Effect of Annealing Temperature on Grain Growth Process in Al-0.3 mass%Mg Alloy

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Grain growth behavior of an Al-0.3 mass%Mg alloy was investigated focusing on annealing temperature dependence of the changes of grain orientation and local grain boundary character distribution (GBCD).

Primary recrystallized specimens were isothermally annealed at 623 or 773 K and their microstructures were observed with an optical microscope and a SEM. Grain orientations and grain boundary structure were characterized using the SEM-ECP method.

The texture change can be divided into two stages during annealing. At the first stage, the cube texture developed as grain growth proceeded. Then, the cube texture declined with the development of these grains accompanied by remarkably abnormal growth of non cube grains at the second stage. This tendency was accelerated at lower temperature. The cause of abnormal growth of non cube grains is discussed with the change of local GBCD and the temperature dependence of grain boundary energies at the triple junction.

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I. Introduction

The nature of the grain boundary is very important for controlling and improving mechanical, chemical and physical properties of polycrystalline materials. Several approaches have been made to improving these properties by controlling the grain boundary character (GBC) which is evaluated using Σ values based on the coincidence site lattice theory⁽¹⁾⁻⁽⁷⁾.

The grain growth process has also been shown to be influenced by GBC; abnormal growth of Goss grains in Fe-Si alloys is closely related to the behavior of $\Sigma 9$ coincidence boundaries with high mobility and low energy⁽⁸⁾. Moreover, grain boundary character distribution (GBCD) is suggested to depend on the average grain size of several alloys; the frequency of coincidence site lattice boundaries increases with decreasing average grain size⁽⁹⁾. An inverse relationship was found⁽⁵⁾⁽¹⁰⁾, however, the details are not yet clear.

We have proposed that changes of grain orientation and GBCD during grain growth are strongly dependent on the initial microstructure such as spatial distribution of grain orientation, GBCD, and grain size for Al-Mg alloys⁽¹¹⁾⁻⁽¹³⁾. This paper presents the effect of annealing temperature on the change of grain orientation and GBCD in the growth process of Al-0.3 mass%Mg alloy focusing on the effect of spatial distribution of GBCD (local GBCD).

II. Experimental Procedure

An Al-Mg alloy ingot containing 0.3 mass%Mg was made by melting pure Al (99.99%) and Mg (99.98%) and homogenized at 773 K for 86.4 ks. The ingot was hot-rolled to 70% reduction at 773 K and then annealed at 623 K for 0.37 ks. Subsequent cold rolling of 90% reduction was carried out and a sheet with a final thickness of 0.9 mm was prepared. After primary recrystallization annealing at 558 K for 3.6 ks, further annealing for grain growth was performed at 623 K or 773 K for a period of 0.2 ks to 2.6 Ms.

Microstructures were investigated in the central portions of annealed specimens using an optical microscope and a scanning electron microscope (SEM), JSM-840A. Orientations of 200-330 grains were measured using the SEM-Electron Channeling Pattern (ECP) technique for each specimen. The grain size was determined by measuring the area of individual grains. Grain boundary structure was characterized by Σ values determined from the ECPs of two adjacent grains. Brandon's condition⁽¹⁴⁾ was used as the criterion of the exact coincidence orientation relationship.

III. Results

1. Changes of grain orientation distribution during grain growth

Figure 1 shows the distribution of grain orientation during grain growth. Primary recrystallization texture was mainly composed of cube {001}<100> and several

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Fig. 1 $\{111\}$ pole figures of Al-0.3 mass%Mg with various average grain sizes after annealing. (a) 558 K-3.6 ks, $18 \mu\text{m}^{(11)}$, (b) 623-0.7 Ms following 558 K-3.6 ks, $170 \mu\text{m}$, (c) 623 K-2.6 Ms following 558 K-3.6 ks, $220 \mu\text{m}$, (d) 773 K-0.2 ks following 558 K-3.6 ks, $160 \mu\text{m}^{(11)}$, (e) 773 K-10 ks following 558 K-3.6 ks, $480 \mu\text{m}^{(11)}$.

Fig. 2 Volume fraction of cube, normal direction-rotated cube (R-cube) and non-cube components as a function of the average grain size.

non-cube components. As grain growth proceeded, the cube component showed sharp distribution at both annealing temperatures. Grains with non-cube components became fewer and the non-cube texture became weaker at 773 K than at 623 K.

