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Recrystallization of Al−5.8Mg−Mn−Sc−Zr alloy

Ying WANG[1], Qing-lin PAN[1,2], Yan-fang SONG[1], Chen LI[1], Zhi-feng LI[1], Qin CHEN[1], Zhi-min YIN[1]

1. School of Materials Science and Engineering, Central South University, Changsha 410083, China;
2. Key Laboratory of Nonferrous Materials Science and Engineering of Ministry of Education, Central South University, Changsha 410083, China

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Abstract: Al−5.8Mg−0.4Mn−0.25Sc−0.1Zr (mass fraction, %) alloys were prepared by water chilling copper mould ingot metallurgy processing which was protected by active flux. The recrystallization temperature and nucleation mechanism of the alloy were studied by means of hardness tests, observations of optical microscopy and transmission electron microscopy. The results show that the anti-crystallization ability can be significantly improved by adding minor Sc and Zr into Al−Mg−Mn alloy. This can be proved by a much higher recrystalliztion temperature (450 °C) than Al−Mg−Mn alloy without Sc and Zr (150 °C). The main reason of the great increase of recrystallization temperature can be attributed to the strong pinning effect of highly disperseded $Al_3(Sc,Zr)$ particles on dislocations and sub-grain boundaries. The recrystallizing process reveals itself the nucleation mechanism of the alloy involving not only the sub-grain coalescence but also the sub-grain growth.

Key words: Al−Mg−Mn−Sc−Zr alloy; recrystallization; $Al_3(Sc,Zr)$ particle; nucleation

1 Introduction

Al−Mg−Mn alloys have a wide range of applications in fields of aviation, spaceflight and machine-building due to their medium strength, good corrosion resistance, easy formability and good welding property [1−3]. This kind of alloy cannot be strengthened by heat treatment, as the Al_3Mg_2 precipitates will rapidly coarsen when the alloy is heated to a higher temperature. It was reported that the addition of alloying elements can modify the microstructure and thus improve the properties of Al−Mg−Mn alloys. The addition of rare earth elements, especially Sc, to Al−Mg−Mn alloy results in the refinement of as-cast grain sizes and improvements in both weld ability and mechanical properties.

Experimental investigations [4,5] showed that adding Sc as an alloying element was thought to be the most effective way to improve the properties of Al−Mg−Mn alloy. Moreover, the effect of Sc is found to be greatly amplified by Zr. The increase in strength mainly comes from grain refining strengthening, substructure strengthening and precipitation strengthening caused by $Al_3(Sc,Zr)$ particles [2,6,7]. In addition, the precipitates of the complex phase $Al_3(Sc,Zr)$ are supposed to have an even stronger anti-recrystallization effect [8−10]. This phenomenon indicates that it is very difficult to obtain a fully recrystallized microstructure even when the alloy is annealed at a very high temperature.

Recent studies have focused on the relationship between the microstructure and properties of Al−Mg−Mn alloy [11,12], but there are very few reports concerning the effects of the Sc and Zr double additives on the recrystallization behavior. In this work, the recrystallization temperature of Al−5.8Mg−Mn−Sc−Zr alloy is studied by means of optical microscopy and hardness measurement. The aim of this work is to investigate the effect of precipitates on recrystallization and the recrystallization nucleation mechanism.

2 Experimental

The Al−5.8%Mg−0.4%Mn−0.25%Sc−0.1%Zr alloy (mass fraction, %) was cast via metallurgy with pure Al, Mg and master alloys (Al−2.23%Sc, Al−4.48%Zr, Al−8.5%Mn) into a 260 mm×150 mm×30 mm ingot. The

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Corresponding author: Qing-lin PAN; Tel/Fax: +86-731-88830933; E-mail: pql@csu.edu.cn
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specimens were taken from the middle of the ingot and homogenized at 460 °C for 24 h. Then a hot-rolling process was applied to 5.7 mm (77% rolling reduction) at 470 °C, which was followed by intermediate annealing at 400 °C for 2 h. Subsequently, the hot-rolled sheets were cold-rolled to a thickness of 2.0 mm (65% reduction). The cold-rolled sheets were annealed at 100, 150, 200, 250, 300, 350, 400, 450, 500, 550 and 580 °C for 1 h, respectively.

