THERMOMAGNETIC TREATMENT OF ZINC: SELECTIVE GRAIN GROWTH AND TEXTURE MODIFICATION

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Abstract. Highly textured Zn-1.1%Al alloy with fine-grained microstructure was annealed in a high magnetic field of 32 T. The texture of the samples was characterized by the two 0002 components tilted at 15-20° from the normal to the rolling direction of the sheet. The annealing of samples parallel to the field preserved the maximum intensity of texture components and redistributed the intensity between original orientation of 0002 components and the normal direction. Annealing of samples at ~20° to the field resulted in the increase or retention of texture components with higher magnetic susceptibility and in the complete disappearance of the components with lower susceptibility. It has been shown that the difference in magnetic susceptibility creates an additional magnetic driving force for boundary migration. The magnetically induced boundary migration was studied using Zn bicrystals. Bicrystals with symmetric 90°<10T0> tilt boundaries were annealed in a high magnetic field of 25 T. The grains in the bicrystal had asymmetrical orientation with respect to the field. For the Zn polycrystalline alloy, the driving forces of magnetically induced and capillary driven boundary migration were found comparable at the average grain size of ~0.1 mm. This allowed interpreting the observed texture modification in terms of selective grain growth exerted by magnetic driving force for boundary migration.

1. INTRODUCTION

Usually, the driving force for grain growth or boundary migration (motion of the grain boundary normally to its plane) is created by internal structural factors such as boundary curvature, the difference in density of lattice defects in the neighboring grains, and others (e.g., [1]). In magnetically anisotropic materials placed in a magnetic field, the boundaries experience an additional driving force for their motion. This driving force, often called magnetic driving force, is the difference between magnetic free energies of differently oriented grains having conjoint boundaries [2]. In most of the anisotropic diamagnetic materials, that difference is extremely small. Therefore, magnetically induced boundary migration in those materials can be initiated in the case of sufficiently high fields [3]. Boundary migration is an important process of the evolution of grained structure and crystallographic texture during recrystallization and grain growth. Although a wide range of boundary disorientations and grain orientations with respect to the field affects effectively the motion of only certain groups of boundaries, for certain types of crystallographic texture a high probability for significant structural alterations can be expected.

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In this paper, we report the investigations of texture evolution in a Zn-1.1% wt Al alloy and migration of individual grain boundaries in Zn bicrystals during annealing in high magnetic fields.

2. EXPERIMENTAL PROCEDURES

Zn-1.1%Al alloy was prepared from high purity metals (99.995% Zn and 99.99% Al). The ingot was homogenized at 623K for 150 hours, rolled at 573K with a reduction of 50%, and finally rolled at room temperature for a total reduction of up to 99%. For each pass, reduction in thickness was about 5%. The direction of rolling was reversed after each pass. The final sheet thickness was 0.5 mm. In order to prevent coarsening of the Al-rich phase, the material was stored in a refrigerator at 203K before the annealing. The experiments used a resistive, steady state 32 T Bitter magnet with a 32 mm bore diameter. The samples of Zn-1.1%Al alloy were annealed at a temperature of 663K in a magnetic field of 32 T. The annealing time was 55 min. Pole figures from the surface area of each sheet sample were determined by the Schulz method using Cu Kα radiation and a Philips texture goniometer before and after annealing. For statistically reliable data, a sample oscillation of 10 mm was used during texture measurements. Pole figures were calculated using Philips X’Pert Texture software by calculating orientation distribution functions on the basis of five different raw pole figures. The specimens were oriented differently with respect to the magnetic field (Fig. 1). For one specimen, RD coincides with the direction of magnetic strength H (Fig. 1a) and for the other set of specimens RD is tilted at +19° (Fig. 1b) and -19° (Fig. 1c) to the field direction. Zinc bicrystals (99.995%) containing a 88.7±0.5°<1010> symmetrical tilt boundary were used (Fig. 2a). A bicrystalline plate was grown by the horizontal Bridgman method from molten Zn in a boat consisting of a polished graphite plate and mica flanges in an argon atmosphere. Specimens were cut from a bicrystalline plate at an angle of 60° with respect to the boundary using an electrical discharging machine. Basal planes show a 15±1° deviation from the parallel and perpendicular directions to the long side of bicrystal. The damaged layer adjacent to the surfaces was removed by chemical polishing on an acid-resistant cloth. Final polishing was performed electrolytically. The experiments were carried out using a direct current resistive, steady-state 27 T and 33 T Bitter magnets at the National High Magnetic Fields Laboratory in Tallahassee, Florida, USA. The bicrystal was first annealed with no field at a temperature of 663K for 20 min and then was removed from the furnace for surface observations. Subsequently, it was annealed in a field having a strength of H=1.99 10⁷ A/m at 663K for 5 min. The specimen was inclined at 15±1° with respect to the field H (Fig. 2b) making the hexagonal axes of grains A and B almost parallel and normal to the field. Additionally, the same type of specimen was annealed at the same temperature without a magnetic field for 100 hours. The annealing was interrupted after different periods for surface observations.

