The effect of solutes on grain boundary mobility during recrystallization and grain growth in some single-phase aluminium alloys

Y. Huang, F.J. Humphreys

BCAST, Brunel University, Kingston Lane, Uxbridge, Middlesex UB8 3PH, UK
Manchester Materials Science Centre, The University of Manchester, Grosvenor Street, Manchester M1 7JL, UK

ARTICLE INFO

Article history:
Received 8 July 2011
Accepted 11 November 2011

Keywords:
Alloys
Annealing
Electron microscopy
Interfaces
Thermodynamic properties.

ABSTRACT

The effect of solutes (Si, Mn, Mg) in quantities typical of commercial aluminium alloys, on grain boundary mobility in aluminium, has been investigated with in situ annealing and electron backscattered diffraction in the SEM, and grain growth experiments. The in situ experiments provided information on the migration of the high mobility tilt boundaries of misorientations close to 40° (1 1 1). Grain growth experiments were used to investigate boundary migration in alloys of high solute content (1–5wt%Mg), and a comparison between the in situ and bulk experiments is made. The relationship between boundary velocity and driving pressure was found to be linear in all cases, and the activation energies for boundary migration were higher than those controlled by lattice diffusion of the solutes at higher solute concentrations.

© 2011 Elsevier B.V. All rights reserved.

1. Introduction

It has long been recognised that solute elements segregate to grain boundaries in order to lower the energy and that during boundary migration, the solute atmosphere diffuses with, but lags behind the boundary, resulting in a dragging pressure on the boundary. Even very small additions of solute elements to high purity metals can drastically reduce the mobility (M) of boundaries, e.g. [1–3]. In order to understand and control the processes of recovery, recrystallization and grain growth during thermomechanical processing it is important that the effects of solute drag are quantified. Although the general effects of solute on boundary migration have long been known, quantitative measurements have been made only on a limited number of systems. Careful research carried out on curvature-driven boundary migration in bi-crystals of known orientation has clearly demonstrated the effects of boundary character, orientation and purity on boundary mobility, particularly at high temperature [4–8]. However, recent work [9] has suggested that there may be differences in boundary behaviour during recrystallization and curvature-driven growth, which could be attributed to differences in the type and magnitude of the driving pressure, the lower temperatures involved and the fact that a recrystallization front generally comprises small areas of boundary of different character, rather than a single boundary type.

The prime reason for the present work was to assess the effect of the solutes of the type and quantity typically found in industrial aluminium alloys on boundary migration during recrystallization. Such a study should provide some of the measurements of boundary mobilities which are required as inputs to models of recrystallization, e.g. [10–13]. It should however be emphasised that it is much more difficult to interpret the results of such an investigation in terms of the fundamental physics of boundary migration than is the case for the fundamental studies on curvature driven growth referred to above, in which only trace additions of solute were made.

2. Experimental

2.1. Materials

High purity (HP) aluminium and a series of high purity based single-phase aluminium alloys were investigated in this study. The compositions of the alloys are given in Table 1.

2.2. Specimen preparation

Cylindrical single crystals of size Ø16 × 100–140 mm were grown from the melt in a graphite mould under a vacuum of 10⁻³ Pa from 7 mm diameter cylindrical seed crystals of (1 1 0) orientation, and the crystal orientations were determined from X-ray diffraction to an accuracy of ±1°. The crystals were carefully cut and polished into Goss-oriented (1 1 0) (0 0 1) rectangular specimens of dimensions (4.5, 6.0, 10) mm × 10 mm × 15 mm and then given the homogenisation heat-treated specified in Table 2. The crystals were cooled after homogenisation to 300 °C at a rate of 10⁻³ °C h⁻¹ and then water quenched, except the high purity Al, which was cooled to room temperature at 10⁻³ °C h⁻¹.

The rectangular single crystals of Goss orientation were deformed in plane strain compression on an Instron-4405 test machine using a stainless steel channel-die. The deformation was carried out at room temperature and at a strain rate of 10⁻³ s⁻¹ for all materials to reductions (strain): 30% (0.36), 50% (0.69) and 70% (1.2). In order to reduce friction, PTFE tape, which was replaced at an increment of ~20% reduction, was used as a lubricant.
Table 1
Compositions of the alloys.

