Quantitative measurements of small scaled grain sliding in ultra-fine grained Al–Zn alloys produced by friction stir processing

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1. Introduction

Grain boundary sliding (GBS) is an important mode of deformation in creep and superplasticity. Some of detailed reviews are reported in recent years [1–3]. Many efforts have been made over the past years to produce reliable and reproducible procedures for estimating the contribution of grain boundary sliding to the total strain. The GBS contributions may be determined directly by measuring offsets in marker lines on the specimen surface or from the displacement of adjacent grains perpendicular to the specimen surface.

In recent years nanocrystalline (nc) and ultrafine-grained (ufg) metals and alloys synthesized in different ways have been extensively investigated [4–6]. There is indirect evidence suggesting that grain boundary sliding may occur more easily in metals at room temperature in nc and ufg metals and this may lead to a high strain rate sensitivity and a reasonable level of GBS contribution [7,8]. In contrast to the superplastic and creep deformation at elevated temperatures, the grain boundary sliding occurs in nc and ufg metals and alloys at room temperature, showing that the ratio of the strain to the total strain due to GBS, δ, is not inhomogeneous plastic deformation over the gauge length. In addition, the width of marker lines scratched using diamond powders are far oversize for ufg and nc materials. Therefore, all determinations of δ obtained by the traditional experimental methods to estimate the contribution of GBS are not feasible.
It is known that GBS can be enhanced by grain boundary diffusion. It is the reason that Zn is chosen to add into an Al matrix in large quantities so as not to alter its FCC microstructure and to enhance the GB diffusion rate [9]. The available data for grain boundary diffusion of Zn in Al gives the value of $sD_\delta (300K) \approx 10^{-19} m^3 s^{-1}$ (15 wt.% Zn) [10] for a grain boundary thickness of $=0.5$ nm and a segregation factor of $s=1$. Grain boundary diffusion for Al [11], Mg [12], Cu [13] in an aluminum matrix at 300 K are $5 \times 10^{-28} m^3 s^{-1}$, $1.3 \times 10^{-24} m^3 s^{-1}$, $6.15 \times 10^{-26} m^3 s^{-1}$, which are respectively much smaller than Al-Zn alloys. The purpose of this paper is to present a new technique developed to measure GBS displacements and strains for samples deformed at room temperature using Focus Ion Beam machining. Also, an attempt to estimate the contribution of GBS to total strain in deformed fine-grained Al alloys is conducted. Three advantages of the proposed method are: firstly, the marker lines can be scratched exactly parallel to the tensile direction; secondly, the scratched markers with scaled divisions are used to evaluate the true strain in non-uniform and uniform strain fields over a short distance of less than 25 $\mu$m so that more precise estimation of the true contribution of sliding to total strain is possible; thirdly, the width of the marker line can be easily adjusted through controlling the FIB parameters on the ufg and nc materials. Thus, it is possible to estimate quantitatively the small contribution of GBS to the total strain at low temperature and/or room temperature.