Figure 2 shows the changes in volume fractions of the

cube, normal direction (ND)-rotated cube and non-cube components during grain growth. In the present paper, the volume fraction of the cube component was calculated for grains with a deviation of less than 15° from an exact cube orientation, while all the ND-rotated cube orientations excluding the cube component were defined as rotated cube component. At 773 K, the volume fraction of the cube component remarkably increased at the first stage (stage 1), and this turned into a decrease at the second stage (stage 2) as grain growth proceeded. This was closely related to the spatial distribution of grain orientation, GBCD and grain size^{(11)–(13)}. In contrast, a notable increase in the volume fraction of non-cube components occurred in the second stage at 623 K after a rapid decrease in cube components.

2. Changes of grain size during grain growth

Figure 3 shows the relationship between average grain size with annealing temperature to explain the changes in frequency of the orientation component as shown in Section III.1. At the beginning of stage 2 (indicated by arrows) where the volume fraction of cube component was the largest, there was little difference between the average sizes of grains annealed at 623 and 773 K. The average size of grains with the non-cube component was larger than that with the cube component. The growth rate of non-cube grains was larger than that of cube grains and it was very remarkable at lower temperature. This suggests that an abnormal grain growth of the non-cube component must be accelerated at lower temperatures.

To clarify the difference in the texture change behavior during grain growth, the changes of GBCD were investigated, and the results are shown in the following section.

3. Changes of GBCD during grain growth

Figure 4 shows changes of GBCD during grain growth. The $\Sigma 3$ -51 boundaries in this study were classified as the coincidence boundaries. Two stages of GBCD change were observed in GBCD changes during grain growth. At stage 1, the frequency of the $\Sigma 1$ boundary increased with increasing grain size, while the opposite tendency oc-

Fig. 3 Average grain sizes of cube and non-cube components as a function of the annealing time.

Fig. 4 Variation in the frequency of the grain boundary character with the average grain size of Al-0.3 mass%Mg annealed at (a) 623 K and (b) 773 K⁽¹¹⁾.

curring at stage 2. Random boundaries showed an inverse tendency. The frequency change of the $\Sigma 1$ boundary corresponded well with that of cube grain.

It has been reported that coincidence boundaries with low energy and high mobility play an important role in selective grain growth⁽¹⁵⁾. However, there was no significant difference in the change of GBCD between the two temperatures during grain growth. Even the frequency of $\Sigma 3$ -51 boundaries did not change during grain growth at either temperature. Thus, more detailed analysis of microstructure is required on the basis of spatial distribution of orientation and local GBCD to understand the grain growth behavior at both temperatures.

4. Changes of local GBCD during grain growth

Figures 5 and 6 show the microtextures at the beginning and in the middle of stage 2 at two annealing temperatures. At the beginning of this stage, the microstructure consisted mainly of the cube grain matrix containing isolated non-cube grains at both temperatures (Figs. 5(a) and 6(a)). In the cube grain matrix, high frequency of $\Sigma 1$ boundary was recognized. In Fig. 5 coincidence boundaries with high mobility were more frequently seen around the non-cube grains than around other grains at 623 K and abnormal grain growth of non-cube components was accelerated there. To evaluate the local GBCD quantitatively, we classified the boundaries between adjacent grains into three groups as shown in Fig. 7: Cube/Cube (C/C), Cube/Non-cube (C/N), and Non-cube/Non-cube (N/N) grains⁽¹¹⁾.

Figure 8 shows changes of local GBCD in these grain

Fig. 5 Spatial distribution of grain orientations and GBC in Al-0.3 mass%Mg annealed at 623 K with various average grain sizes. (a) 170 μm , (b) 220 μm . The cube and rotated cube grains are shown in the gray shaded and hatched regions, respectively. The $\Sigma 1$ boundary, coincidence boundaries ($\Sigma 3$ to 51) and random boundaries are represented as the bold full, dashed and fine full lines, respectively.

boundary groups during grain growth. In the C/C group the frequency of $\Sigma 1$ boundary increased with increasing average grain size and the increase became remarkable at 623 K, while the coincidence and random boundaries decreased and their decrease was accelerated at 623 K.

In contrast, for the C/N group the frequency of coincidence boundaries maintained a constant value during grain growth at 773 K but sharply increased at 623 K. For the N/N group the frequency of $\Sigma 1$ boundary showed a sharp increase at 623 K. Since the local GBCD depends on average grain size and annealing temperature, abnormal growth of non-cube grains may be accelerated at lower temperature on the basis of local GBCD.