3. RESULTS

Fig. 3 illustrates (0002) pole figure of Zn-1.1%Al sheet after 99% rolling. As seen from this pole figure, two components with c-axis (basal) poles are tilted at some 15-20° from the normal direction to
the rolling direction (RD) around the transverse direction (TD). The texture intensities for different specimens are varied from 28.0 to 34.0 for component A and from 20.2 to 24.9 for component B (Table 1). Annealing specimens without a field slightly changes intensities of the texture components and retains the original type of pole figure after rolling (Fig. 4a). Splitting the texture components into a few subcomponents is ob served after annealing with and without a field. Annealing in a magnetic field changes the type of pole figure. When RD is parallel to the field there is some unification of two peaks into one, although the positions of the most intense subcomponents corresponds to the positions of the original components (Fig. 4b). During magnetic annealing of specimens tilted at +19° from the direction of the field, component B is totally annihilated while the intensity of component A rises by a factor of 1.5 compared to the intensity of the original component A (Fig. 4c). In the case of magnetic annealing of a specimen tilted at -19° from the H component, A disappears completely and component B increases in intensity by 3.8 times as much as the original component B (Fig. 4d). Fig. 5 illustrates specimen microstructures after rolling and after magnetic annealing. The grain size in the sheet surface obtained by linear intercept method is ~2 mm. Annealing results in the increase of the average grain size up to ~150 mm. Optical micrographs of birefringent specimens subjected to magnetic annealing and annealing with no field are shown in Fig. 6. The preliminary annealing of the bicrystal with no field during 20 min. resulted in some displacement of the boundary ends near the lateral surfaces, which can be determined by the position of a boundary groove. In contrast, during magnetic annealing for 5 min, the whole boundary migrated towards grain A (Fig. 6). The distance of migration varied from one end of the boundary to the other ranging from 0.9 to 1.8

Fig. 2. Geometry of a bicrystal (a) and its orientation to the field (b).

Fig. 3. Rolling texture (0002 pole figure) of Zn-1.1%Al sheet.
Table 1. Intensities of basal poles before annealing and their orientations to the field direction during annealing.

<table>
<thead>
<tr>
<th>Spec. No</th>
<th>$I_A$</th>
<th>$I_B$</th>
<th>Orientation of the specimen to magnetic field during annealing</th>
<th>Orientations of the c-axes to the field</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Component A</td>
<td>Component B</td>
</tr>
<tr>
<td>7.2</td>
<td>32.9</td>
<td>20.2</td>
<td>RD is parallel to $H$, (Fig. 1a)</td>
<td>70-75°</td>
</tr>
<tr>
<td>7.0</td>
<td>28.0</td>
<td>24.9</td>
<td>RD is at 19° to $H$, (Fig. 1b)</td>
<td>89-94°</td>
</tr>
<tr>
<td>7.4</td>
<td>34.0</td>
<td>20.3</td>
<td>RD is −19° to $H$, (Fig. 1c)</td>
<td>51-56°</td>
</tr>
<tr>
<td>7.8</td>
<td>29.8</td>
<td>23.9</td>
<td>No field</td>
<td>—</td>
</tr>
</tbody>
</table>

Fig. 4. Pole figures of Zn-1.1%Al sheet specimens after annealing. (a) no field; (b) oriented parallel to the field; (c) tilted at +19° to the field about the TD, (d) tilted at −19° to the field about the TD.
mm. The distance of migration in the middle of the boundary is equal to 1.54 mm. This was determined by averaging the measurements of migration in the middle on the front and back surfaces of the sample. Also, during migration the boundary changed its orientation in such a way that the boundary length decreased by approximately 9%. The orientation of the boundary line became almost perpendicular to the lateral surfaces. This effect was observed both on front and back surfaces of the bicrystalline samples. The position of the boundary after magnetic annealing (Fig. 6a) can be specified by an angle of $\psi = (\theta_a - \theta_b)/2$ which characterizes a deviation of boundary plane from its symmetry position. The magnitude of $\psi$ is measured to be about 25°.

