can be shifted in a vertical direction without altering the offsets, a and b.

When grain-boundary sliding in a direction perpendicular to the sample surface is either taken into account or shown to be negligible the general procedure presented here yields diffusional and sliding components on a microscopic scale and has a number of possible applications four of which are as follows.

1. The diffusional and sliding components can be determined as a function of the orientation of the grain boundary relative to the stress axis.
2. It may be possible to establish the influence of grain-boundary structure on these components as well as their variation along a given boundary.
3. It may be possible to establish the contribution of diffusional accretion and sliding to the total strain during diffusion creep.
4. It may be possible to determine the relative importance of diffusion creep with respect to other modes of deformation in particular cases of interest.

Acknowledgement

Financial support provided by the Commonwealth Scientific and Industrial Research Organization in the form of a University of Melbourne

Fellowship and valuable discussions with Dr R. C. Gifkins are gratefully acknowledged.

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Received 3 July and accepted 27 November 1973.