IV. Discussion

1. Effect of local GBCD on grain growth

A notable abnormal grain growth at low temperature is discussed as affected by grain boundary mobility and driving force for grain growth, focusing on the local GBCD. An increase in the frequency of $\Sigma 1$ boundary with low mobility for the C/C group induced the suppression of the growth rate of the cube grain matrix. This

Fig. 6 Spatial distribution of grain orientations and GBC in Al-0.3 mass%Mg annealed at 773 K with various average grain sizes. (a) 160 μm ⁽¹³⁾, (b) 270 μm . See Fig. 5 for an explanation of the grain orientations and GBC.

Fig. 7 Schematic illustration of microstructure and grain boundary groups.

suppression of the grain growth may aid the abnormal growth of non-cube grains⁽¹⁵⁾. In addition to this effect, the increase in the frequency of coincidence boundaries with high mobility for the C/N group eases the growth of non-cube grains resulting in acceleration of their abnormal grain growth.

The driving force for grain growth due to the strong texture, is given by the modification of Hillert's equation⁽¹⁶⁾:

$$P(T) = k(\sigma_c(T)/R_c(T) - \sigma_n(T)/R_n(T)) \quad (1)$$

Fig. 8 Local GBCD changes for three grain boundary groups in Al-0.3 mass%Mg during grain growth. (a) C/C boundary group, (b) C/N boundary group, (c) N/N boundary group.

where T is the annealing temperature, $P(T)$ is the driving force for grain growth of non-cube components, $\sigma_c(T)$ and $\sigma_n(T)$ are the mean specific interfacial energies of the cube and non-cube grains, respectively, $R_c(T)$ and $R_n(T)$ are the mean radii of the cube and non-cube grains, respectively, and k is a constant⁽¹⁷⁾. At the beginning of Stage 2, where the relationship of $R_c(623\text{ K}) \sim R_c(773\text{ K})$ and $R_n(623\text{ K}) \sim R_n(773\text{ K})$ is obtained from Fig. 3, the driving force for grain growth of the non-cube components is influenced by the difference in mean specific interfacial energy, namely, the grain boundary structure.

As the grain growth advanced, local GBCD changed depending on annealing temperature as shown in Fig. 8. Since the grain boundary energies of this alloy are not available, the effect of the mean specific interfacial energies on the driving force for grain growth of the non-

cube component cannot be quantitatively estimated at present. More measurements of grain boundary energies are required to estimate the driving force for grain growth of strongly textured materials.

2. Effect of temperature on orientation distribution of the cube component during grain growth

GBCD was strongly influenced by grain orientation distribution⁽⁵⁾⁽¹⁰⁾⁽¹⁸⁾⁽¹⁹⁾. The distribution of the grain misorientation angle in the C/C boundary group was investigated to clarify why local GBCD in cube matrix changed with temperature. As seen from Fig. 9, the average grain misorientation angle in the C/C group started from 9.5° for an as-recrystallized specimen, and then the angle decreased as the grain growth proceeded. The decrease was accelerated at a lower temperature. The angles at the beginning of stage 2 at 773 and 623 K, for example, were 8.5° and 7.9°, respectively. The low misorientation may help forming preferentially cube clusters containing a low angle $\Sigma 1$ boundary in the cube matrix. The cube clusters cause the increase in frequency of the low energy $\Sigma 1$ boundary and may suppress the growth of cube grains in the matrix. Furthermore, the non-cube grains which maintained coincidence orientation relationships with cube matrix grew much faster than cube grains. Thus, the orientation distribution of the cube components plays an important role in grain growth behavior of the cube and non-cube components.

The reason the grain misorientation distribution of cube components decreases during grain growth is discussed focusing on the energy balance of the triple junction of grain boundaries in the cube clusters and matrix. Grain boundary migration is known to be driven by an imbalance of grain boundary energies at the triple junction of these boundaries. Figure 10 shows a schematic illustration of the typical triple junction of C/C grain boundaries in a cube grain cluster or matrix. If the misorientation angle between small adjacent grains of similar size is minimal, the following assumption can be made at the triple junction composed of L1, L2 and H1 cube grains:

Fig. 9 Average misorientation angle inside the cube grain clusters in the matrix of Al-0.3 mass%Mg as a function of the average grain size.