2. Experimental Materials and Procedures

The starting materials used were commercially pure Al powders (99.7% purity, 325 mesh) and Zn powders (99.5% purity, 325 mesh). The premixed alloy powders, with Zn content of 15 wt.%, were cold compacted into $12 \times 20 \times 80$ mm billets in a steel die set, under a pressure of 225 MPa. First, a large pin (the diameters of the shoulder and probe of the tool were 16 and 6 mm, respectively) and high spindle rotation speed (1500 rpm) were employed to produce dense large cross section bulk material. Then, the smaller diameters of the shoulder and...
probe of the tool and lower spindle rotation speed were used to produce refined a grain size (~500 nm–1 μm and ~2 μm) [9]. Importantly, the processed samples and fixture were cooled down to room temperature when one processing operation parameter is changed to another in order to avoid accumulative heating during the friction stir passes. The tilt angle was maintained at approximately 3°. A detailed description of the friction stir process (FSP) set-up can be found in several published papers [9,14]. As shown in Fig. 1, the oxygen content of the starting powders and FSPed Al-15Zn samples (Al-15Zn-1, Al-15Zn-2 and Al-15Zn-3) was determined using an oxygen–nitrogen analyzer (HORIBA, EMGA-620W). It was also observed that the oxygen contents of all the Al-15Zn samples and starting powder were negligible. The tensile specimen was machined from the stir zone with the tensile axis parallel to the traveling direction. Multiple dog-bone shaped tensile specimens with a 6 mm gauge length, a 1.5 mm width, and 1.5 mm thickness were electro-discharge machined from the stir zone. The dog-bone samples were first surface polished to a mirror finish using silicon oxide with 20 nm particles, and then the surfaces were polished by Argon ions (Gatan Mode 683 Met-Etch Ion-Polisher) to remove surface defects, such as scratches, embedded polishing grit, and oxide layers. Then, the marker line and crossing line were scribed using FIB (Fig. 2). The width and depth of the scribed fine line were 50 nm and 60 nm, respectively, much finer than the mechanical scratches [15–17] and the distance between two crossing lines used for a reference was 25 μm. Measuring the true local strains and the offsets of marker lines on the surfaces of deformed specimen are crucial to estimate the contribution of GBS. Based on the definition of strain, the true local strains at different locations of the specimen after deformation were obtained by taking the distance between the selected crossing lines along the appropriate direction. It was assumed that the deformation along the thickness direction was the same as along the transverse direction. In each case, at least six 6×6 μm areas were analyzed and determination of the GBS component perpendicular to the specimen surface (the u component in Fig. 3) was made on these areas. For the observation of grain size, the grain boundary was etched by a GATAN Model 683 Met-Etch Ion-Polisher and then examined by scanning electron microscopy (JSM-6330TF). A computer program (Optimas-6.1) was used to determine the grain size and size distribution. The tensile tests were conducted using an Instron 5582 universal testing machine with initial strain rates of $1 \times 10^{-3}$ s$^{-1}$ at room temperature. For each

Fig. 4 – The XRD pattern of Al-15Zn with before and after FSP. The inset show the enlarged portion of the XRD patterns which the Al (420) peak of the Al and Al-15Zn samples.

Fig. 5 – Macrographs showing the homogeneous microstructure in the processed zone in FSP (a) Al-15Zn-1, (b) Al-15Zn-2, and (c) Al-15Zn-3 samples (samples were lightly etched).
specimen, more than 500 grains were measured. X-ray diffraction analysis (XRD) was performed with a Siemens D-5000 diffractometer operating with CuK$_\alpha$ radiation ($\lambda=0.15406$ nm). Samples were scanned between $20^\circ$–$90^\circ$ ($2\theta$) and $116^\circ$–$117.2^\circ$ using a step size of 0.02$^\circ$.

3. Results and Discussion

The X-ray diffraction patterns of Al+Zn powder samples and the friction stirred UFG Al-15Zn samples are plotted in Fig. 4. It clearly shows that peaks in the pattern of the friction stirred Al-15Zn sample are related to the aluminum single FCC phase. Such a result also indicates that all of the Zn added into the green sample had been dissolved into the aluminum matrix after FSP. The inset of Fig. 4 shows the (420) peaks of Al peaks in the patterns of the UFG Al-15Zn and UFG Al, were shifted to slightly higher angles with increased Zn content. It is commonly known that the atomic radius of Zn is smaller than that of Al ($r_{\text{Al}}=0.1432$ nm, $r_{\text{Zn}}=0.1332$ nm)[18], when some of the Al atoms in the lattice are replaced by Zn, the resultant Al (Zn)solid solution will have a smaller lattice parameter. Since solid-state friction stir processing does not result in solute loss by evaporation and segregation by solidification, solute

Fig. 6 – (a)(c)(e): SEM micrographs for the Al-15Zn-1, Al-15Zn-2 and Al-15Zn-3 samples, and (b)(d)(f): grain size distribution charts of the Al-15Zn-1 and Al-15Zn-2 and Al-15Zn-3 samples.
elements are homogeneously distributed in the processing zone. As shown in Fig. 5(a–c) show the optical micrographs of Al-15Zn-1, Al-15Zn-2 and Al-15Zn-3, revealing the FSP technique was very effective in creating a homogeneous microstructure.

In this experiment, the Al-15Zn samples were processed at the specific operation parameters [10]. Fig. 6(a)(c)(e) show the SEM micrographs of samples Al-15Zn-1, Al-15Zn-2 and Al-15Zn-3, revealing a well-defined equiaxed grain nature. In parallel, Fig. 6(b)(d)(f) show the grain size distribution with an average grain size of 480, 900 and 1750 nm, respectively. Friction stir processing (FSP) led to dynamic recrystallization in all conditions to achieve equiaxed grains with high-angled grain boundaries of more than 85% in volume fraction [19,20]. High-angled boundaries and fine grains are necessary in order to achieve GBS, and to increase contribution of GBS in the total strain [21].

