Sub-grain Boundary Mobility of 6XXX Alloys during Annealing

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Abstract: In this paper, 6061 aluminum alloy and a new type of dispersion-strengthened 6XXX aluminum alloy (named WQ) were compressed to a true strain of 1.2 at a strain rate of 0.1/s at 300/400/500°C and then annealed at different temperatures. The microstructure evolution of both heat treated alloys were characterized by EBSD. The results showed that during the annealing process of the two deformed alloys, the sub-grain size did not increase significantly. Due to the pinning effect of the dispersoids in WQ alloy, the sub-grain size of WQ was smaller than that of 6061 alloy. Under the same deformation conditions and annealing process, the deformation stored energy of WQ alloy was higher than that of 6061 alloy. The deformation stored energy of 6061 compressed alloy was more sensitive to the annealing temperature. Combining theoretical formulas and EBSD data, the sub-grain boundary migration rate curves of 6061 and WQ deformed alloys during the annealing process were obtained, which could provide a theoretical basis for the future study of the sub-grain growth process of multi-element aluminum alloys.

1. Introduction

6XXX aluminum alloy (Al-Mg-Si-Cu alloy) is widely used in the manufacture of auto parts due to its high specific strength, good formability and corrosion resistance, including car bodies, seats and floor panels [1,2]. 6XXX aluminum alloy generally requires thermal deformation after homogenization heat treatment. Subsequent annealing and heat treatment are required to further improve the overall performance of the deformed alloy. In the process of low-temperature annealing of deformed alloy, the static softening mechanism is mainly recovery and recrystallization, and its kinetics research is very important [3,4]. During the static recovery process of the deformed alloy, the evolution of the microstructure is based on the sub-grain boundary (LAGBs). Low angle grain boundary migration is not only important in the recovery process, but also has a significant impact on recrystallization nucleation [5-7]. Yet, there is a lack of comprehensive and systematic research on the growth kinetics of sub-grains. With the development of high-resolution EBSD technology, people can describe the evolution of sub-grains orientation and misorientation, which will help to further understand the sub-grain growth process of 6XXX deformed alloy. Researcher [8,9,11] conducted a lot of exploration of the relationship between sub-grain structure and sub-grain boundary migration. In summary, previous studies on sub-grain growth and sub-grain boundary
migration of deformed aluminum alloys were more focused on single crystal or binary alloys with low solute elements, and there was almost no report correlated with the sub-grain growth behavior of deformed aluminum alloys with multiple high solute elements.

This paper selected two kinds of 6XXX aluminum alloys, namely the commonly used 6061 aluminum alloy and the new dispersion strengthened WQ Al alloy. Both alloys were conducted hot compression tests, which was followed by annealing heat treatment experiments at different temperatures. Then static recovery behavior of the two alloys during annealing process was studied and compared. The microstructure evolution of annealed alloys was characterized. The paper also analyzed the deformation stored energy evolution of the two deformed alloys during annealing process and calculated of the low angle grain boundary mobility of the two deformed alloys and reported the sub-grain growth behavior of deformed aluminum alloy with multiple high solute elements.

2. Materials and experiments
The 6061 and WQ Al alloys were homogenized at 550°C for 10 hours then air cooled to room temperature. The chemical composition of two studied alloys was measured and listed in Tab.1. After the homogenization heat treatment was completed, the samples of both alloys were machined into a compressed sample of φ10×15mm (Fig.1). The previous study [10] has been confirmed that WQ alloy containing transitional elements (Mn/Cr) would precipitate a large number of nano-scale α-Al(MnCr)Si dispersoids after homogenization heat treatment, which was not found in 6061.