<table>
<thead>
<tr>
<th>Alloy</th>
<th>Si</th>
<th>Fe</th>
<th>Cu</th>
<th>Mn</th>
<th>Mg</th>
<th>Zn</th>
<th>Ti</th>
<th>B</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>HP–Al</td>
<td>&lt;.01</td>
<td>.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td></td>
<td></td>
<td>Bal</td>
</tr>
<tr>
<td>Al–0.05Si</td>
<td>.05</td>
<td>&lt;.01</td>
<td>.001</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>Bal</td>
</tr>
<tr>
<td>Al–0.05Mn</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>.01</td>
<td>.001</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>Bal</td>
</tr>
<tr>
<td>Al–0.3Mn</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>.30</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>Bal</td>
</tr>
<tr>
<td>Al–0.1Mg</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>Bal</td>
</tr>
<tr>
<td>Al–1Mg</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>Bal</td>
</tr>
<tr>
<td>Al–3Mg</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>.5</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>Bal</td>
</tr>
<tr>
<td>Al–5Mg</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>.5</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>&lt;.01</td>
<td>Bal</td>
</tr>
</tbody>
</table>

Bold values in Table 1 represent the amount (in weight percentage) of the purposely added individual solute element in the corresponding binary aluminium alloy.

Disc specimens were spark-cut from the deformed single crystals, with the plane of the disc parallel to the compression (rolling) plane. The disc specimens were deeply etched to remove any damage from the spark erosion, and mechanically polished and electropolished to a final size of 0.3 mm × 1 mm.

2.3. In situ annealing experiments

The in situ annealing experiments were carried out using similar procedures to those of an earlier investigation [9]. A single fine scratch was initially made across a disc sample, which was mounted on a hot stage in a CAMSCAN MX4000 field emission gun scanning electron microscope (FEGSEM). The disk was then heated to a temperature of 270–280 °C and held for ~10 min to promote recovery of the dislocations to form subgrain structures. The disk was then heated to the required temperature (between 300 °C and 420 °C) and isothermally annealed. Recrystallization was initiated at the scratch (Fig. 1a), and the experiment was continued until grains had grown a distance of ~500 μm from the scratch.

The partly recrystallized disk was then cooled to room temperature and EBSD mapping was carried out using an HKL Channel EBSD system. In some cases where the size of recrystallized grains was suitable for further annealing, the disk was annealed again at a different (generally lower) temperature and measurements were made on the same boundaries as those at the previous higher temperature. After annealing at the second temperature, the subgrain size and misorientation in the recovered matrix were again measured. During in situ annealing, the microstructural evolution was monitored using backscattered electron imaging. A U-matic video recorder was used to record the microstructural changes in samples where boundary migration was too rapid for normal image recording.

2.4. Microstructural characterisation and EBSD data analysis

The rate of growth of the recrystallizing grains was measured in the rolling plane. After the first stages of growth (∼50 μm) in which the recrystallization was influenced by the deformation close to the scratch, the boundary velocity was found to remain almost constant, although a slight decrease due to recovery in the matrix was sometimes observed [9].

Because all the recrystallized grains originated close to the surface at the scratch, it is reasonable to assume that measurements of boundary velocity in the disc plane

Table 2
The homogenisation treatment of the single crystals.

<table>
<thead>
<tr>
<th></th>
<th>HP–Al</th>
<th>Al–0.05Mn</th>
<th>Al–0.3Mn</th>
<th>Al–0.05Si</th>
<th>Al–0.1Mg</th>
<th>Al–1Mg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (°C)</td>
<td>630</td>
<td>600</td>
<td>580</td>
<td>580</td>
<td>600</td>
<td>580</td>
</tr>
<tr>
<td>Time (h)</td>
<td>4</td>
<td>5</td>
<td>12</td>
<td>4</td>
<td>5</td>
<td>12</td>
</tr>
</tbody>
</table>
are representative of the true boundary velocity. In order to check this assumption and to assess the influence of the free surface on the recrystallization, partly recrystallized samples were sectioned normal to the disc plane and metallographically examined. In most cases, the migrating boundary was approximately perpendicular to the specimen surface as shown in Fig. 2, indicating that the free surface had little influence on the boundary migration.

The EBSD data in the form of orientation maps (e.g. Fig. 1b) were analysed using the post-processing software, VMAP [14]. The subgrain size and misorientation in the recovered matrix were calculated, and from these, the driving pressures \( P \) for recrystallization was calculated as discussed in Section 3.4. Correlation of the driving pressures with the boundary velocities \( V \) measured just before the in situ annealing was stopped enabled the boundary velocities \( M \) to be determined from the relationship:

\[
V = MP
\]

(1)

The crystallographic relationships between recrystallized grains and the recovered matrix were also determined from the EBSD data.

