SEM observations of grain boundary mobility under thermal and plasticity effects

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Abstract. Grain growth in high purity aluminum was studied by in situ experiments in a SEM in order to follow grain boundary mobility under thermal and plasticity effects. After a first cold rolling stage followed by a first heat treatment, samples are submitted to one or two sequences of 3% plastic strain followed by an additional heat treatment. SEM observations allow following the resulting mobility for representative sets of grain boundaries with either low or large misorientations between differently oriented grain pairs. This paper reports on the quantitative investigation of the driving force contributions due, on the one hand, to grain boundary curvatures as measured from experimental observations and, on the other hand, to differences of dislocation densities between the adjacent grains of each examined boundary, as estimated from numerical simulations of polycrystalline plasticity. A “migration diagram” representing these two contributions is presented and discussed with regard to existing velocity laws and models.

1. Introduction

Grain boundary straining and mobility features are studied in polycrystals because they play an important role in the thermo-mechanical properties of metallic materials that is still far from being understood. In order to study grain boundaries experimentally, it is necessary to overcome the difficulty of accessing within a polycrystal several key parameters for a great number of boundaries, such as the plane of the boundary, its curvature and structure. Up to now, among the few well assessed characteristics, it has been shown that boundary migration was depending on the locally stored energy within the material [1], according to a velocity law that can be simply written as \( V = MP \). In this formula, \( V \) is the grain boundary velocity, \( M \) is the boundary mobility which depends on temperature and boundary misorientation, and \( P \) is the driving force necessary for the boundary mobility, which can be decomposed into two contributions: one is due to the stored energy during plastic strain and is simply proportional to the differential of dislocation densities \( \Delta \rho_{ij} \) between the adjacent \((i,j)\) grains of a \(i/j\) boundary, that we here denote \( E_{\Delta\rho} \) and the other one, associated with curvature, , is proportional to the grain boundary energy and will be denoted \( E_c \).

If the mobility of the grain boundaries is known to furthermore depend on the grain orientations and misorientations [2],[3], the way all these dependencies combine with each other is still a challenging point. Scanning Electron Microscope (SEM) techniques combined with electron backscattering diffraction (EBSD) [4] provides an efficient tool to study the migration of grain boundaries, although...
the 3D information (about curvature for instance) remains out of reach. In this study, also based on SEM+EBSD experimental examinations, polishing techniques have been used in order to at least verify that the grain dimensions in the sample depth were similar to the surface one. The estimates of the dislocation densities stored in the individual grains after a 3% plastic strain have been obtained from now classical plasticity models for polycrystals.

2. Experimental method

From a pure aluminum ingot characterized by an average grain size about 2 centimeters, a first sample was cold rolled to 85% reduction (from 15 millimeters to 2.25 millimeters) and a second one to 90% reduction (from 15 millimeters to 1.5 millimeters). For in situ annealing and further mechanical and microstructural characterization, both samples were then spark cut to the dimensions shown in figure 1. The final useful surface for observation has thus a length of 5 mm and a width of 2 mm. In order to obtain good images from the SEM, each sample was subjected to mechanical polishing with several silicon carbide papers from 1200 to 4000 mesh and to a final electrolytic polishing within a solution composed of 5% of perchloric acid, 65% of ethanol, 10% of 2-butoxyethanol and 15% of water.

After polishing, the 85% reduced sample (to be denoted as S85) was annealed at 400°C for 3 minutes and the 90% reduced sample (to be denoted as S90) at 350°C for 30 minutes [5]. Both were then characterized by electron backscattering diffraction (EBSD). Then, these samples were deformed in tension up to 3% plastic deformation, in order to introduce sufficient energy into the material for triggering grain boundary mobility during subsequent annealing. The S85 sample was annealed 5h at 400°C and the S90 sample 1h at 350°C. For the S85 sample, a second sequence of a 3% plastic straining followed by the same thermal treatment (5 hours at 400°C) was applied in situ inside the SEM. For the S90 sample, the first sequence of straining + heating (1 hour at 350°C) was performed in situ. All these treatments are presented in Table 1.

| Sample | Rolling step | T1 treatment (recrystallization) | 1st critical strain step | T2 treatment (grain growth) | 2nd critical strain step | T3 treatment (grain growth) |
|--------|--------------|----------------------------------|--------------------------|-----------------------------|--------------------------|-----------------------------|
| S85    | 85%          | 400°C 3 minutes                  | 3%                       | 400°C 5 hours               | 3%                       | 400°C 5h                    |
| S90    | 90%          | 350°C 30 minutes                 | 3%                       | 350°C 1 hour                | -                        | -                           |

For in situ testing, a tension micro machine system was used (see figure 2) [6]. A resistive furnace is placed below the sample for heating and the heating rate is assigned to be 10°C/s. Images of the sample surface were taken every five minutes in order to follow grain growth, and grain boundary displacement.
At the end of the test, the microstructure was analyzed by EBSD to quantify the grain growth and the change of the crystallographic texture. Additionally, the EBSD software was used to calculate the Schmidt factors of the possible slip systems. Then, it could be checked that the observed traces did correspond to the system(s) associated with maximal Schmidt factor.

