The influence of cube grain stored energy evolution on recrystallization texture and grain growth in 99.99% high purity Aluminum foil

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The high purity aluminum sheet was subjected to cold rolling and final annealing at 500℃, which was investigated by technique of X-ray diffraction (XRD) and electron backscattered diffraction (EBSD). The results show that an increasing of the energy gap between cube-{001}<100> grain and other orientated grain with an increased cold reduction, which supplies the energy to subsequent grain growth, especially for secondary grain growth, and the energy of orientated dependent acts as a driving force for grain growth abnormally during recrystallization after annealing, the mechanism of orientation energy difference which can explain the phenomenon for abnormal grain growth in annealed aluminum foil. And the cube-{001}<100> grain is prior to nucleate and grow. Therefore, the cube grain stored energy plays an important role for grain growth through the whole microstructure evolution during deformation and annealing.

(Received December 1, 2014; accepted June 24, 2015)

Keywords: High purity aluminum foils, Stored energy, Grain growth

1. Introduction

In our daily life, aluminum foil is widely used as package and high voltage anode electrolytic capacitor [1, 2]. However, using as high voltage anode electrolytic capacitor, which has the significant aspects about how to improve the specific capacitance of aluminum foil, and texture evolution, especially for cube texture nucleation and grain growth. Therefore, the cube texture component is a key factor to increase the specific capacitance for electrolytic capacitor. The previous work of inter-granular corrosion behavior in high purity aluminum has been studied by Kim et *al* [3], and information about the grain boundary character distribution (GBCD) as a parameter to describe the characteristic of investigated high purity aluminum foil.

On the one point of stored energy, as it knows that the stored energy existed in the crystal lattice as the form of defects such as dislocation, dislocation cell wall, vacancies, stacking faults and twinning [4]. During the cold working deformation, the grain orientation dependent stored deformation energy as a driving force for recrystallization and subsequent grain growth, which store in the deformed specimen. So the cube grain dependent stored energy act as an important role in the texture development and grain growth in annealed aluminum foils.

The cube texture evolution in aluminum foil of investigated material, which affected the continuous recrystallization (CRX) and grain growth was studied by Engler [1], the foil contains high stored energy which subjected to a heavily plastic cold reduction, and then the cube grain oriented dependent energy governs the mechanism of nucleation and grain growth. The 99.99% high purity aluminum foil is widely used in manufacture the high voltage electrolytic capacitors. However, the crystallographic <001>-directions as the effective channel for etching into this foil, a lot of researches have been made by investigators on the high purity aluminum, including the evolution of microstructure and texture [1, 5-7], the precipitation or second phase particle [8-10], grain boundary migration during grain growth [11-18]. The stored energy which being in aluminum and its alloys, Fe53%-Ni alloys and low-carbon steel [4, 19-27]. The typical dynamic and static recrystallization, subsequent grain growth has studied [28-35]. So nowadays, the aluminum and its alloy developed maturely many decades in the wide industry of metal materials, and its applications penetrated into every place of our daily lives.

In this paper, the important aspect of research is to investigate recrystallization and grain growth having relationship with variation of stored energy for cube texture, this texture evolution during rolling and annealing process was tracked by X-ray macro-texture [20] and EBSD micro-texture [36, 37], and optimize the processing parameters, finally it makes contributions to controlling and promoting the actual production, also promoting the production efficiency as well as good prospect for aluminum foil.

2. Experimental

The present investigated material of 99.99% high purity aluminum sheet (thickness～8mm) were subjected to a high level of cold reduction, the detailed two experimental routes were shown in Fig. 1, and which have been investigated after cold-rolling (CR) and annealing.

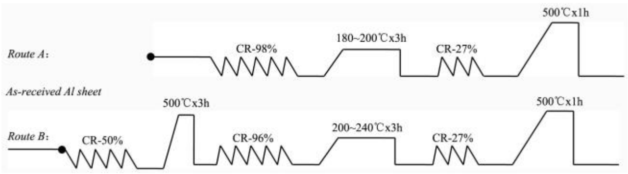

Fig. 1. Schematic description of the cold rolled and heat treatment routes for investigated material.

In *route A*, the first step of the samples were subjected to 98% cold rolling (～0.15mm) and subsequently annealed at 180℃ or 200℃ for 3 hours, which have been stored heavy energy during the high level of cold rolling reduction, and it acts as a driving force for nucleation and subsequent grain growth. The sample were cooled by water completely after annealed, then the second step of previous annealed samples were subjected to 27% cold reduction, and then it was annealed at 500℃ for 1 hour, these two steps are typical cold-rolling/annealing process.