2.5. Grain growth experiments on bulk samples

It was of interest to measure the boundary mobilities of alloys containing large solute concentrations, and Al–Mg, which is of industrial importance, is an ideal alloy system. However, it was not possible to carry out in situ recrystallization experiments on alloys with larger concentrations of aluminium than 1%. This was mainly due to the difficulty in determining accurately the driving pressures for recrystallization from the deformed and recovered microstructures in alloys containing larger concentrations of magnesium, and in which well-recovered subgrain structures may not be formed before recrystallization. It was also thought that the evaporation of magnesium at the surface via bulk or grain boundary diffusion might result in surface artefacts.

Therefore boundary migration in these alloys was investigated by measuring grain growth rates in polycrystals of Al–3%Mg and Al–5%Mg in addition to the Al–12%Mg alloy used for the in situ experiments. By using the Al–15%Mg alloy for both types of experiment, it was also possible to compare results from the two methods.

The 50 mm thick cast ingots were homogenised at 500 °C and slow cooled at 10 °C h\(^{-1}\) to room temperature. They were then hot rolled to 20 mm slab and annealed for 2 h at 360 °C for Al–1%Mg, 400 °C for Al–3%Mg and 440 °C for Al–5%Mg respectively. The hot rolled and annealed slabs were then cold rolled to a reduction of 60%, and annealed at 360 °C for 1 h. The average grain sizes were Al–1%Mg: 64 μm, Al–3%Mg: 92.1 μm, Al–5%Mg: 58.7 μm.

For the grain growth experiments, samples were annealed in air at temperatures between 300 °C and 450 °C. The annealed samples were sectioned in the ND–RD plane, metallographically prepared and examined in the FEGSEM using channelling contrast imaging and EBSD. The grain size, misorientation and their distributions were determined from the reconstructed EBSD data using VMAP. Grain sizes were determined from the grain area as an equivalent circle diameter (ECD), which, for an equiaxed grain structure is 0.816 of the diameter of spherical grains.

3. Results of in situ experiments

3.1. General features of the annealing

The Goss orientation is stable during channel die plane strain compression [15] and the resultant subgrain structure in which there are neither short range nor long-range orientation gradients and therefore no sites for originating recrystallization, provides an ideal matrix into which the recrystallized grains originating at the scratch, can grow. The microstructure in the deformed and recovered matrix was a uniform subgrain structure, for details see [15,16], and the range of microstructural parameters of the various recovered materials is given in Table 3.

The basic features of recrystallization and grain boundary migration were found to be similar for all the materials, and are typified by those shown in Fig. 1. As many aspects of the recrystallization behaviour are qualitatively similar to those reported in an earlier investigation on Al–0.05%S [9], we will not discuss them in detail, but will concentrate on the quantitative effects of the alloying elements.

Recrystallization originated along the scratch. The initial grains were found to have a reasonably random orientation relationship with the matrix, and the distribution of misorientation axes for the specimen of Fig. 1 is shown in Fig. 1c. However, a few grains are seen to have grown more rapidly and anisotropically.

3.2. Grain shape

The fast growing grains appeared to be lenticular, and this was confirmed by microstructural examination after progressive removal of the surface layers. The fastest growth direction of the rapidly growing grains was found to be perpendicular to the axis of misorientation between the grains and the recovered matrix, showing that the most mobile boundaries were of tilt character, whereas...
the twist boundaries along the broad faces of the grains were the least mobile. For those grains whose rotation axes were approximately parallel to the sample surface, both the fastest and slowest growth directions are parallel to the sample surface and therefore measurement of the grain aspect ratio gives a good measure of the anisotropy of the boundary growth rate. As shown in Table 3, the growth anisotropy ratio was found to be ∼10 and to be almost independent of the solute type and concentration. However, it may be seen that there is some small increase with increasing strain for the alloy crystals, and a larger effect for the high purity aluminium.