The axial thermal compression experiment was conducted at 300/400/500°C with a strain rate of 0.1/s to a true strain of 1.2. The compressed samples were immediately quenched in water to retain the deformed structure and deformation storage energy. The deformed samples of two studied alloys were kept at 180°C, 240°C and 300°C for 0-4h, and then quenched to room temperature. The EBSD analysis data were used to calculate the misorientation distribution and size of sub-grain. EBSD experiment was carried out in a specified central area of the thermally compressed sample, as displayed in Fig.1b. The grain boundary with a misorientation greater than 15° was defined as a high-angle grain boundary (HAB), and a grain boundary with a misorientation between 1 and 15° was defined as a low-angle grain boundary (LAB), which was also known as sub-grain boundary. The grain boundary below 1° was not taken into consideration. To characterize the evolution of the microstructure during the annealing process, the grain boundaries with different misorientation range were illustrated by lines with different color: 1-5° green line, 5-10° red line, 10-15° blue line, > 15° black line.

![Fig.1. (a) Samples at different strains, (b) Observation area for EBSD](image-url)
3. Results and discussion

3.1 Sub-grain growth
In order to show the evolution of the (sub)grain boundary of the two deformed alloys during low-temperature annealing, the 400°C-deformed alloys were held at 240°C for different periods of time, and the samples were observed by EBSD to visualize the changes in the (sub)grain boundaries with different misorientation during annealing process.

Fig.2 shows the evolution of the sub-grain structure of the 6061 alloy deformed at 400°C and held at 240°C for different durations. The corresponding statistical evolution result of the grain boundary misorientation distribution was shown in Fig.4a. As shown in Fig.2, during the annealing process of 6061 deformed alloy, the sub-grain structure did not change significantly, and no obvious sub-grain growth and merging can be observed. The statistical results of the sub-grain size were shown in Fig.5c.

![Fig.2](image)

Fig.2. 6061 sample deformed at 400°C held at 240°C for (a)0.5h, (b)1h, (c)2h and (d)4h

WQ alloys were annealed treatment at 240°C following being compressed at 400°C. With the extension of annealing time at 240°C, the sub-grain structure did not show a significant change but a growth trend of sub-grain size can be observed. The result of grain boundary misorientation at different holding time was shown in Fig. 4b, and the statistical result of sub-grain size evolution was shown in Fig. 5d.
Fig. 3. WQ sample deformed at 400°C held at 240°C for (a) 0.5h, (b) 1h, (c) 2h and (d) 4h.

Fig. 4. Grain boundary misorientation evolution of deformed (a) 6061 and (b) WQ annealed at 240 °C for different time.

Fig. 4 showed that the 6061 and WQ deformed alloys presented similar evolution patterns of grain boundary misorientation during annealing heat treatment. In the initial stage of annealing, the frequency of sub-grain boundaries of 1-5° decreased, while the frequency of high-angle grain boundaries (>15°) increased, and then as the holding time was extended, the frequency of the two remains basically stable, while frequency of 5-15° low-angle grain boundaries has basically not changed. After deformation and quenching treatment, a large number of deformation dislocations were generated within the alloy, therefore large deformation stored energy were retained. In the initial stage of annealing at 240 °C, the deformation stored energy provided dynamic conditions for static recovery, causing rotation between adjacent sub-grain, leading to the further increase of (sub)grain boundaries misorientation. Thus the transition from low-angle grain boundaries to high-angle grain boundaries occurred, so the proportion of low-angle grain boundaries decreased and the proportion of high-angle boundaries increased. In addition, it was worth noting that during the annealing process, the proportion of low-angle grain boundaries in WQ alloys has always been higher than that of 6061, while the proportion of high-angle grain boundaries in WQ alloys has been lower than that of 6061. This was related to the thermally stable α-Al(MnCr)Si dispersoids in WQ alloys. Dislocation movement and sub-grain boundary migration as well as rotation were strongly pinned by dispersoids, thus restricting the transition from low-angle grain boundaries to high-angle grain boundaries.
After 6061 and WQ alloys were thermally compressed at 300/400/500°C, they were annealed at 180/240/300°C for different lengths of time respectively, and the evolution of the sub-grain size was counted and shown in Fig.5. It can be found that as the deformation temperature increased from 300°C to 500°C, the sub-grain size gradually increased. This was attributed to the fact that the increase in the deformation temperature further activated the dynamic softening mechanism and promoted the annihilation and migration of dislocations, resulting in a decrease in dislocation density and the increase of the sub-size size. However, it should be noted that due to the pinning effect of the dispersoids in the WQ alloy, the sub-grain size was smaller than that of 6061 under the same deformation condition. The sub-grain size of the two alloys increased with the extension of the holding time and the increase of the annealing temperature, but the increase was limited. The sub-grain size increased rapidly in the initial annealing stage, because a large amount of deformation stored energy was generated during the deformation process, which acted as the source for sub-grain growth energy. As the deformation stored energy was gradually consumed along with the later stage of holding period, the size of the sub-crystal appeared stagnant.