Fig. 6b illustrates boundary migration and reorientation after annealing with no magnetic field for 100 hours. The boundary migration started on the lateral surfaces spreading gradually to the central part of the bicrystal. It can be seen that the angle of boundary reorientation is lower than that in the case of annealing in the field, and it takes much longer to reorient in this way. The boundary migration rate during annealing with no field is more than two orders of magnitude lower than that inside the field and the migration stopped after 5 hours of annealing. Further annealing for 95 hours made almost no change in the boundary position and orientation.

4. DISCUSSION

The results obtained demonstrate that annealing a Zn-1.1%Al sheet without a field makes a minor change in texture. Retaining the type of pole figure and intensity of texture peaks during annealing without a field can be predicted for the most hexagonal materials having no phase transformations. In contrast, annealing in a high magnetic field drastically changes the texture depending on the orientation of the specimen with respect to the direction of the magnetic field. Table 1 shows orientations of specimens and texture components during magnetic annealing. For different specimens shown in Fig. 1, basal poles are tilted at three different angles to the field. For the specimen with RD parallel to the direction of the magnetic field H (Fig. 1a), the angle between the a-axes of both components and the direction of the field is about 70-75°. The tilt of RD at +19° from the direction of the field (Fig. 1b) makes the c-axis of component A nearly perpendicular to the field. In turn, by tilting RD at -19°, the specimen is mounted in a position where the c-axis of component B is perpendicular to the magnetic field. The magnetic annealing of samples in both tilt-positions results in an increase of the texture peak corresponding to grains with the c-axis perpendicular to the field, while the other texture component disappears completely. The observed change in the type of texture can be understood from the experiments on zinc bicrystals. Annealing of bicrystals with no magnetic field resulted in boundary reorientation or rotational migration mainly near the lateral surfaces (Fig. 6b). It is worth noting that the boundary element, situated in the middle of the boundary length, did not move in the longitudinal direction and did
not rotate. Only the peripheral parts of the boundary rotated about the axis perpendicular to the specimen plane. The motion of the boundary in this case corresponds to the well-known bicrystal technique (so-called reversed-capillary technique) for measuring the grain boundary motion by applying the capillarity (reduction of boundary energy with displacement) as the driving force for grain boundary migration [4-6]. The direction of the boundary motion is normal to the boundary towards to the center of curvature. The driving force \( p_s \) is given by \( p_s = \sigma \cdot k = \sigma R \), where \( \sigma \) is the surface tension on the boundary, \( k \) the curvature, and \( R \) the radius of curvature. In the case of such geometry and under the assumption of shape invariance during migration, the curvature is inversely proportional to the distance from the vertex of \( a \) in Fig. 2a and the driving force decreases with increasing boundary displacement. Therefore, the boundary migration did not reach the center of the boundary and no rotational migration of the central part of the boundary was observed. The capillary driving force at the left and right sides of our specimens (Fig. 2) acts in opposite directions, rotating the peripheral portions of the boundary counterclockwise. In contrast, the relatively short (5 minutes) annealing of the specimen in a high magnetic field leads to much larger reorientation of the boundary plane (Fig. 6a) and to considerable movement of the boundary in the direction of grain A or, in other words, to the growth of grain B at the expense of grain A. This effect can be understood in terms of magnetic driving force for boundary migration created by the anisotropy of the magnetic susceptibility in Zn. Mullins [2] considered the origin of this driving force. If the volume density of the magnetic free energy \( \omega \) in a crystal induced by a uniform magnetic field is independent on crystal shape and size (the condition for this is \( \chi < 1 \)) then the magnetic driving force acting on the boundary of two crystals that have different magnetic susceptibilities is given by:

\[
\rho = \omega_1 - \omega_2 = \frac{\mu_0 H^2}{2} (\chi_1 - \chi_2),
\]

where \( \chi_1 \) and \( \chi_2 \) are the susceptibilities of crystal 1 and 2, respectively, along the magnet field \( H \). For the case of zinc bicrystals, Eq. (1) is transformed to

\[
\rho = \mu_0 \frac{\Delta \chi}{2} H^2 (\cos^2 \theta_1 - \cos^2 \theta_2),
\]