3.3. Orientation dependence of growth

The fast growing grains in all the materials were found to have orientation relationships with the recovered matrix close to 40°(1 1 1) and no other fast growing orientation relationships were detected except for the HP–Al in which the fastest growing grains had a 28–38°(1 0 0) relationship to the recovered matrix at recrystallization temperatures below 150 °C. Fig. 3a shows the dependence of the boundary velocity of the fast growing grains on the misorientation angle to the matrix for grains of misorientation axes <15° from (1 1 1). Fig. 3b shows the dependence of velocity on the deviation of the misorientation axis from (1 1 1), for grains whose misorientation to the matrix is 35–45°. In Fig. 3, the boundary velocity is expressed as a ratio of the velocity of a particular boundary relative to that of the fastest growing grain in the sample. Note that the measurements of the 28–38°(1 0 0) misoriented grains found at annealing temperatures below 150 °C in high purity aluminium are excluded from Fig. 3, but will be discussed elsewhere. Fig. 3 shows that although the maximum boundary mobility occurs close to a 40° rotation about a (1 1 1) axis as has commonly been observed during recrystallization [9,17,18], the peaks are very broad, and Table 5 shows the half-height half-widths of the mobility peaks for the various materials.

The half widths of the mobility peaks as a function of angular deviation (Fig. 3a) are seen to be ∼1°, and generally similar for all materials. Although there is scatter in the data, some trends may be seen. The Al–0.3Mn peak is the narrowest (5.1°) and that of the high purity aluminium the widest (11.2°), with those of the other alloys being intermediate and similar. The half widths of the mobility peaks as a function of axis deviation from (1 1 1) (Fig. 3b) are also ∼1°. However in this case, the Al–0.3Mn peak is broadest (13.4°) and that of the high purity aluminium is sharpest (7.3°).

The broad mobility peaks of Fig. 3 are in contrast to the very sharp peaks (<1°) found for boundaries moving by curvature driven growth, e.g. [3,7]. However, the broad peaks are similar to those found in an earlier investigation of recrystallization [9], where it was suggested that they were attributable to the effect of boundary excess volume rather than to the crystallography of high coincidence boundaries.

3.4. Grain boundary velocity and mobility

The velocities of the fastest growing grains in their fast growing direction (i.e. the tilt boundaries of the ∼40°(1 1 1) misoriented grains) were measured from the in situ annealing experiments for various driving pressures and temperatures as described earlier. The driving pressure (Pd) is provided by the recovered subgrains, and is given by [9]

\[ P_d = \frac{2\gamma_m}{L} \frac{\theta}{\theta_m} \left( 1 - \ln \frac{\theta}{\theta_m} \right) \]  

(2)

where \( \gamma_m \) is the energy of a high angle grain boundary (taken as 0.324 J m\(^{-2}\) [19]), \( \theta \) is the low angle misorientation, \( \theta_m \) is the misorientation above which the boundary energy become independent of misorientation (taken as 15°) and \( L \) is the mean linear sub grain intercept. Such a relationship, which is based on the Read–Shockley relationship, is valid only if the low angle boundaries are well recovered (i.e. not cell walls), and if the dislocation density within the subgrains is low, and this has been verified.

The migrating boundaries have a curvature (R), and this provides a retarding pressure (\( P_C \)) given, e.g. [2] by

\[ P_C = \frac{2\gamma_m}{R} \]  

(3)

and therefore the resultant pressure on the boundary (P) is \( P = P_0 - P_C \). P\(_{0w}\) was calculated from Eq. (2) using values of \( \theta \) and \( L \) determined from EBSD of the unrecrystallized microstructure after the in situ annealing, and \( P_C \) was calculated from the measured curvatures of the growing grains. \( P_C \) was found to be typically ∼0.02 MPa and to vary only slightly during the growth of the grains.

The variation of boundary velocity with temperature and net driving pressure (P) for the various materials is shown in Fig. 4. The data show some scatter, but give reasonable fits to a straight line as predicted by Eq. (1). As was found in a previous investigation [9], there appears to be a small threshold pressure (∼0.02 MPa) for migration which varies only slightly with material and temperature, and whose origin is unclear. It could be associated with interaction of the migrating boundaries with the surface by grooving or oxide film formation, although as shown in Fig. 2, there is little evidence of such interaction.

Table 3
Typical microstructural parameters for the recovered specimens.