In *route B*, it has an additional pre-annealing process before the two step annealing of *route A*, the *route A* and *route B* of these final annealed textures are investigated by X-ray diffraction (XRD) using a Rigaku D/MAX 2500 PC X-ray diffractometer. This orientation distribution functions (ODFs) of the deformed and recrystallized specimens are calculated from three complete quarter pole figures (111), (200), and (220) according to the series expansion method. The microstructure and orientation image microscope (OIM) are carried out by electron backscattered diffraction (EBSD) equipped with FEI Nova 400 FEG-SEM, these two steps of evolution of texture and microstructure are observed and shown in Fig.3 and Fig.5. Zeiss Libra 200 FE transmission electron microscopy (TEM) is used to observe the development of sub-grain and dislocation configuration, and the detailed experimental results are exhibited in the next sections.

3. Results and discussion

The samples are cold rolled along the original normal direction (ND) to final thickness of 4.0 mm (the deformation reduction is 50% and showing in Fig. 2b) and obtained the optical microstructure in Fig. 2a. The observed grain size of investigated material before cold rolling is uniformly distributed in crystallographic spatial (mean grain size～223μm) showing in Fig. 2a, the EBSD map of 50% reduction is given the information of the deformation micro-banding existed between the adjacent grain, the grains are squashed and elongated along the rolling direction. In order to obtain the information of rolling texture, the {111} pole figure [38, 39] shows that the rolling texture distribute randomly in this cold rolling sample (Fig. 2c,d), that is to say it does not form a strong texture, because of the low level of cold reduction, without preferred orientation. So there is not a strong texture before the sample annealing at 500℃ in the *Route B*, these results determine the final annealing texture, therefore the cube texture volume fraction is lower than the *Route A* via studying from another approach and shown in Fig. 5.

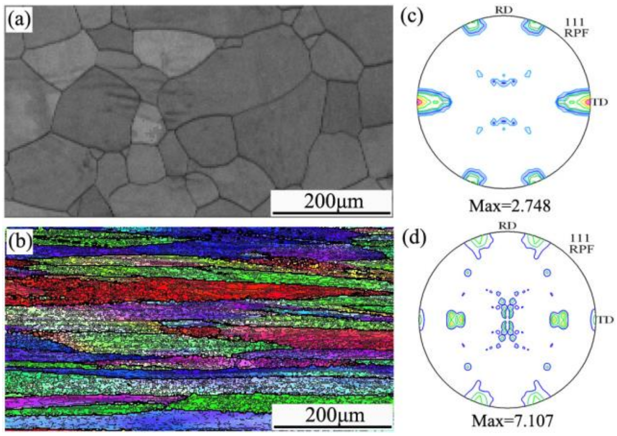

Fig. 2. Microstructure of aluminum foil for (a) optical microstructure of original states; (b) EBSD maps of 50% cold rolled reduction; (c) and (d) {111} pole figure are corresponding to (a), (b).

After the low level of cold reduction, orientation imaging microscope showed the information of slight deformation banding across different oriented grains, according to earlier investigation of deformation banding or shear banding by some researchers, such as Lee, Wagner *et al* [40, 41], the deformation banding or shear banding, which provides for a nucleation site to promote nucleation and grain growth, the deformed zone is covered with high level of distortion energy. On the one hand, this part of energy is consumed for recovery before recrystallization, on the another hand the surplus energy of annealed sample acts as a driving force for recrystallization and subsequent grain growth or secondary recrystallization (abnormal grain growth) [42, 43], the final grain size is becoming small or large.

The microstructure and textures of annealed foil of two routes are investigated by the technique of electron backscattered diffraction (EBSD), X-ray diffraction (XRD), we obtain the final microstructure and texture of annealed foil, which are shown in Fig. 3. According to the typical texture analysis theories, we can see that a classical recrystallization microstructure and strong plane-texture existed in the samples, the obviously complete recrystallization grain is shown in Fig. 3a and b, the corresponding (111) pole figure and inverse pole figure shown in Fig. 3(a1) and (b1), and inverse pole figure gave the information of typical (001) basal texture [42], P. Sonnweber-Ribic has studied the abnormal grain growth about texture evolution in thin Cu films, this basal texture is examined as a cube texture, the red color represents cube texture and showing in Fig. 3a and Fig. 3b, this texture component evolution from Fig. 2 to Fig. 3, and the grain size is ~241μm, finally the equiaxed recrystallization grains uniformly distribute in crystal spatial.