The hardness test was carried out on a 401MVD MICRO-VICKERS. The microstructure characterization was performed using a POLYVER-MET optical microscope (OM) and a 200 kV Tecnai $G^2$20 transmission electron microscope (TEM). The optical microscopy of the specimens was observed by polar light after electro-polishing using an electrolyte solution consisting of 10% $HClO_3$ and 90% ethanol (volume fraction). Thin foils for TEM analysis were prepared by twinjet polishing with an electrolyte solution consisting of 25% HNO_3 and 75% methanol (volume fraction) below −25 °C.

3 Results

3.1 Recrystallization temperature measurement of alloy

The recrystallization is studied by microhardness measurements, which is a function of isothermal annealing temperatures. Figure 1 shows the variations of hardness of the cold-rolled Al−Mg−Mn−Sc−Zr and Al−Mg−Mn alloy, after isothermal recrystallization treatments at different temperatures. It can be seen from Fig. 1 that the starting recrystallization temperature of Al−Mg−Mn alloy is 250 °C, while the ending recrystallization temperature is 350 °C. This result is consistent with the observation of optical microstructures. In contrast to Al−Mg−Mn alloy, the alloy with minor Sc and Zr has no obvious starting and ending recrystallization temperature.

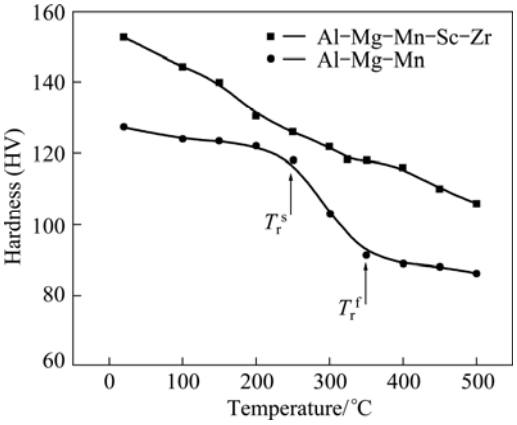

Fig. 1 Variations of hardness at different annealing temperatures for 1 h

3.2 Optical microscopy observation of alloy

The optical microstructures of Al−Mg−Mn alloy are shown in Fig. 2. The alloy presents obvious fibrous rolling deformation organization (Fig. 2(a)). After being annealed at 300 °C for 1 h, partial recrystallization with fine equiaxed grains can be observed (Fig. 2(b)). When the temperature increases to 350 °C, the alloy is completely recrystallized, producing fine and nearly equiaxed microstructure, as shown in Fig. 2(c). No grain size change can be found when the alloy is treated at 450 °C (Fig. 2(d)). But when annealed at 500 °C for 1 h, there is an obvious growth of grains (Fig. 2(e)).

In addition, the specimens of Al−Mg−Mn−Sc−Zr alloy are also observed in an identical manner for a comparison of the recrystallization progress (Fig. 3). The microstructure of the alloy treated at 400 °C maintains fibrous rolling deformation organization and the surface of the deformation organization is undulating(Fig. 3(a)). After being annealed at 450 °C for 1 h, partial recrystallization with very fine recrystallization grains is observed in Fig. 3(b). When the temperature increases to 550 °C, the cold rolled alloy has not yet fully recrystallized, and still keeps fibrous rolling deformation organization, as shown in Fig. 3(d). Increasing the temperature to 580 °C, the complete recrystallization structure with nearly equiaxed grains is found (Fig. 3(e)). It can be concluded from the above observations that addition of Sc and Zr can retard recrystallization during annealing, and the recrystallization temperature of Al−Mg−Mn−Sc−Zr alloy is 150 °C higher than that of Al−Mg−Mn alloy without Sc and Zr.