<table>
<thead>
<tr>
<th></th>
<th>Al–(N)</th>
<th>Al–0.05Si</th>
<th>Al–0.05Mn</th>
<th>Al–0.3Mn</th>
<th>Al–0.05Si</th>
<th>Al–0.3Mn</th>
<th>Al–0.1Mg</th>
<th>Al–1Mg</th>
</tr>
</thead>
<tbody>
<tr>
<td>True strain</td>
<td>0.36–1.4</td>
<td>0.36–1.2</td>
<td>0.36–1.2</td>
<td>0.36–1.3</td>
<td>0.36–1.2</td>
<td>0.36–1.2</td>
<td>0.36–1.2</td>
<td></td>
</tr>
<tr>
<td>Subgrain size (µm) (mean intercept)</td>
<td>1.8–4.8</td>
<td>2.4–4.3</td>
<td>2.1–4.2</td>
<td>1.5–3.4</td>
<td>1.5–3.78</td>
<td>1.5–4.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mean subgrain misorientation (°)</td>
<td>1.5–2.5</td>
<td>1.7–2.6</td>
<td>1.5–2.8</td>
<td>1.8–2.9</td>
<td>1.6–2.75</td>
<td>1.6–2.6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Orientation spread (°)</td>
<td>~10</td>
<td>~8</td>
<td>~9</td>
<td>~11</td>
<td>~7</td>
<td>~9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Net driving pressure (MPa)</td>
<td>0.03–0.26</td>
<td>0.01–0.13</td>
<td>0.03–0.21</td>
<td>0.035–0.3</td>
<td>0.038–0.2</td>
<td>0.035–0.2</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 4
The average aspect ratio of fast-growing grains.

<table>
<thead>
<tr>
<th>Reduction (%)</th>
<th>Aspect ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>HP–Al</td>
</tr>
<tr>
<td>30</td>
<td>8.8</td>
</tr>
<tr>
<td>50</td>
<td>11.3</td>
</tr>
<tr>
<td>70</td>
<td>14.7</td>
</tr>
<tr>
<td>Average</td>
<td>11.6</td>
</tr>
</tbody>
</table>
Fig. 4. The correlation between boundary velocity and driving pressure: (a) HP–Al, (b) Al–0.05Si, (c) Al–0.1Mg, (d) Al–1Mg, (e) Al0.05Mn and (f) Al–0.3Mn.

The boundary mobilities were calculated from Eq. (1), using the data from Fig. 4, and the effect of temperature on mobility is shown in Fig. 5. It is seen that all the solute elements have strongly reduced the boundary mobilities in the alloys and the mobility values for the alloys are relatively similar compared to the large differences to the mobilities of the high purity aluminium. However, if the mobilities of the alloys alone are plotted on an expanded scale as in Fig. 5b, differences between the alloys are clearly seen. Fig. 5 shows that
the variation of the mobilities with temperature is a reasonable fit to the equation

\[ M = M_0 \exp \left( \frac{-Q}{RT} \right) \]  

(4)

and the calculated activation energies (Q) and the pre-exponential term (M0) for each material are listed in Table 6.

4. Results of bulk grain growth experiments

For the reasons discussed in Section 2.5, boundary migration could not be investigated by in situ annealing in alloys of high magnesium content. However, some information could be obtained from bulk grain growth experiments, and for the Al–1%Mg alloy, both techniques were employed for comparison.

![Fig. 5](image1.png)  
**Fig. 5.** Temperature dependence of mobility, (a) all data including those of 28–38° (100) boundaries in the HP–Al and (b) data only for the alloys.

![Fig. 6](image2.png)  
**Fig. 6.** A typical misorientation distribution in the Al–Mg alloys used for grain growth experiment. The data are from Al–5%Mg annealed at 400 °C for 1 h.

4.1. Grain growth kinetics and boundary mobility

The material processing routes (Section 2.5) produced polycrystals with weak textures which were similar in all cases. The data are summarised in Table 6, where it is seen that the only significant texture component is S (123)634. The distribution of boundary angles was similar in all cases, and an example is shown in Fig. 6 where it can be seen that the distribution is rather similar to that predicted for a randomly oriented grain assembly [20], but with a slightly larger frequency of boundaries with misorientations between ~10° and 20°.

Grain growth was observed in all the bulk annealed samples. The grain structure remained equiaxed and no abnormal growth was detected. As seen from Table 7, there were no significant changes to the texture or grain boundary distribution during grain growth, and the standard deviation of the grain size relative to the mean grain size remained almost constant. The results discussed in Section 3 referred to the migration rates of boundaries close to the 40° (111) or Σ7 relationship. In the bulk annealed samples, the number of such boundaries was small and the length percentages of Σ7 boundaries (defined according to the Brandon criterion) was between 0.6% and 0.8%, with no significant change during grain growth.