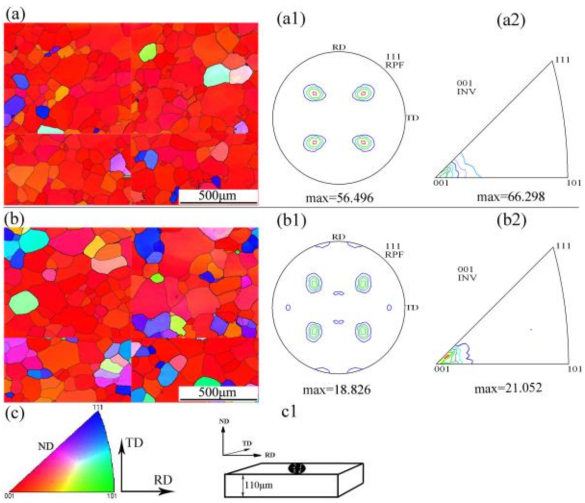

Fig. 3. Route A: (a) the final microstructure of two steps of cold-rolling/annealing; (a1), (a2) are corresponding to (a) (111) pole figure and (001) inverse pole figure. Route B: (b) the final microstructure of three steps of cold-rolling/annealing; (b1), (b2) are corresponding to (b) (111) pole figure and (001) inverse pole figure. (c) Color code and (c1) is the sampling point corresponding to (a) and (b).

Corresponding to Fig. 3, the high level of low misorientation angle distributes in the selected four areas (black area of 1.2.3.4), and the volume fraction is obtained by software of Channel 5.0.9, it is clearly to check the low and high angle grain boundary. Therefore, it is well to explore the mechanism and kinetic of recovery and recrystallization during annealing process.

According to the analysis of microstructure and texture from the data of Fig. 3, and corresponding to that the volume fraction of texture component is obtained via statistical analysis software, the recrystallized state (Fig. 3a and Fig. 3b) are shown as fraction of grain boundary in dependence on misorientation angle in Fig. 4a and Fig. 4b respectively.

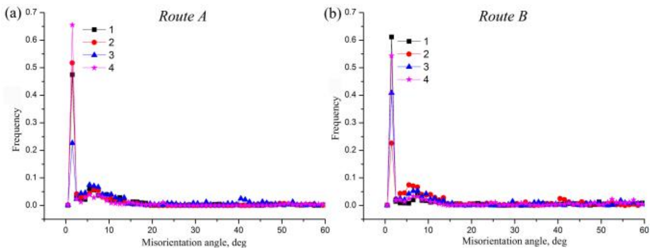

Fig. 4. Volume fraction of grain boundaries as a function of the misorientation angle; (a) the route A (seen Fig.3a); (b) the route B (seen Fig. 3b).

Both the distributions are similar within the statistical scatter of the oriented imaging maps, and it is obvious that the majority grain boundaries in both two routes are below 15°. Therefore, it is classified as low angle grain boundaries (LAGBs), The data need to be combined with the EBSD maps in Fig. 3, the small misorientation, which represent LAGB (seen in Fig. 4), there are much more frequent than they would be in a sample with the same texture and same orientation, it is clearly to have occurred completely recrystallization.

The tracks of cold rolled and annealed process for aluminum foil have been followed, then gave the information of the texture component volume fraction and showed in Fig. 5, the original states are mostly consist of random texture (R-texture) and a small fraction of Cube-{001}<100>, Brass-{011} <211>, Copper-{112} <111>, Goss-{011}<100> and S-{123}<634>, seen in Fig. 5a and b respectively. Finally, the texture component is mainly containing cubic texture, and the volume fraction is 60%, 31% (the red color rectangular bar is shown in Fig. 5) in *route A*, *route B*, respectively. The difference of volume fraction of cube texture between these two routes implies that the stored energy plays an important role with orientation dependent during microstructure evolution, the difference of orientation dependent energy between texture and texture, the relationship is plotted in Fig. 7, the energy gap (Fig. 7b) between cube and the other main orientations is a key factor for subsequent normal grain growth or abnormal grain growth [1].

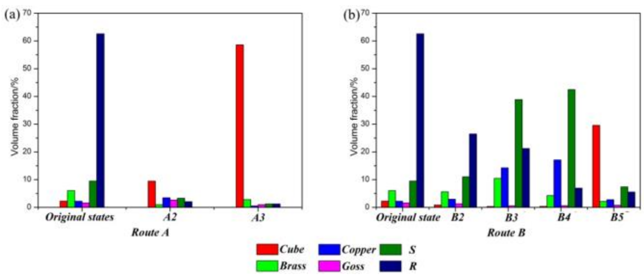

Fig. 5. Comparison of texture components volume fraction of (a) route A; (b) route B.

