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Evolution of recrystallization textures in high voltage aluminum capacitor foils①

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[Abstract] The evolution of recrystallization textures in high voltage aluminum capacitor foils which are produced with a high level of cold reduction was tracked by analysis of microstructure and crystallographic texture. The results show that the deformation textures are mainly composed of S-orientation, Cu-orientation and a little B_s-orientation. During the low temperature stages of final annealing, the iron precipitates first along the sub-grain boundaries, and the Fe concentration in the matrix becomes low. Then, the cube grains nucleate preferably into the sub-grains. At high temperature stages, the cube nuclei can grow preferably because of their 40° ⟨111⟩ orientation relationship to the S orientation, the main component of the rolling texture. Finally, the cube texture is sharply strong and the R orientation is very weak in the foils.

[Key words] aluminum capacitor foils; deformation textures; recrystallization textures

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1 INTRODUCTION

The capacitance of electrolytic capacitors depends on the effective surface of aluminum foils from which they are made[1]. The effective surface can be greatly increased by up to two orders of magnitude through an etching process that forms narrow channels along the crystallographic ⟨001⟩-direction into the foils. Since this "tunnel etching" follows the crystallographic ⟨001⟩-direction, a strongly textured material consisting only cube-oriented grains is favourable and therefore the aim of the processing. The cube texture in cold rolled high purity aluminum foils developed by recrystallization is mainly influenced by trace elements[2], hot rolling processing[3], final cold rolling reduction[4], heat treatment[5] and impurity contents[6], and so on.

And the work by Suzuki et al[6] demonstrated that the several factors were considered the origin of the suppression of the development of textures such as the existence of precipitates, clusters, and solute atoms. And the concentration of solute atoms in the matrix is the dominant factor, which has a strong influence on recrystallization behavior. In this paper we investigated the evolution of the recrystallization textures of high voltage aluminum capacitor foils during the final multi-stage annealing.

2 EXPERIMENTAL

In the present investigation a DC cast slab with the dimensions of 450 mm × 1 000 mm × 5 000 mm of high purity Al (99.99%) was used; the impurities present in that material are given in Table 1. After a homogenization treatment for 20 h at 600 ℃, the slab was reheated to 520 ℃ and held at this temperature for 2 h. Then the material was hot-rolled on a reversing breakdown mill to 16 mm thickness, and then hot-finishing-rolled to 7 mm thickness with an exit temperature of 310 ℃. The hot band was reversibly cold rolled with a thickness reduction of as much as 98% to a final thickness of 0.11 mm. Finally, the cold rolled specimens were annealed in a vacuum furnace at 200 ℃ for 3 h plus 520 ℃ for 2 h.

Table 1 Chemical composition of investigated aluminum (mass fraction, %)

Fe	Si	Cu	Mg	
0.0010	0.0008	0.0033	0.0019	
Mn	Zn	Ti	Ni	Al
< 0.002	< 0.002	< 0.002	< 0.002	Bal.

For a microstructural characterization of the cold rolled and recrystallized foils TEM was employed. The crystallographic textures were determined by measuring four incomplete pole figures {111}, {200}, {220}, {311} by means of fully automatic X-ray diffractometer using CuK_α radiation. The experimental orientation distribution functions (ODF) $f(g)$ were computed according to Bunge's series expansion method[7]. All ODFs were ghost corrected by using Gauss-type scattering functions[8]. In order to combine the information on the concurrent evolution

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of microstructure and texture during rolling and annealing, the EBSP was employed.