3.3 TEM observation of alloy

For further investigation on the nucleation mechanism of the Al−Mg−Mn−Sc−Zr alloy, TEM observations were conducted. Figure 4 shows the morphology of specimens after stabilizing treatment at different annealing temperatures for 1 h. After 200 °C stabilizing treatment, a large number of dislocation tangles are found, and the dislocations sparsely distribute near grain boundaries, forming clear cellular substructure shown in Fig. 4(a). Increasing the temperature to 350 °C (Fig. 4(b)), the recovery process in the matrix of the alloy begins with some low-angle sub-grains formed in some areas. At the same time, spherical morphology of the second phase particles is clearly visible. The matrix has been completely recovered and high-angle grain boundaries are formed when the annealing temperature goes to 450 °C. In this case, both the number and the size of sub-grains increase and some recrystallized grains appear. It is the evidence that recrystallized procedure begins at this temperature, which accords with the results of optical microscopy. When being annealed at 550 °C, the recrystallization develops but the sizes of

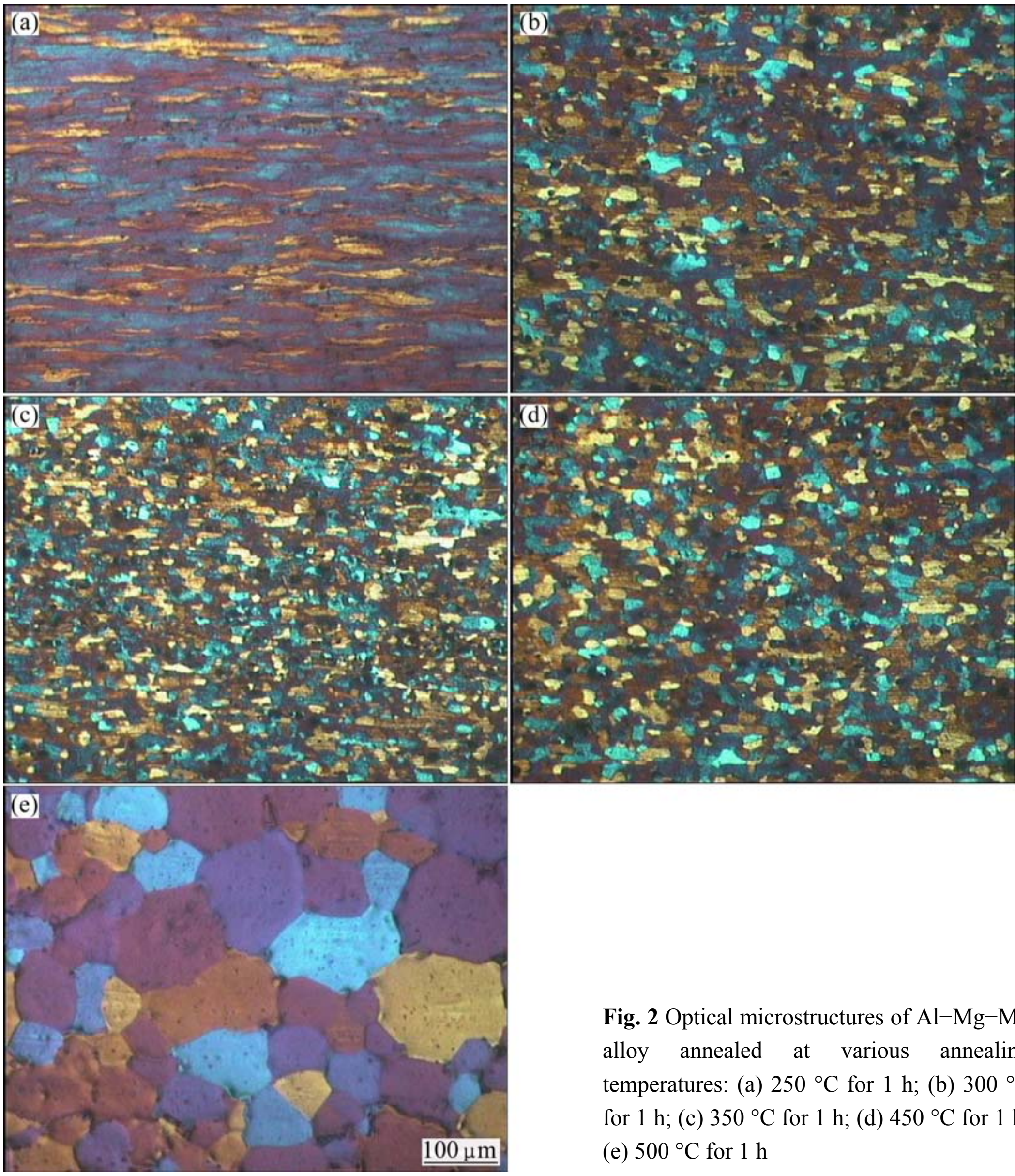

Fig. 2 Optical microstructures of Al−Mg−Mn alloy annealed at various annealing temperatures: (a) 250 °C for 1 h; (b) 300 °C for 1 h; (c) 350 °C for 1 h; (d) 450 °C for 1 h; (e) 500 °C for 1 h

recrystallized grains remain close to those of the sub-grains(Fig. 4(e)). With a higher magnification, most of sub-grains disappear while smooth trinary grain boundary appears, as shown in Fig. 4(f).