The EBSD analyzes were performed using a scanning electron microscope with a field emission gun Zeiss Supra 40VP. Kikuchi diagrams were obtained by tilting the samples at 70° with respect to the incident electron beam. Data acquisition was performed using the software Nordif TSL-OIM data collection, with a step of 1 micrometer. Data processing was then done using the OIM data analysis software.

3. Data analysis

In order to examine the correlation between the driving force and the grain boundary motion, this driving force is simply estimated from a relation of the type \( P = \alpha E_c + \beta E_{\Delta \rho} \). \( E_c \) is the boundary energy resulting from the curvatures of the boundaries and is taken of the form \( 2\gamma R \), where \( \gamma = \gamma_m \theta / \theta_m (1 - \ln \theta / \theta_m) \) for a Low Angle Grain Boundary (LAGB) and \( \gamma = \gamma_m \) for a High Angle Grain Boundary (HAGB) and \( R \) is the curvature radius of the boundary. It is estimated locally through image processing. It most often corresponds to the maximal value observed for a given grain boundary. \( \gamma_m \) is taken as 0.324 J/m² [7]. \( E_{\Delta \rho} \) is the energy part which depends on the dislocation densities in the grain pair adjacent to the considered boundary. This energy part is given by the formulae \( E_{\Delta \rho} = 0.5\mu b^2 \Delta \rho \) where \( \mu \) is the shear modulus, equal to 25.4 GPa for aluminium, and \( b \) the Burgers vector, equal to 2.76 \( \times 10^{-10} \) m. \( \rho \) is the grain dislocation density and \( \Delta \rho = \rho_i - \rho_j \) is the difference for a \((i,j)\) grain boundary.

The dislocation density within the individual grains is calculated by using a viscoplastic “Relaxed Constraint” Taylor model which includes the single crystal constitutive law developed by Tabourot and al. [8] and further modified by Bacroix and Brenner to account for the observed anisotropy of the recovery processes [9]. This model allows the determination of the reorientation and the hardening of every plastically deformed crystal. The evolution of the dislocation density during straining is given by the equation

\[
\frac{d\rho^s}{dt} = \frac{1}{b} \left\{ \sqrt{\sum a^s i^s \rho^s} - 2 \sum \left( \frac{b^s i^s j^s}{\rho^s} \right) \rho_j \right\} |\gamma^s|, \]

in which \( \rho^s \) stands for the dislocation density and \( \gamma^s \) for the slip rate on the system \( s \). The hardening \( a^s i^s \) and recovery \( b^s i^s \) coefficients, as well as the parameter \( K \) are obtained through an identification procedure on available stress – strain tensile curves for aluminium single crystals [10]. These tests, as well as the experimental ones described earlier, are simulated by prescribing a uniaxial stress state to each crystal. The identified coefficients are gathered in Table 1. The two hardening and recovery matrices are characterized by only 5 distinct coefficients, which describe the interaction types between systems (from 1 to 5: self-hardening, collinear, coplanar, orthogonal and others) [8]. The initial dislocation density \( \rho_0^s \) is taken equal to \( 10^9 \) m⁻³ for all systems. It is worth noting that the parameters are not strictly the same for all single crystals, but two main classes can be distinguished: the <111> and <100> orientations (these indices represent the Miller indices of the tensile direction) which correspond to the simultaneous activation of at least 6 systems on one hand and the <123> and <112> orientations for which one or two systems only are simultaneously activated on the other hand.

| Orientation | \( K \) | \( a^1 \) | \( a^2 \) | \( a^3 \) | \( a^4 \) | \( a^5 \) | \( b^1 \) | \( b^2 \) | \( b^3 \) | \( b^4 \) | \( b^5 \) |
|-------------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|
| 111         | 90      | 0.4     | 0.5     | 0.6     | 0.6     | 0.5     | 5.0     | 0.5     | 1.0     | 4.0     | 0.5     |
| 100         | 95      | 0.3     | 0.5     | 0.6     | 0.6     | 0.5     | 5.0     | 0.5     | 0.5     | 5.0     | 0.5     |
| 112         | 105     | 0.2     | 0.4     | 0.6     | 0.6     | 0.35    | 1.5     | 0.0     | 0.0     | 3.0     | 0.0     |
| 123         | 105     | 0.25    | 0.4     | 0.6     | 0.6     | 0.35    | 1.5     | 0.0     | 0.0     | 3.0     | 0.0     |
4. Results
We first examine and report on the S85 sample. Figures 3, 4 and 5 represent respectively the sample surface after 85% rolling and T1 treatment, after the 2nd critical strain step, and after T3 treatment (Table 1). They show (according to the color convention of the reported reference triangle) three main orientations: the <111> orientation, and the <112> and <102> orientations which are close to <100>. Grain sizes of 200 to 500 µm are obtained. The grains are still elongated in the rolling direction. Figures 3, 4 and 5 summarize the grain boundary evolution due to the assigned strain + heat treatment sequences. In figure 5, the new shapes of the same grains after T3 treatment characterized by EBSD are superimposed to the grain contours prior to annealing (in red). We clearly observe the Strain Induced Boundary Migration (SIBM) of a part of the GBs present in the material. Furthermore, in most cases, we clearly see red grains growing at the expense of blue ones.