3 RESULTS AND DISCUSSION

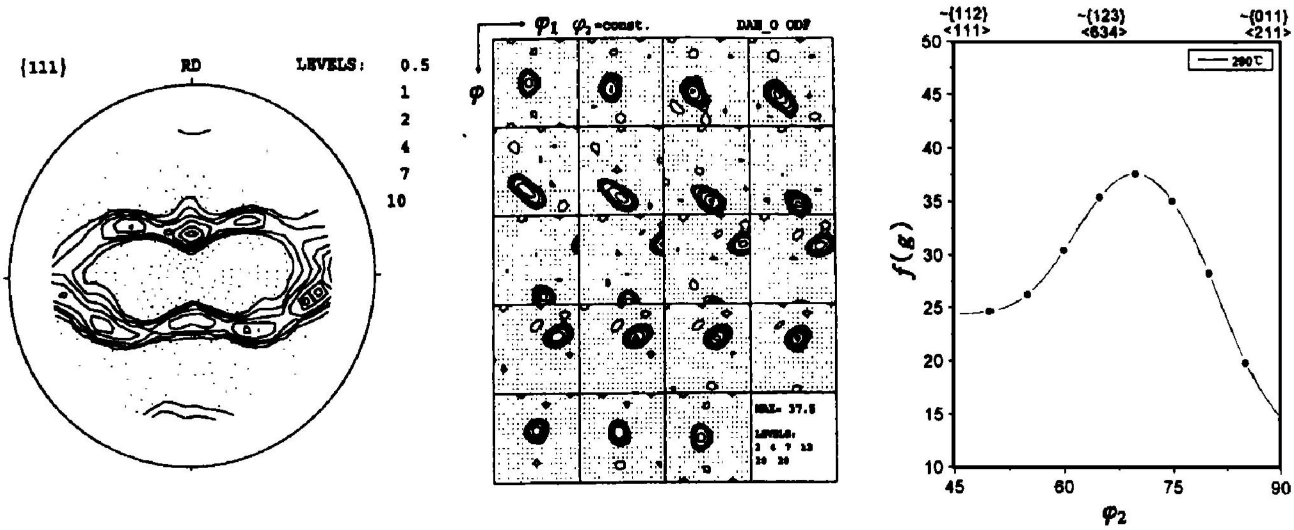
3.1 Experimental results

Fig. 1 gives the (111) pole figure, ODF and β-fibre of cold rolled foils in thickness of 0.11 mm. It shows that the deformation textures of high purity aluminum are mainly composed of S-orientation {123}〈634〉, Cu-orientation {112}〈111〉 and B_s-orientation {110}〈112〉. But the densities of Cu-orientation and B_s-orientation are very weak. The (111) pole figure and ODF of Al foils recrystallized at 200 ℃ for 3 h plus 500 ℃ for 2 h are shown in Fig. 2. The cube texture develops strongly, and the R orientation is very weak.

Fig. 3 shows a cube-oriented nucleus and its diffraction pattern of the foils annealed at 200 ℃ for 3 h according to TEM. Fig. 4 exhibits the appearance of precipitates which formed along the subgrain boundaries at low temperature stage of final annealing. Fig. 5 shows the completely recrystallized cube-oriented grains which are separated by approximately planar grain boundaries.

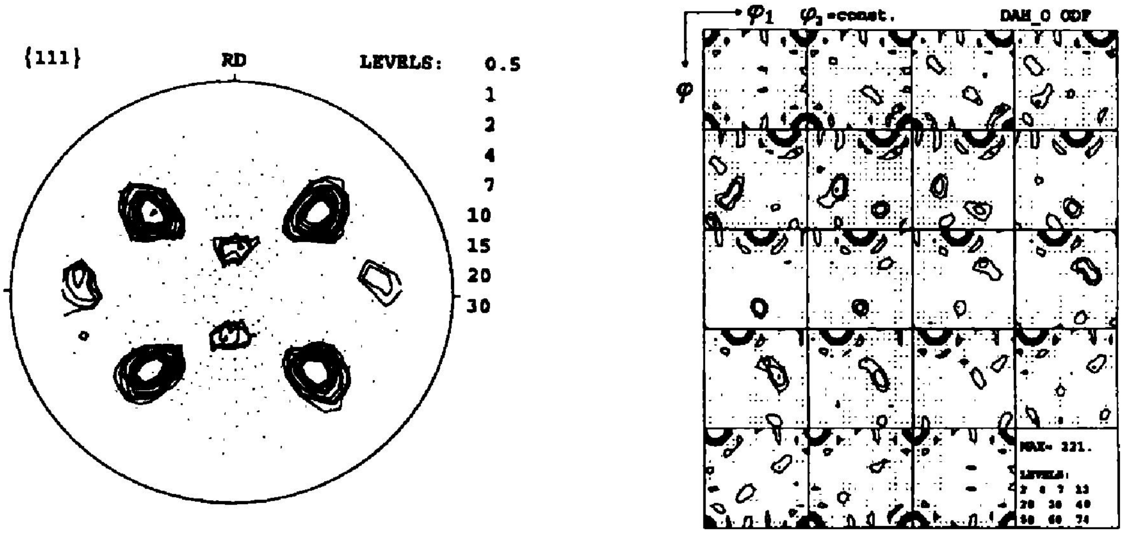
3.2 Discussion

It is known that the recrystallization textures are influenced by deformation texture[9], annealing temperature, heating rate[10], amount of iron in solution, and so on. But the origin of recrystallization textures during annealing of a deformed metal has been a matter of controversy for decades, with the discussion focusing on the rival theories[11], oriented nucleation and oriented growth. The oriented nucleation is assumed that the nuclei possess preferred orientations which by their growth will determine the texture. The oriented growth is assumed that a broad spe-



(a) (b) (c)

Fig. 1 (111) pole figure (a) and ODF (b) and β-fibre (c) for samples 0.11 mm in thickness (ε= 98%)



(a) (b)