The isothermal grain growth kinetics are shown in Fig. 7. The good fit of the data to straight lines indicates that the variation of mean grain size (D̄) with time (t) follows the well known relationship, see e.g. [2,3]

\[ D^2 - D_0^2 = kt \]  

(5)

where k is a constant equal to αMy, where α ~ 2 [2].

It is interesting that this relationship, which is predicted for an ideal grain assembly (equiaxed grains with identical
boundary energies in a single-phase alloy) [21] is often found not to be obeyed [2]. However, as the grain growth kinetics in the present investigation clearly follow Eq. (5), the growth rates may be used to determine the boundary mobilities using this equation and the data of Fig. 7. The effect of temperature on the mobility of boundaries in the Al–Mg alloys is shown in Fig. 8, where it may be seen that the data are a good fit to Eq. (4). The calculated activation energies (Q) and the pre-exponential term (M₀) for the materials are shown in Table 6.

5. Discussion

The boundary velocities were found to be proportional to the driving pressures for all the conditions investigated, confirming the validity of Eq. (1) under the range of conditions investigated. This result supports the recent Monte Carlo simulation outcomes that grain boundary mobility is independent of the driving force under certain conditions [22,23] and casts doubts on the widely accepted solute drag theory [24–26], which implies that grain boundary mobility is a function of driving force. In addition, there is no evidence in the materials and conditions of the present investigation of a transition to high boundary velocity associated with the breakaway of a boundary from its solute atmosphere predicted by the solute drag theory. The driving pressures and annealing temperatures in the present work are typical of those encountered during recrystallization of aluminum, and the type and concentration of solutes are similar to some industrial alloys. This suggests that during the recrystallization of most aluminum alloys, boundary breakaway (if there is any) is unlikely to play a significant role.

5.1. Comparison of in situ and bulk experiments

Fig. 8 and Table 6 show that for the 1%Mg alloy, for which the boundary mobilities were measured by both in situ annealing and bulk grain growth experiments, there is a large difference in the mobilities. Although the activation energies are similar, the mobilities at any temperature measured from the in situ experiments are larger by a factor of ~10. The reason for this is that the in situ experiments measure the mobilities of tilt boundaries with orientation relationships close to 40°[1 1 1] to the recovered matrix (M₅₋₂), whereas the grain growth experiments measure the mobilities of “random” boundaries (Mₐ). Previous measurements of the ratio M₅₋₂/Mₐ from in situ [9] or bulk [16] experiments show it to be ~10 and to be close to the aspect ratio of the 40°[1 1 1] platelet grains. As seen from Table 4, the average value of the aspect ratio for the 1%Mg alloy is 9.5, and if the in situ measurements in Fig. 8 are divided by this factor so as to obtain the mobility of “random” boundaries, the mobilities obtained by in situ and bulk methods are comparable as shown by the crosses in Fig. 8.

5.2. Effect of temperature on mobility

The behaviour of the high purity aluminum alloy was quite different to the alloys, and the boundary mobility was not only much larger, but showed two regimes, both of low activation energy, in which boundaries of different type grew most rapidly. The mobilities of boundaries in the alloys appear to be controlled by the same thermally activated process throughout the temperature range investigated. It is seen from Table 6 that the reduction of mobility in the alloys with increasing solute is not due to a decrease in M₀ but to an increase of both M₀ and Q, and as a result, the effect of solutes on mobility is less at higher temperatures. This is in agreement with the experiments of Gordon and Vandermeer [27] on Al–Cu alloys. Recent work of the Aachen group [3,7] has clearly demonstrated that for boundaries of a particular structure, M₀ and Q are often related by the relationship

\[ Q = \alpha \ln(M_0) + \beta \]  

(6)

where α and β are constants. The boundary mobilities measured in the present work (Table 6), apart from the Al–0.3Mn alloy, are in agreement with such a relationship, with \( \alpha = 6.7 \) and \( \beta = 119 \text{ kJ mol}^{-1} \). This coupling of M₀ and Q, which is often referred to as the “compensation effect”, can be explained in terms of theories of thermally activated boundary migration [3,7,28].

### Table 6

Parameters of grain boundary migration obtained in this investigation.