4 Discussion

4.1 Effects of precipitates on recrystallization

The observation of microstructures above indicates that the recrystallization temperature can be increased from 150 °C to 450 °C by adding Sc and Zr to Al−Mg−Mn alloy which proves that the researched alloy has a good thermal stability. Research in Refs. [13,14] showed that the retarding recrystallization of alloy had close relationship with the Al_3(Sc,Zr) particles. Figure 5(c) shows that there are many dispersed Al_3(Sc, Zr) particles in the alloy, which are petal-shaped. The selected area diffraction (SAD) illustrates that the Al_3(Sc,Zr) particles are coherent with the matrix (Fig. 5(d)). The orientation relationship of the Al_3(Sc,Zr) particle is $[110]_{Al_3(Sc,Zr)}//[110]_{Al}$ and $(002)_{Al_3(Sc,Zr)}//(002)_{Al}$. In addition, Zener's theory [15,16] shows that the coherent interface between the dispersoid and matrix can increase the resistance to recrystallization. Based on the theory and experiment results, we can conclude that the Al_3(Sc,Zr) precipitates play an important role in hindering the recrystallization of Al−Mg−Mn−Sc−Zr alloy.

The dispersed Al_3(Sc,Zr) precipitates can be obtained inside sub-grain and at sub-grain boundaries after the alloy was annealed at various temperatures. TEM observation indicates that these Al_3(Sc,Zr) particles exert a strong pinning effect (Zener pinning) on sub-grain boundaries (Fig. 5(a)) and dislocations (Fig. 5(b)),