A twin-jet electro-polishing sample is put into TEM to obtain the bright field image as seen in Fig. 6a and Fig. 6b. The former map of grain boundary for thin aluminum foil is a small quantity of substructure and dislocation cell, it is containing equiaxed grain, the grain boundary edge sharpened. However, it exhibits that larger grain size in the Fig. 6b, the number of grain boundary is decreasing with the interface spacing increased, dislocation configuration is lamellar and non dislocation tangles [44], these parts of recrystallization structure after recovery and its energy has released partly, the only left energy which acts as a driving force for recrystallization and subsequent grain growth, this mechanism has been used to explain the normal grain growth, nevertheless, the mechanism of abnormal grain growth have a long way to explore.

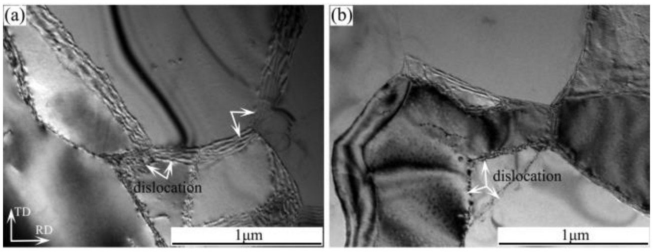

Fig. 6. Bright field TEM images: (a) dislocation configuration of the route A of cold rolled to 0.15mm then annealed at 200℃ for 3 hour; (b) the route B for pre-cold rolling to 4mm and annealed at 500℃ for 3 hour then cold rolled to 0.15 mm and annealed at 240℃ for 3 hour.

The experimental evidence of TEM maps is applied to analyze the evolution of dislocation configuration, and it is contributed to studying the slip system in the deformed materials. However, in next section, the stored energy of distribution as a function of the reduction rate R (55%, 77% , 95%) imparted by cold rolling [4] shown in Fig. 7a, compute the energy gap between Copper-C, S, Brass-B orientations and Cube shown in Fig. 7b, this deformed materials store high distortion energy, and value of C, S, B textures are increasing with the cold reduction increased.

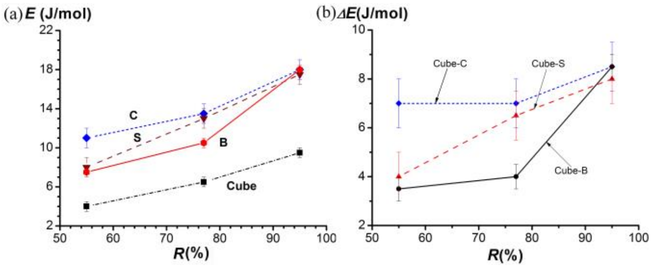

Fig. 7. Stored energy (J/mol): (a) the C-Copper, S, B-Brass and Cube orientations as a function of the reduction rate R (%) imparted by cold rolling [4]; (b) energy gap between C, S, B orientations and Cube versus rolling reduction R(%).

The researchers [4] had found that the energy gap between the cube and the other main orientations increases for a cold rolling reduction greater than 80%, it can be seen from Fig.7b, and confirm his research, while such an increase can promote the cubic grain growth during recrystallization by supplying the necessary driving force for cube grain boundary migration into the surrounding matrix. So the abnormal grain growth may be easy to occur, and it will grow larger.

The energy gap is provided to a driving force for grain growth, and the energy equation is showed in following:

$$\Delta P = (\Delta E) = (E_{other}\text{-}E_{cube}) = \frac{\alpha M\gamma}{R} = V/M$$

Where the ΔP is driving force, ΔE is energy gap, E_{-other} is the other grain oriented energy, E_{-cube} is cube oriented energy, M and γ are the grain boundary mobility and interfacial energy per unit of area, R is the radius of mean grain size, α is a non-dimension geometric factor, V is the grain boundary migration rate. The cubic grain orientation dependent energy is lower than the others as seen in Fig.7a, this difference of the orientation dependent energy is a necessary requirement for cube oriented grain easily to nucleate and grow.

4. Conclusions

The evolution of microstructure and texture during rolling/annealing of the aluminum foil was tracked by optical microscope, TEM investigations as well as X-ray macrotexture and EBSD microtexture analysis.

(1) In two routes of annealed aluminum foil, the cube grain nucleation and growth play an important role, the final microstructure and texture mostly consist of cube component.

(2) In deformed materials the cold reduction more than 80%, the energy gap between cube grain and other orientation grain is increasing and it supplies the energy for subsequent grain growth, especially for abnormal grain growth, during the process of recrystallization the energy with orientation dependent acts as a driving force for grain growth abnormally, this mechanism can explain the phenomenon of cube oriented grain growth quickly and abnormally in high purity aluminum foil.

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