The result of the strain rate change test, or jump test, for three samples (Al-15Zn-1, Al-15Zn-2 and Al-15Zn-3) at room temperature can be seen in Fig. 7(a), and tests were performed at the base strain rate of 10^{-4} and 10^{-3}s^{-1}. The strain rate sensitivity m was estimated using the relation

$$m = \frac{\partial \ln(\sigma)}{\partial \ln(\dot{\varepsilon})} = \frac{\ln(\sigma_2 / \sigma_1)}{\ln(\dot{\varepsilon}_2 / \dot{\varepsilon}_1)}$$

(1)

where $\sigma_2$ and $\sigma_1$ are the flow stresses corresponding to the strain rate $\dot{\varepsilon}_2$ and $\dot{\varepsilon}_1$, respectively. For the Al-15Zn-1 sample (grain size of 0.5 μm), the measured m-value was about 0.12 within the strain range of 5%–30%, which is five times more than that (m=0.022) for a pure Al sample with a similar grain size [22], while the measured m-value of Al-15Zn-2 sample (grain size of 1 μm) was about 0.047 which is also three times more than that (m=0.015) for a similar grain size pure Al sample [22]. However, it should be noted that the m-value obtained for the Al-15Zn-3 sample (grain size of 2 μm) was about the same as that reported for pure Al (m=0.011) with a similar grain size [22]. The increase of m-value with a decrease of grain size in the region of grain sizes less than 2 μm is plotted in Fig. 7(b) to compare with SRS for pure Al and Al alloys obtained from previous literature [22–27]. The m-value

$$m = \frac{\partial \ln(\sigma)}{\partial \ln(\dot{\varepsilon})} = \frac{\ln(\sigma_2 / \sigma_1)}{\ln(\dot{\varepsilon}_2 / \dot{\varepsilon}_1)}$$

Fig. 7 – (a) Strain-rate jump test between the strain rate of $\dot{\varepsilon} = 1 \times 10^{-3}s^{-1}$ and $\dot{\varepsilon} = 1 \times 10^{-4}s^{-1}$ for Al-15Zn samples (b) strain rate sensitivity as a function of $d^{-1/2}$, where $d$ is the average grain size. The reference and its first author corresponding to each data point are also given in the plot.

Fig. 8 – AES depth profiles showing the influence of the FIB on the resulting Ga and O concentration profile on (a) scribe line and (b) Al-15Zn matrix.
for Al-15Zn alloys is extraordinarily larger than that for conventional Al alloys.

To study the mechanisms of plastic deformation, the surface relief of the deformed samples was carefully analyzed by scanning electron microscopy (SEM). However, since the marker line and crossing line were scribed using FIB, it is necessary to consider whether the ion beam damage from FIB milling could lead to implantation of Ga, as well as other microstructural damage that can penetrate in the order of several tens of nanometers [28]. The Auger electron microscopy (AES) measurement using a Jeol Jamp-9500 Field Emission Auger Microprobe at an accelerating voltage of 10 kV, a beam diameter of 5 nm, and a beam current of 10 nA was carried out for reliable information on the depth profile of Ga concentration. Fig. 8 shows the AES depth profiles for the Al-15Zn sample. No implantation of Ga found was due to fast line etching rates.

Figs. 9-11 show the SEM surface topographies taken at different position where true strains were measured for the Al-15Zn-1, Al-15Zn-2 and Al-15Zn-3 pulled at a strain rate of $1 \times 10^{-3}$ s$^{-1}$. Typical offsets of marker lines of the local regions marked by the white rectangles on the deformed specimen in Al-15Zn-1 and their true strains of 5%, 13%, 21%, 40%, and 60%, respectively are shown in Fig. 9(a-e). The grains remained equiaxed in shape during deformation. A closer inspection revealed that the marker lines were sharply defined and exhibited apparent sliding offsets at many grain boundaries. The sliding offsets tended to increase with the elongation. Focusing on the region of small strains (5% and 13%), the occurrence of numerous small offsets, which took place when tiny marker lines were used, would not obscure the general trend in grain displacement. These results indicate that the method is capable of differentiating relatively small displacements and providing a quantitative measurement of the total sliding displacement. In the past, conventional methods [2,3] would have more difficulty to analyze sliding displacement at lower strains or non-uniform regimes on such fine-grained materials. Similar measurement for Al-15Zn-2 and Al-15Zn-3 from Figs. 10 and 11 are summarized in Table 1. The strain caused by GBS, $\varepsilon_{\text{gbs}}$, as characterized by Eq. (2) can be determined by measuring the sliding offset ($u$) combined with measurements of the mean linear intercept grain size ($\ell$) [23].