where \( \theta_1 \) and \( \theta_2 \) are the angles between the direction of magnetic field and the hexagonal (or c or <0001> axis) in both neighboring grains, \( \Delta \chi \) is the difference in susceptibilities parallel and perpendicular to the hexagonal axis. The force \( \rho \) is directed towards the grain with smaller value of \( q \) and does not depend on the sign of the magnetic field. The magnitude of the difference in the magnetic free energy of different grains in the Zn-1.1%Al alloy investigated can be estimated using the measurements of the crystal diamagnetism of Zn crystals [7]. According to [7] the gram susceptibility of Zn parallel and perpendicular to hexagonal (or c or <0001>) axes are \( \chi_q = -0.190 \times 10^{-6} \text{ cm}^3/\text{g} \) and \( \chi_i = -0.145 \times 10^{-6} \text{ cm}^3/\text{g} \), respectively. Conversion of these data from Gauss unit system gives for volume susceptibility in SI units as \( \chi_q = -1.695 \times 10^{-6} \) and \( \chi_i = -1.294 \times 10^{-6} \). According to Eq. (1) the maximum magnetic driving force grain growth in Zn in the case of the field strength of 2.55 \times 10^7 \text{ A/m}, and the difference in volume susceptibilities of \( \Delta \chi = \chi_i - \chi_q = 0.401 \times 10^{-6} \) is \( \rho_{\text{max}} = 1.65 \text{ kJ/m}^3 \). This force is related to a bicrystal with the angle of 90° between basal planes (or <0001> directions) oriented parallel and perpendicular to the field direction. For boundaries with other disorientation the driving force should be lower. It is reasonable to expect that grains corresponding to texture components A and B should have the highest proportion of common boundaries with disorientation angles ranging between 30° and 40° and TD as the rotation axis (Figs. 1 and 3). If the c-axis of the grains of one texture component is oriented perpendicular to the field, then these grains experience an additional driving force for the growth (or for the motion of their boundaries) in the direction of grains of another component. In this case the orientation of the c-axis of the second component, with respect to the field direction, ranges between 50° and 60° and the magnetic force according to Eq. (1) varies from 0.4 to 0.7 kJ/m^3. This force can be compared with usual capillary driving force for grain growth determined as

\[
\rho_s = \frac{2\sigma}{R},
\]

where \( \sigma \) is grain boundary energy and \( R \) the mean grain radius. The value of the ratio of the average grain boundary curvature to the inverse of the mean linear grain intercept has been experimentally found for Al as 0.31 throughout grain growth [8]. Applying the same relationship for Zn alloy with mean grain size of 150 mm we obtain \( R = 0.5 \text{ mm} \). Assuming a grain boundary energy of typically 0.3 J/m^2 [9], the
capillary force amounts to $p_c \equiv 1.2 \text{ kJ/m}^3$. Thus, a comparison of the respective driving forces reveals that the magnetic force is at least of the order of the capillary forces and able to make a strong influence on grain growth increasing the growth rate of those grains whose \(<0001>\) axis is perpendicular to the field. When the sample in our experiment is tilted at +19° the c-axis of component A becomes almost perpendicular to the field and the c-axis of component B is at 51-56° in respect to the field. Magnetic free energy of grains A reaches its minimum and becomes lower than the energy of all other orientations. Consequently, the additional magnetic driving force arises and enhances the growth of grains A. At the same time texture component B disappears. The growth in intensity of grains A is close to the drop in intensity of grains B. Therefore, it is reasonable to suggest that the disappearance of grains B is related to the growth of grains A, which grow mainly at the expense of grains B. When the sample is tilted at -19° to the field, the c-axis of component B becomes perpendicular to the field and grains B grow at the expense of grains A. In addition, some growth can naturally occur at the expense of other orientations. In a position where the sample is parallel to the field direction, the c-axes of both components are tilted at 70-75° to the field. The magnetic free energy of the grains of both components is equal and neither grains A nor B have additional force to grow. The experiment reveals that although the intensity of texture subcomponents after magnetic annealing in the orientation of RD parallel to the field (Fig. 4) remains almost the same as the intensity of texture components before the annealing, their c-axes become closer to the normal direction. Such behavior can be explained by preferable growth of those grains within the same component whose c-axis is closer to the normal direction of the field that results in some redistribution in texture intensity to the normal direction of the sheet.

5. CONCLUDING REMARKS
For the first time, it has been demonstrated that annealing a zinc alloy sheet in a high magnetic field can make significant changes in crystallographic texture. Depending on the orientation of the specimen to the applied field, the texture components can be strengthened, disappear or retain the original intensity. The intensity of the components related to the grain orientations with lowest diamagnetic susceptibility increases during magnetic annealing, whereas components related to higher susceptibility completely disappear. The experiments on bicrystals promote understanding of magnetically induced texture evolution. It is demonstrated that under the simultaneous action of capillary and magnetic driving forces the initially planar symmetrical 89° tilt boundary migrates reorienting its plane along the whole length nearly perpendicular to the lateral surfaces. The individual grain boundary in zinc bicrystal moves under high magnetic field in the direction of grain with the highest diamagnetic susceptibility. Detailed analysis of texture changes and the motion of individual boundaries shows that evolution of crystallographic texture in Zn alloy occurs due to selective grain growth induced by a high magnetic field.

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