<table>
<thead>
<tr>
<th>Material</th>
<th>Driving pressure (MPa)</th>
<th>Temperature (°C)</th>
<th>Orientation (±10°)</th>
<th>Q (kJ mol⁻¹)</th>
<th>M₀ (M⁺⁻¹ s⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HP–Al</td>
<td>0.03–0.15</td>
<td>50–170</td>
<td>28–38/⟨1 0 0⟩</td>
<td>66</td>
<td>0.0022</td>
</tr>
<tr>
<td>Al(5N)</td>
<td>0.03–0.15</td>
<td>100–340</td>
<td>40/⟨1 1 1⟩</td>
<td>87</td>
<td>0.15</td>
</tr>
<tr>
<td>Al–0.05Si</td>
<td>0.01–0.12</td>
<td>205–400</td>
<td>40/⟨1 1 1⟩</td>
<td>151</td>
<td>1.84</td>
</tr>
<tr>
<td>Al–0.1Mg</td>
<td>0.05–0.23</td>
<td>300–370</td>
<td>40/⟨1 1 1⟩</td>
<td>136</td>
<td>3.16</td>
</tr>
<tr>
<td>Al–1Mg</td>
<td>0.05–0.2</td>
<td>300–400</td>
<td>40/⟨1 1 1⟩</td>
<td>171</td>
<td>376</td>
</tr>
<tr>
<td>Al–1Mg*</td>
<td>0.0032–0.016</td>
<td>300–450</td>
<td>Random</td>
<td>170</td>
<td>367</td>
</tr>
<tr>
<td>Al–3Mg*</td>
<td>0.0034–0.012</td>
<td>300–450</td>
<td>Random</td>
<td>189</td>
<td>3333</td>
</tr>
<tr>
<td>Al–5Mg*</td>
<td>0.0052–0.018</td>
<td>300–450</td>
<td>Random</td>
<td>196</td>
<td>3715</td>
</tr>
<tr>
<td>Al–0.05Mn</td>
<td>0.05–0.24</td>
<td>300–400</td>
<td>40/⟨1 1 1⟩</td>
<td>175</td>
<td>44355</td>
</tr>
<tr>
<td>Al–0.3Mn</td>
<td>0.05–0.32</td>
<td>300–400</td>
<td>40/⟨1 1 1⟩</td>
<td>212</td>
<td>37731279</td>
</tr>
</tbody>
</table>

* Measured by grain growth during bulk annealing of recrystallized structures.

### Table 7

The initial microstructures and textures of the Al–Mg alloys used for grain growth experiments, compared with those annealed at 400°C for 30 min (Al–1Mg), 120 min (Al–3Mg) and 180 min (Al–5Mg) when the grain sizes are approximately double the initial ones.

<table>
<thead>
<tr>
<th>Material</th>
<th>Initial</th>
<th>Annealed</th>
<th>Initial</th>
<th>Annealed</th>
<th>Initial</th>
<th>Annealed</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al–1Mg</td>
<td>64 (0.55)</td>
<td>33.2</td>
<td>77.6</td>
<td>5.4</td>
<td>10.2</td>
<td></td>
</tr>
<tr>
<td>Al–3Mg</td>
<td>151 (0.59)</td>
<td>33.4</td>
<td>78.4</td>
<td>5.1</td>
<td>9.4</td>
<td></td>
</tr>
<tr>
<td>Al–5Mg</td>
<td>92 (0.54)</td>
<td>33.8</td>
<td>80.5</td>
<td>6.3</td>
<td>8.9</td>
<td></td>
</tr>
<tr>
<td></td>
<td>165 (0.51)</td>
<td>34</td>
<td>81.2</td>
<td>5.5</td>
<td>9.5</td>
<td></td>
</tr>
<tr>
<td></td>
<td>58 (0.48)</td>
<td>35.1</td>
<td>83.3</td>
<td>6.7</td>
<td>10.7</td>
<td></td>
</tr>
<tr>
<td></td>
<td>115 (0.51)</td>
<td>35.5</td>
<td>85.4</td>
<td>6.2</td>
<td>10.3</td>
<td></td>
</tr>
</tbody>
</table>

% material within 15° of important texture components (no other components were >5%)

<table>
<thead>
<tr>
<th>Brass</th>
<th>$s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.4</td>
<td>10.2</td>
</tr>
<tr>
<td>5.1</td>
<td>9.4</td>
</tr>
<tr>
<td>6.3</td>
<td>8.9</td>
</tr>
<tr>
<td>5.5</td>
<td>9.5</td>
</tr>
<tr>
<td>6.7</td>
<td>10.7</td>
</tr>
<tr>
<td>6.2</td>
<td>10.3</td>
</tr>
</tbody>
</table>
Fig. 7. Isothermal grain growth kinetics of Al–Mg polycrystals plotted according to Eq. (4): (a) Al–1Mg, (b) Al–3Mg, (c) Al–5Mg.