3.2 Evolution of stored energy
During the deformation of the alloy, the multiplication and entanglement of dislocations were formed in the Al matrix, and the deformation stored energy was retained in the matrix after instant quenching. During the static recovery process, the size of the sub-grain formed by the migration of the sub-grain boundary increased, and the dislocation density decreased, which in turn caused the stored energy to decrease. The storage energy $P$ can be represented by the average sub-grain boundary energy $<\gamma>$ and the sub-grain size $<d>$ [10]:

$$P = 2 <\gamma>/<d>$$ (1)

In the formula-1, $<\gamma>$ is the average subgrain boundary energy, which can be obtained by the Read-Shockley equation [11]:

$$\gamma = \frac{\sum_i n_i \gamma_i}{n} = \frac{\sum_i n_i \theta_i}{n} \left(1 - \frac{\theta_m}{\theta_a}\right)$$ (2)

In the formula-2, $\gamma_n = 0.324$ J m$^{-2}$ is the high-angle grain boundary energy of the aluminum matrix, $\theta_m=15^\circ$ is the high-angle grain boundary misorientation; $\gamma_i$ represents the energy held by sub-grain
boundary with misorientation of \( \theta \). Based on the assumption that the frequency of sub-grain with different misorientations was evenly distributed, \( \gamma \) was generally considered to be the average value, and formula 2 can be simplified as:

\[
\langle \gamma \rangle = \gamma_m \frac{\bar{\theta}_{\text{sub-grain}}}{\theta_m} \left( 1 - \ln \frac{\bar{\theta}_{\text{sub-grain}}}{\theta_m} \right)
\]  

(3)

However, with the help of EBSD technology, it can be known that the frequency of sub-grain boundaries with different misorientation are not evenly distributed. According to the EBSD results, the specific frequencies of the sub-grain boundaries with different misorientation can be obtained. Therefore, formula 2 can be modified to accurately calculate the sub-grain boundary energy:

\[
\gamma = \int_0^{\pi/2} \gamma_m \frac{\theta_i}{\theta_m} \left( 1 - \ln \frac{\theta_i}{\theta_m} \right) f(\theta_i) d\theta_i
\]

(4)

In the formula, \( f(\theta_i) \) is the proportion of the sub-grain boundary whose misorientation is \( \theta_i \).

According to the revised formula, the stored energy evolution process of 6061 and WQ deformed alloys during annealing can be obtained, and the result was shown in Fig.6.

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Fig.6. Stored energy evolution of 6061(a,c,e) and WQ(b,d,f) during annealing after deformed at 300°C (a,b), 400°C (c,d) and 500°C (e,f)

Obviously, the stored energy \( P \) of the hot compression alloy was related to the alloy type, annealing temperature, annealing time and hot compression conditions. According to the following formula:

\[
P \approx \frac{1}{2} \rho \mu b^2
\]

(5)

Where \( \rho \) is the dislocation density, \( \mu \) is the shear modulus and \( b \) is the Burgers vector.