Fig. 10 Schematic illustration of a triple junction of grain boundaries inside the cube grain cluster or matrix.

$$\theta_{L1/L2} < \theta_{L1/H1}, \quad \theta_{L2/H1} \quad (2)$$

where $\theta_{i/j}$ is the misorientation between the two adjacent grains i and j . When the misorientation angle is less than 15°, the grain boundary energy may change and be proportional to the misorientation angle. Therefore,

$$\sigma_{L1/L2} < \sigma_{L1/H1}, \quad \sigma_{L2/H1} \quad (3)$$

where $\sigma_{i/j}$ is the energy of the grain boundary formed by the two adjacent grains, i and j . In general, the equilibrium dihedral angles balanced with individual boundary energies at the triple junction are given by Herring's equation as follows:

$$\sigma_{L1/L2} / \sin(\alpha_{H1}) = \sigma_{L1/H1} / \sin(\alpha_{L2}) = \sigma_{L2/H1} / \sin(\alpha_{L1}) \quad (4)$$

where α_K is the dihedral angle⁽²⁰⁾. Consequently, from (3) and (4), these dihedral angles are derived as the following relation:

$$\alpha_{L1}, \quad \alpha_{L2} < \alpha_{H1} \quad (5)$$

In this type of triple junction which consists of one low energy boundary and two high energy boundaries, the relative critical size relationship for grain growth is given as follows⁽¹²⁾⁽¹³⁾:

$$d_{L1crit}, \quad d_{L2crit} < d_{H1crit} \quad (6)$$

where d_{Kcrit} is a relative critical size for grain growth. For instance, the grain H1 with $d_{H1} < d_{H1crit}$ will shrink and that with $d_{H1} > d_{H1crit}$ will grow. Relation (6) suggests that the grain H1, which has larger misorientation than the other two grains and requires larger relative grain size for growth, is easily shrunk by the triple junction migration, resulting in a sharp distribution of misorientations of the cube component. Therefore the average misorientation of cube grain cluster or matrix decreases as grain growth proceeds.

The relative critical size for grain growth also shows temperature dependence because the grain boundary energies depend on temperature. From the relations (3) to (6), the following can be derived:

$$\sigma_{L1/H1} / \sigma_{L1/L2} \propto d_{H1crit} / d_{L1crit} \quad (7)$$

From this relationship, the relative critical sizes can be ob-

tained by evaluating the ratio of grain boundary energies at the triple junction. Although grain boundary energy is not available for this alloy, the temperature dependence of grain boundary energies for pure Al was obtained⁽²¹⁾. The results of [100] tilt boundaries of pure Al were used as grain boundary energies of C/C group in Al-3 mass%Mg alloy. The ratio of the boundary energy ($\sigma_{L1/H1}(773\text{ K})/\sigma_{L1/L2}(773\text{ K})$) with a larger misorientation to the smaller one at 773 K is estimated to be 1.202, while the ratio ($\sigma_{L1/H1}(623\text{ K})/\sigma_{L1/L2}(623\text{ K})$) at 623 K is 1.210. Thus, the grain boundary energy ratio becomes larger at lower temperature, resulting in an increase in the relative critical size for the grain growth of highly misoriented cube grains. The highly misoriented cube grains therefore shrink more rapidly as the annealing temperature decreases. This result is not exactly correct but an addition of 0.3 mass%Mg may not drastically influence the tendency.

In summary, the orientation distribution of cube matrix, which strongly influences local GBCD, is very important to understand grain growth behavior of the present alloy.

V. Conclusions

The temperature dependence of grain size, grain orientation and grain boundary character distribution during grain growth for an Al-0.3 mass%Mg alloy was investigated at 623 and 773 K focusing on the local GBCD. The results are summarized as follows.

(1) Two stages in the change of orientation distribution during grain growth were observed at both temperatures. Cube components developed in the first stage and then decreased slightly in the second stage at 773 K. In contrast, non cube components remarkably increased and their abnormal grain growth was accelerated in the second stage at 623 K after a rapid decrease of the components in the first stage.

(2) Spatial distribution of GBC was dependent on the annealing temperature. In the cube grain matrix, the frequency of $\Sigma 1$ boundary with low mobility increased with decreasing temperature, while the frequency of coinci-

dence boundaries with higher mobility increased around the cube grain matrix at lower temperature. The changes of local GBCD accelerated the abnormal grain growth of non cube components as temperature decreased.

(3) The change of local GBCD inside and around the cube matrix during grain growth corresponded to the change in misorientation distribution of the cube component. The average misorientation of this component decreased with decreasing temperature. The temperature dependence of the imbalance of boundary energies at the triple junction of cube grain cluster and matrix was closely related to the difference in grain growth behavior at both temperatures.

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