5.3. Effect of magnesium concentration on boundary mobility

Due to the limited data available, a direct correlation between solute concentration and mobility could only be made for Al–Mg alloys. The data from the Al–Mg alloys, which is shown in Fig. 9, shows that the relationship between mobility ($M$) and concentration ($X$) at a given temperature can be described by the relationship

$$\ln M = aX + b$$

where $a = 0.7332$ (300°C), 0.5989 (350°C) and 0.05512 (400°C), $b = 29.08$ (300°C), 26.267 (350°C) and 23.95 (400°C) respectively.

Fully quantitative theories of the effects of solutes on the mobility of grain boundaries have not yet been developed. For the case of boundaries whose migration rate is controlled by the diffusion of a solute atmosphere, it is predicted [3,24–27] that

$$M = \frac{q}{X}$$

where $q$ is a constant and relationships close to this have been found for metals containing trace additions of solute, see [2,3,28], and in particular, by the work of Gordon and Vandermeer [26]. The present results showed much larger reductions in mobility with increasing magnesium concentration as shown in Fig. 9. The calculated activation energies at high magnesium concentrations (>1%) are much larger than the values obtained from lattice diffusion controlled

Fig. 8. Temperature dependence of mobility in Al–Mg alloys. Data from bulk grain growth experiments are represented by solid shapes and the in situ results by open shapes. The crosses represent the mobilities of “random” boundaries in Al–1Mg calculated from the in situ data as discussed in the text.

Fig. 9. Grain boundary mobility (random boundaries) as a function of the concentration of Mg in Al–Mg alloys.
migration process [29,30]. Unfortunately, there is little experimental data with which the current results can be compared, as most investigations of the effects on solute mobilities of boundaries in aluminium have considered only trace additions.

5.4. The effect of different solutes on boundary mobility

The in-situ annealing experiments measured the mobilities of tilt boundaries close to 40° (1 1 1) for a number of solutes: silicon, magnesium and manganese, over a limited range of concentrations. The migration of boundaries under the influence of solute drag is expected to be affected by a number of solute parameters, which will depend on the specific solute and solvent. In addition to the solute concentration which is discussed above, the interaction energy between solute and boundary, the shape of the solute profile and the diffusivity of the solute atoms are all predicted to be important parameters in determining boundary mobility [3,23,31,32]. The boundary character is also particularly important, and as shown first by the classic work of Aust and Rutter [4,5] the mobilities of boundaries with misorientations close to coincidence site relationships are less affected by solute than other boundaries. There is experimental evidence, see e.g. [3] that for trace additions of solute to aluminium, different elements may have greatly different effects on boundary mobility. However, even for very low solute concentrations it is difficult to quantitatively predict the effect of a specific solute on boundary mobility.

The present investigation is concerned with large solute additions to aluminium. Attempts to correlate boundary mobility with parameters such as solubility and solute diffusivity have not been successful, and, as shown in Fig. 10, for the in situ experiments, the boundary mobilities correlate reasonably well with solute concentration alone, suggesting that for these elements and concentrations, other factors are secondary.

6. Conclusions

1. In-situ recrystallization experiments on deformed aluminium crystals containing large solute additions have shown that the orientation dependence of tilt boundaries with misorientations close to, but deviating from 40° (1 1 1) is dependent on the nature of the solute.

2. Grain growth experiments have been used to measure the mobilities of "random" boundaries in a range of Al–Mg polycrystalline alloys. The results can be correlated with those from the in situ recrystallization studies.

3. A relationship between the activation energies and pre-exponential terms similar to that found for more dilute alloys has been found.

4. The mobility of boundaries in the Al–Mg alloys decreases more rapidly with solute concentration than the inverse relationship predicted by early theories.

5. The mobilities of alloys containing Mg, Si or Mn solutes were found to depend primarily on solute concentration and temperature, the specific solute type having limited influence.

Acknowledgements

The authors would like to acknowledge the financial support of the EPSRC for this project and Alcan International, Banbury, for the provision of materials, facilities and helpful discussions.

References