$$\varepsilon_{\text{gbs}} = \frac{\Psi}{\ell} \bar{u},$$  \hspace{1cm} (2)

where $\bar{u}$ is the average value of $u$, $\ell$ is the mean linear intercept grain size, and $\Psi$ is a geometrical constant, which is
taken as 0.8 [29]. Finally, the GBS contribution, \( \delta \), is determined in Eq. (3) [30]:

\[
\delta = \frac{\epsilon_{gbs}}{\epsilon_t};
\]

where \( \epsilon_t \) is the total strain of the specimen. The above data for deformed specimens are summarized in Table 1.

Fig. 12(a) shows the measured GBS contribution to the true strain at different strains regions. The GBS contribution increased from \(-4\%\) for Al-15Zn-3 to \(-18\%\) for Al-15Zn-1. During the initial deformation stage, the GBS contribution increased with increasing strain and reached a saturation value when the strain was higher than 20\%. The saturation of GBS contribution at higher strain is consistent with previous reports [30–32]. Furthermore, while the GBS contribution is plotted as a function of \( d^{-1/2} \), where \( d \) is the average grain size as shown in Fig. 12(b), it is apparent that the GBS contribution increases to a greater extent as grain size decreases, which is consistent with this paper’s present m-value result. It is interesting to note that the values of strain rate sensitivity and GBS contribution are within the range of similar order of magnitude regardless of grain sizes.

The Al-15Zn, as demonstrated in the present study, has some unique features different from typical superplastic metallic alloys. They are follows: (1) the increase in the m-value with decrease in grain size and (2) the occurrence of grain boundary sliding after deformation. In the following, the mechanism of flow for current Al-15Zn will be explored through the comparison of the experimental data with those anticipated from available deformation-mechanism models.

Several deformation mechanisms have been proposed to describe GBS. Among the currently available theoretical models based on GBS accommodated by diffusion [33] and slip [34–36], the model proposed by Ashby and Verrall (called A-V model) is in best agreement with the experimental results of Al-Zn alloy. Ashby and Verrall [33] developed a model based on observations of an oil emulsion. According to their model, accommodation of GBS occurs as a result of diffusional mass transfer along the boundaries or through the body of grains. The accommodation method is the superplastic analog of Nabarro-Herring [37,38] and Coble creep [39]. As proposed by Ashby and Verrall, grain shape is preserved during deformation by a “grain-switching” mechanism that also produces a material strain. Grain switching is illustrated in Fig. 13, which shows a two-dimensional configuration of four grains before (Fig. 13(a)), after (Fig. 13(c)), and at an intermediate stage of the grain-switching process.
There is an increase in grain-boundary area in the intermediate state compared to the initial and final state. Ashby and Verrall, considering volumetric mass transport (lattice diffusion) and mass transport through grain boundary (grain boundary diffusion), developed the following constitutive equation to describe the grain-switching creep rate:

\[
\dot{\varepsilon} = \frac{1000\Omega}{kTd^2} \left( \alpha - 0.72 \frac{\gamma}{d} \right) \left( 1 + \frac{3.3\delta D_{GB}}{D_L} \right) (4)
\]

where \(\dot{\varepsilon}\) is the steady-state creep rate, \(d\) is the grain size, \(T\) is the absolute temperature, \(\alpha\) is an effective grain boundary thickness for mass transport, \(\sigma\) is the flow stress, \(\Omega\) is the atomic volume, \(k\) is the Boltzmann constant, \(D_{GB}\) is an effective grain boundary diffusivity, and \(D_L\) is a lattice diffusivity. If only boundary transport is important (if \(D_{GB} \gg D_L\)) [11,40], Eq. (4) reduces to

\[
\dot{\varepsilon} = \frac{3300\Omega}{kT} \left( \frac{\alpha D_{GB}}{d^2} \right) \left( \alpha - 0.72 \frac{\gamma}{d} \right) (5)
\]

It can be immediately note that \(m\)-value is governed by the grain boundary diffusivity \(D_{GB}\), and strain rate and grain size \(d\). Region I is considered to be dominated by grain switching.