Fig. 2 (111) pole figure (a) and ODF (b) for samples 0.11 mm in thickness after final annealing

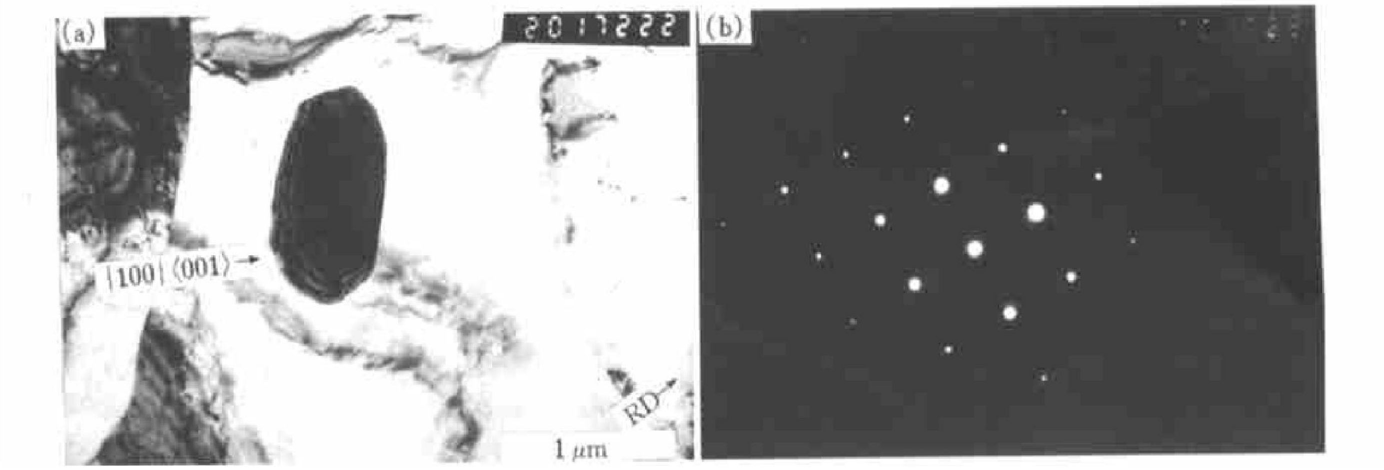


Fig. 3 Micrograph and diffraction pattern of cube nucleus
(a) —A cube-oriented nucleus annealing at 200 ℃ for 3 h;
(b) —Diffraction pattern taken at the cube-oriented nucleus with a arrow

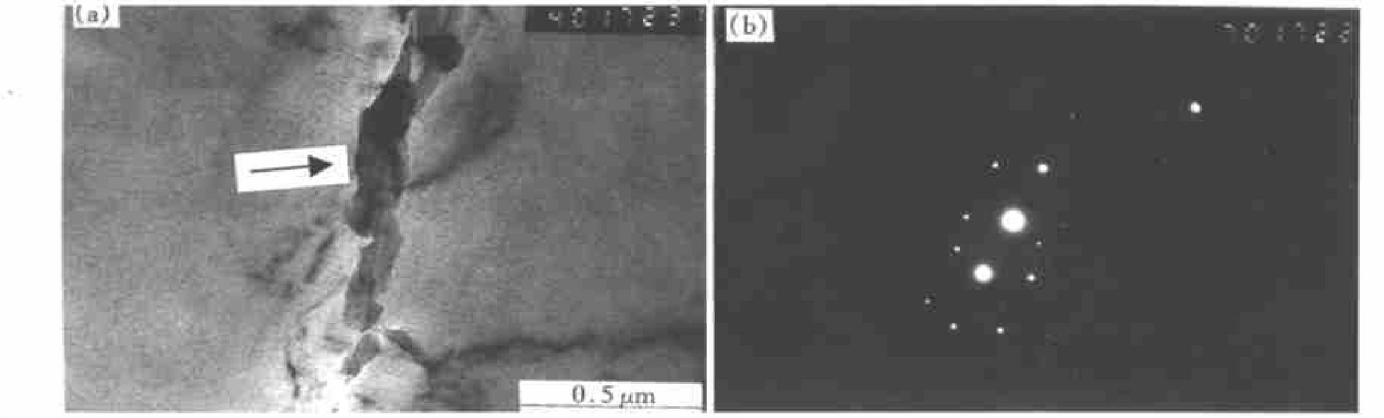


Fig. 4 Microstructure with precipitates along subgrain boundaries (a) and diffraction pattern taken at the precipitate with a arrow (b)