However, other orientations exist below the observed surface of the sample, which present smaller grain sizes and which can affect the growing sequence observed on the extreme surface. The microstructures characterized at 200 and 400 µm below the surface are thus presented in figures 6 and 7. It is observed that the dimension of the grains through the thickness of the sample is of same order of magnitude as the grain size on the surface with a larger number of small grains.

Figure 3. S85 sample surface after 85% rolling and T1 treatment.
Figure 4. S85 sample surface after T2 treatment + 3% strain.
Figure 5. S85 after final T3 in situ annealing with grain contours of figure 4 (red).

Figure 6. Microstructure at a depth of 200 µm with grain contours on the surface (red).
Figure 7. Microstructure at a depth of 400 µm with grain contours at depth of 200µm.
In all these figures, the grains have been numbered to follow the boundaries between adjacent pairs: the characteristics of these boundaries are collected in Table 2. For the S85 sample, 30 such boundaries have been followed, for which the misorientation ($\theta$), boundary displacement (L) and curvature have been extracted from the EBSD maps, whereas the dislocation density gradient has been calculated with the identified constitutive law. These calculations show a higher stored energy ($E_{\Delta \rho}$) for the $\langle 111 \rangle$ grains in which multiple slip occurs. The superposition of the inverse pole figure mapping before and after grain growth shows that these most deformed grains tend to shrink while the surrounding grains expand.

A similar examination is performed on the S90 sample. The number of examined boundaries in this case is 50. It is observed in Figures 8 and 9 that (i) the grains are globally smaller than in the S85 sample; (ii) the orientations are more on the $\langle 100 \rangle$ side of the reference triangle (only a few $\langle 111 \rangle$ oriented grains are present); it is also seen in Table 2 that the boundary curvatures are more marked and the difference of dislocation densities are in contrast much weaker than for the S85 sample. The grains with the largest growth are those (of white color as grain 70 or 116) oriented in the center of the reference triangle. About half of the collected data are reported in Table 2 to save room.

5. Discussion

By convention, a grain boundary displacement from a grain $i$ to a grain $j$ is always counted positive. Both the related curvature (the $E_c$ energy contribution) as well as the related differential of the dislocation density (the $E_{\Delta \rho}$ energy contribution) can either be positive or negative which yields four possibilities. Since the grain boundary motion is expected in the direction where the energy $P = \alpha E_c + \beta E_{\Delta \rho}$ is positive, only three of the four possibilities can a priori be associated with GB motion. If the
migration rate (or distance) is proportional to this energy, isovales \( \alpha E_c + \beta E_{\Delta \rho} = \text{cst} \) are straight lines parallel to a line \( \alpha E_c + \beta E_{\Delta \rho} = 0 \) passing through the origin of a \((E_c,E_{\Delta \rho})\) plot. It is also expected that the slope of these lines \( E_{\Delta \rho}/E_c = -\alpha/\beta \), is negative.

The «migration diagram» (an approximate of a velocity map) shown in figure 10 collects all the examined grain boundaries for the two analyzed samples on a \((E_c,E_{\Delta \rho})\) plot: each point in this diagram has been given a dimension proportional to the measured boundary displacement \(\Delta L\) after a same time interval \(\Delta t\).

This displacement would be an estimate of the velocity \(V\) if the displacement rate was constant, which is not the case. Nevertheless, it is clearly observed at first when plotting this displacement in terms of the two energy contributions, that there are no points in the negative “forbidden sector” \((E_c < 0,E_{\Delta \rho} < 0)\) as expected. It is further observed in the limits of the data uncertainty and dispersion, that: (i) most of the points are in the positive sector; (ii) the points with a large radius (most mobile boundaries) are farther from the diagram origin than the points with a small radius (weakly mobile boundaries); (iii) the dislocation density effect seems less marked than the curvature effect, at least for the S90 material for which the more isotropic distribution of grain orientations reduces the dislocation density gradient and the grain size is much smaller leading thus to higher curvatures.