It can be seen from Fig. 6 and Formula-5 that under the same deformation conditions and annealing process, the deformation stored energy of WQ alloy was higher than that of 6061 alloy, which was resulted from the hindrance of dislocation movement. On the one hand, Mn and Cr were added into WQ alloy, which lead to stronger interaction between solute atoms and dislocations. On the other hand, pinning effect caused by dispersoids will also further enhance accumulation of deformation stored energy. The stored energy of the same deformation alloy was related to the deformation conditions. As the deformation temperature increased, the stored energy of corresponding alloy decreased, which was owing to the fact that gradually intensified dynamic softening mechanism in the deformed alloy got the dislocation density to decrease. The deformation stored energy of 6061 alloy was more sensitive to the annealing temperature. As the annealing temperature rised, the deformation stored energy of the 6061 alloy decreased rapidly. The high annealing temperature provided higher dynamic conditions for the static softening mechanism and promoted the movement of dislocations and consequently accelerated the consumption of stored energy. The WQ deformation stored energy exhibited a similar evolution
curve in the studied annealing temperature range (180-300 °C) but higher energy, indicating that the thermally stable dispersoids can effectively inhibit the static softening mechanism in this temperature range. As the annealing time increased, the stored energy in the deformed alloy shows a downward trend, which was consistent with the increase in the size of the sub-grain in the studied alloys.

3.3 Sub-grain boundary mobility
The sub-grain boundary mobility $M_{sb}$ can be determined by continuous sub-grain growth [12]:

$$M_{sb} = \frac{d^2 - \bar{d}^2}{k\gamma_{sb}}$$  \hspace{1cm} (6)

Where $k$ is the geometric constant ($k=1.34$), $\gamma_{sb}$ can be determined by the modified Read-Shockley Formula [4] and $t$ is the time.

Plotted the relationship between $d^2-d_0^2$ and $t$ can be used to evaluate the alloy $M$ value at 180/240/300°C. The experimental results of this paper were compared with and high-angle grain boundary mobility of Al-0.3Mn binary alloy as well as low-angle grain boundary mobility of Al-0.1Mn and Al-0.3Mn in references [10], which was shown in Fig.7.

$$\text{Fig.7. Sub-rain mobility of (a) 6061 and (b) WQ}$$

In Fig.7, the solid lines were the results of this experiment, while the dotted lines were the results cited from the reference. The comparison indicated that, overall, the mobility of low-angle grain boundaries was much lower than that of high-angle grain boundaries. Compared with Al-Mn binary alloys, 6061 and WQ alloys showed poorer sensitivity to annealing temperature. With the increase of the annealing temperature, the low-angle grain boundary migration rate of the two alloys increased. The low-angle grain boundary mobility was related to the deformation conditions, that was the sub-grain boundary mobility of the alloy was higher under the higher deformation temperature. The sub-grain boundary mobility of WQ was lower than that of 6061 alloy, which was related to the existence of dispersoids and higher solute element content.

4. Conclusion
(1) During the annealing process of the two deformed alloys, the sub-grain size did not increase significantly. The pinning effect of the dispersoids limited the migration of dislocations in the WQ alloy, leading to smaller sub-grain size of WQ than that of the 6061 alloy.

(2) Deformation stored energy provided dynamic conditions for the static recovery of deformed alloys. Under the same deformation conditions and annealing process, the deformation stored energy of WQ alloy was higher than that of 6061 alloy. The deformation energy storage of 6061 alloy was more sensitive to the annealing temperature.

(3) The sub-grain boundary migration rate curve during the annealing process of 6061 and WQ deformed alloys was obtained, which provides a theoretical basis for the further study of multi-element aluminum alloy sub-grain growth process.
Acknowledgements
The authors acknowledge the financial support provided by Project of Shanxi Agricultural University (No.2020BQ80), Shanxi Province (SXBYKY2021021) and National Natural Science Foundation of China (Grant No. U1864209).

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