### Table 1 – Measurements of GBS in true strains for the Al-15Zn-1, Al-15Zn-2 and Al-15Zn-3 samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>(\dot{\varepsilon}(s^{-1}))</th>
<th>(\varepsilon_t(%))</th>
<th>(\bar{u}(nm))</th>
<th>(\bar{L}(nm))</th>
<th>(\delta(%))</th>
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<tbody>
<tr>
<td>Al-15Zn-1 1.0 \times 10^{-3}</td>
<td>5</td>
<td>3</td>
<td>480</td>
<td>10.2</td>
<td></td>
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<tr>
<td></td>
<td>13</td>
<td>11</td>
<td>480</td>
<td>14.1</td>
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<td></td>
<td>21</td>
<td>21</td>
<td>480</td>
<td>16.6</td>
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<td></td>
<td>40</td>
<td>41</td>
<td>480</td>
<td>17.2</td>
<td></td>
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<tr>
<td></td>
<td>60</td>
<td>62</td>
<td>480</td>
<td>17.2</td>
<td></td>
</tr>
<tr>
<td>Al-15Zn-2 1.0 \times 10^{-3}</td>
<td>10</td>
<td>5</td>
<td>900</td>
<td>4.4</td>
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<tr>
<td></td>
<td>15</td>
<td>10</td>
<td>900</td>
<td>5.9</td>
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<tr>
<td></td>
<td>26</td>
<td>19</td>
<td>900</td>
<td>6.8</td>
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<tr>
<td></td>
<td>48</td>
<td>38</td>
<td>900</td>
<td>7.0</td>
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<tr>
<td></td>
<td>60</td>
<td>47</td>
<td>900</td>
<td>7.0</td>
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<tr>
<td>Al-15Zn-3 1.0 \times 10^{-3}</td>
<td>8</td>
<td>3</td>
<td>1750</td>
<td>1.7</td>
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</tr>
<tr>
<td></td>
<td>12</td>
<td>5</td>
<td>1750</td>
<td>2.3</td>
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<td>1750</td>
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strain rate ($10^{-3}$ to $10^{-5} \text{s}^{-1}$), the $m$-value increases drastically from 0.1 to about 0.8 in submicron and ultrafine regimes. However, no evidence of a pronounced $m$-value at room temperature was found in ultrafine-grained materials. Therefore it can be concluded that the pronounced $m$-value seen at room temperature is associated with very high in Al–Zn alloys. The results are consistent with the experimental data of this study. The overall scheme as envisaged by Ashby and Verrall is summarized in Fig. 14. This shows that the proposed explanation of the pronounced $m$-value by the occurrence of GBS explained in terms of grain-switching is viable.

4. Conclusions

This study demonstrates that the method of scribing marker lines with scaled division using Focus Ion Beam micromachining is a suitable method to evaluate grain boundary sliding and its contribution to true strain in fine-grained and conventional grained materials. In the case of Al–15Zn alloys with an average grain size in the range of 0.4–2 μms subjected to tensile testing at room temperature, the grain boundary sliding contribution to true strain and $m$-value increases with reducing grain size. The result is very encouraging and suggests the use of this technique, not only for the determination of small offsets in small strains, but also for determination of the small contribution of grain boundary sliding in conventional grains. Therefore, this paper also concludes that grain boundary sliding is a viable deformation process in ultrafine-grained and conventional grained Al–Zn materials.

Acknowledgements

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Fig. 12 – (a) Variation in the GBS contribution with strain (b) the GBS contribution and SRS as a function of $d^{-1/2}$, where $d$ is the average grain size.

Fig. 13 – Schematic of a grain-switching event. Relative grain-boundary sliding produces a strain (c) without a change in grain shape (compared (a) with (c)). However, the intermediate step (b) of the process is associated with an increased grain-boundary area [33].
Fig. 14 – (a) The relationship between grain switching, dislocation creep, and superplasticity as proposed by Ashby and Verrall. Region I is considered to be dominated by grain switching and III by dislocation creep. The total strain rate is the sum due to both processes and this produces a high strain-rate sensitivity (b) in the transition region. Data used in calculations are appropriate for Pb with $d=1 \mu m$ at a temperature of 0.5 $T_m$ [33].

REFERENCES