We can also note, that if we try to plot a straight line which would represent the \(P = 0\) limit and place all measured points on the side of a positive driving force, only a few points remain below, and are thus associated with an overall negative driving force. These few cases need to be further examined in order to see if this is due only to data dispersion.

### Table 3: Characteristic of the examined GBs for both samples.

| Boundary | \(\theta\) | \(L(\mu m)\) | \(\Delta\rho/\rho_0\) | \(E_c(J/m^3)\) |
|----------|----------|---------------|----------------------|-----------------|
| **S85**  |          |               |                      |                 |
| \(B_{16,17}\) | 4 | 0 | 7.74E+02 | 9.43E+02 |
| \(B_{10,31}\) | 43 | 0 | 1.94E+02 | 3.13E+03 |
| \(B_{19,22}\) | 9 | 3 | 9.38E+02 | 5.89E+02 |
| \(B_{21,7}\) | 59 | 7 | 1.18E+03 | 2.59E+03 |
| \(B_{28,30}\) | 10 | 8 | 1.23E+03 | 1.51E+03 |
| \(B_{36,9}\) | 50 | 17 | -1.74E+03 | -1.01E+03 |
| \(B_{35,30}\) | 53 | 18 | 3.28E+02 | 2.60E+03 |
| \(B_{30,29}\) | 12.5 | 18 | 6.82E+01 | 6.72E+03 |
| \(B_{8,5}\) | 7 | 26 | 5.66E+01 | -2.69E+03 |
| \(B_{5,43}\) | 55 | 35 | 1.49E+03 | -4.32E+03 |
| \(B_{9,33}\) | 56 | 36 | 5.72E+02 | -2.82E+03 |
| \(B_{4,42}\) | 45 | 39 | 1.76E+03 | 5.18E+03 |
| \(B_{2,29}\) | 46 | 45 | 3.32E+02 | 4.97E+03 |
| \(B_{3,30}\) | 55 | 50 | 3.27E+02 | 3.59E+03 |
| \(B_{4,18}\) | 46 | 60 | 2.32E+03 | 2.25E+03 |
| \(B_{2,41}\) | 51 | 60 | 5.82E+02 | 6.22E+03 |
| \(B_{3,10}\) | 47 | 60 | -1.20E+03 | 1.80E+03 |
| \(B_{1,15}\) | 60 | 65 | 6.54E+02 | 3.29E+03 |

| Boundary | \(\theta\) | \(L(\mu m)\) | \(\Delta\rho/\rho_0\) | \(E_c(J/m^3)\) |
|----------|----------|---------------|----------------------|-----------------|
| **S90**  |          |               |                      |                 |
| \(B_{97,67}\) | 12 | 5 | 1.85E+02 | -1.10E+04 |
| \(B_{100,101}\) | 6 | 6 | 1.64E+02 | 8.29E+03 |
| \(B_{97,68}\) | 9 | 6 | 4.58E+01 | 9.07E+03 |
| \(B_{13,6}\) | 47 | 10 | 1.26E+01 | 7.56E+02 |
| \(B_{15,21}\) | 53 | 10.5 | 3.07E+01 | 1.26E+04 |
| \(B_{62,73}\) | 51 | 11 | -2.87E+02 | 6.21E+03 |
| \(B_{109,112}\) | 14 | 12 | -2.93E+02 | 1.33E+04 |
| \(B_{13,14}\) | 50 | 43 | 4.21E+01 | 1.26E+04 |
| \(B_{116,114}\) | 43 | 44 | 6.38E+01 | 2.78E+03 |
| \(B_{64,65}\) | 54 | 45 | -3.48E+01 | 1.89E+03 |
| \(B_{31,26}\) | 45 | 46 | -4.21E+01 | 3.70E+03 |
| \(B_{54,52}\) | 41 | 50 | -7.61E+01 | 1.06E+04 |
| \(B_{3,5}\) | 59 | 60 | -8.40E+01 | 4.57E+03 |
| \(B_{70,71}\) | 46 | 64 | -2.11E+02 | 6.87E+03 |
| \(B_{3,10}\) | 35 | 67 | 3.81E+01 | 1.70E+04 |
| \(B_{19,26}\) | 40 | 67 | -1.92E+02 | 4.86E+03 |
| \(B_{76,77}\) | 15 | 73 | 3.35E+02 | 1.79E+04 |
| \(B_{116,115}\) | 52 | 112 | 1.16E+02 | 2.13E+04 |
6. Conclusions

The performed SEM in situ examination of grain boundary migrations in aluminium polycrystals has been combined with numerical simulation of plastic straining in order to obtain boundary curvature measurements and dislocation density estimates. The method has been shown capable to yield a “migration diagram” from which the relative contributions to the boundary migration driving force, i.e. the curvature and density contributions can be compared. For more precision, it appears from these results that with an increased number of examined boundaries a quantitative analysis of the data is at hand. This work is on progress.

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