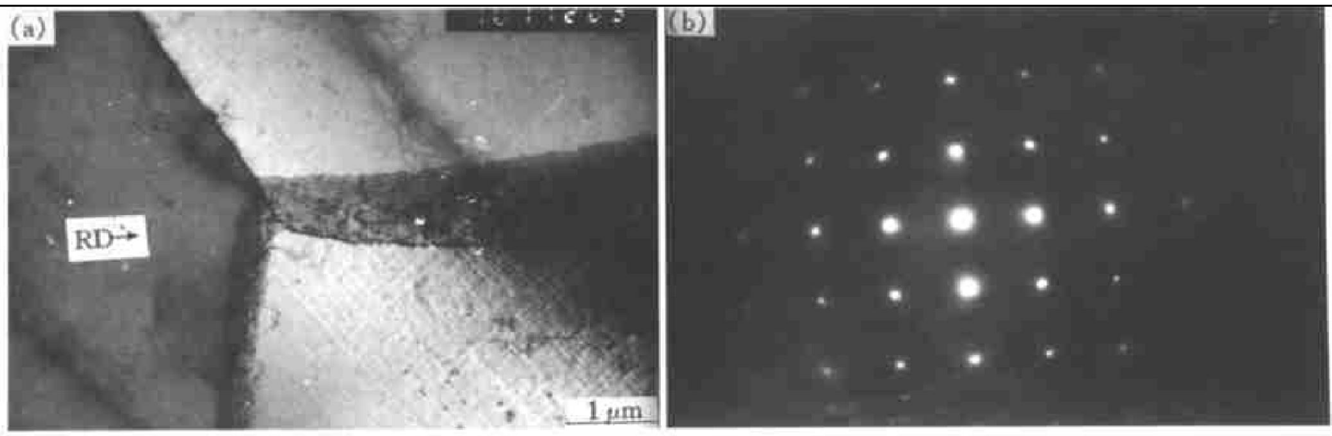


Fig. 5 TEM micrograph (a) and diffraction pattern (b) of recrystallized grains with cube orientation
(Annealed at 200 ℃ for 3 h plus 520 ℃ for 2 h after 98% cold rolling reduction)

ctrum of nucleus orientations is formed, and those with the best growth conditions with respect to the surrounding deformed matrix will determine the recrystallization texture. In Al, the cube texture is interpreted as nucleating in transition bands formed during preceding deformation. And the nuclei with a 40° ⟨111⟩ orientation relationship to the neighbouring deformation matrix were found to possess an especially high growth rate[12].

The presence of cube-oriented bands in the heavily deformed material is of importance to the question whether the cube recrystallization texture is developed by oriented nucleation or oriented growth. Cube-oriented grains presumably nucleate in cube bands that persist in the as-deformed state. TEM micrograph illustrating the formation of cube-oriented nuclei at low temperature stage of the final annealing is shown in Fig. 3(a). At the same time some precipitates are observed in Fig. 4(a). The Al-Fe equilibrium diagram shows that with the low iron content of the investigated material probably only $FeAl_3$ particles are present. The particles are not uniformly distributed as they display a tendency to cluster along the grain boundaries. Because the Fe concentration in the matrix becomes low, no dissolved or a little iron hinders the formation of the cube orientation. And the cube texture is formed so long as the Fe content in solution is sufficiently small. These precipitates explain why cube-oriented nuclei arise during the low temperature stage of final annealing[13].

The work by Ito et al[5] demonstrated that the dissolved iron hinders the formation of any genuine recrystallization and thus leads to the retaining of the rolling orientation, i. e. the R-orientation. The dissolved iron hinders the formation of the cube orientation but not that of the R-orientation by genuine recrystallization. In the present case, at low temperature stage of final annealing indeed very fine precipitates and cube-oriented nuclei have been observed respectively in TEM (Fig. 3(a) and Fig. 4(a)) and then the cube-oriented nuclei can grow preferably by discontinuous grain growth due to their 40° ⟨111⟩ orientation relationship to the S orientation which is a main component of rolling texture at high recrystallization temperature[14]. Whereas the driving force for recrystallization is provided by the lattice defects, in particular the dislocation stored in the as-deformed substructure during the preceding cold rolling deformation, grain growth is driven by the reduction in grain boundary area.

4 CONCLUSIONS

The nucleation sites are of primary importance and it has been found that the initial cube-oriented deformation bands within the original grains are preferential sites for recrystallization cube orientation nuclei in high purity aluminium (99.99%), deformed 98% by cold rolling. At low final annealing temperatures where most of the dissolved iron is precipitated before recrystallization, the cube-oriented nuclei can form. This is because the dissolved iron hinders the formation of the cube grains more strongly than that of the R-grains. And at high annealing temperatures the strong growth of the cube-orientation could be due to the lack of dissolved iron and because of their 40° ⟨111⟩ orientation relationship to the S orientation. Finally, the cube texture sharply increases and the R orientation is suppressed in the foils.

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