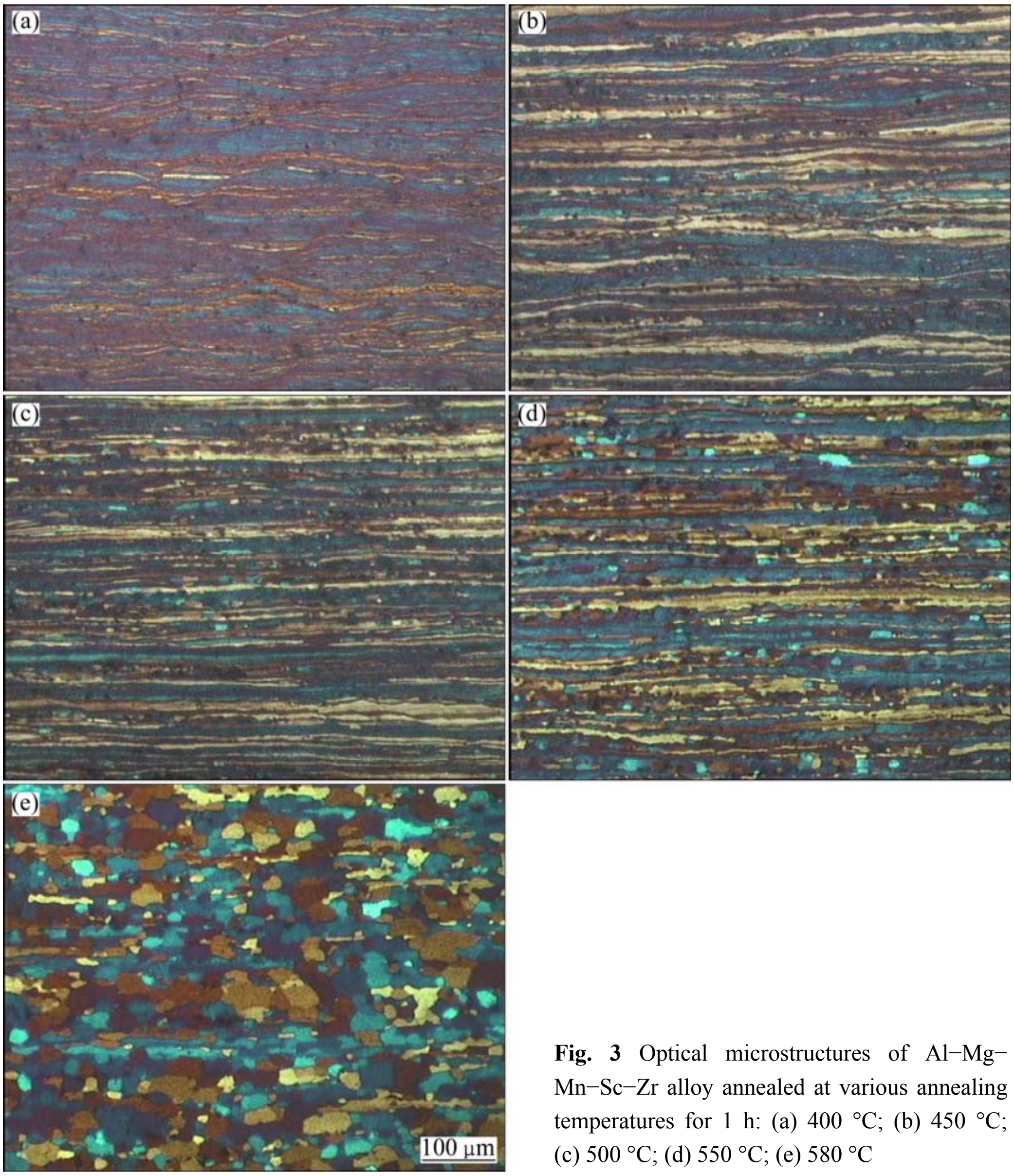

Fig. 3 Optical microstructures of Al−Mg−Mn−Sc−Zr alloy annealed at various annealing temperatures for 1 h: (a) 400 °C; (b) 450 °C; (c) 500 °C; (d) 550 °C; (e) 580 °C

which raised difficulty of the accumulation of sub-grains, thus stabilizing the sub-grains and hampering the nucleus formation of recrystallization. In the annealing treatment above 450 °C, the $Al_3(Sc,Zr)$ particles retard both the formation and growth of the recrystallization. The strong drag effect of these precipitates is due to the fact that they are coherent with Al matrix and very thermally stable against loss of coherency and coarsening [17−19]. Recrystallization process is completely over only after these ultrafine precipitates have coarsen and grown up.

4.2 Nucleation mechanism of recrystallization

It is well-known that there are at least three models to explain the recrystallization of aluminum alloys: sub-grains merging model, sub-grains growing model and grain boundary bending model. Our study with regard to the nucleation mechanism of recrystallization of the researched alloy prefers sub-grains merging mechanism and sub-grains growing mechanism. During the cold-rolling process, high-density dislocations and a large number of dislocation tangles are generated. After stabilizing annealing at different temperatures, the recovery makes the dislocations density ever higher. Thus, the low-angle sub-boundaries and sub-grains are formed gradually. A further increase in the annealing temperature (up to 450 °C) witnesses the merging of sub-grain boundaries in some areas of the alloy (Fig. 4(c)), as well as the merging of dislocations at the border of the two sub-grains (Figs. 4(c,d)). In this case, it indicates that there are mutual reactions between dislocations and sub-grains which are supported by sub-grains merging mechanism. Meanwhile, the matrix has formed high-angle boundaries and grains, as shown in Fig. 4(d). All results above indicate that the nucleation

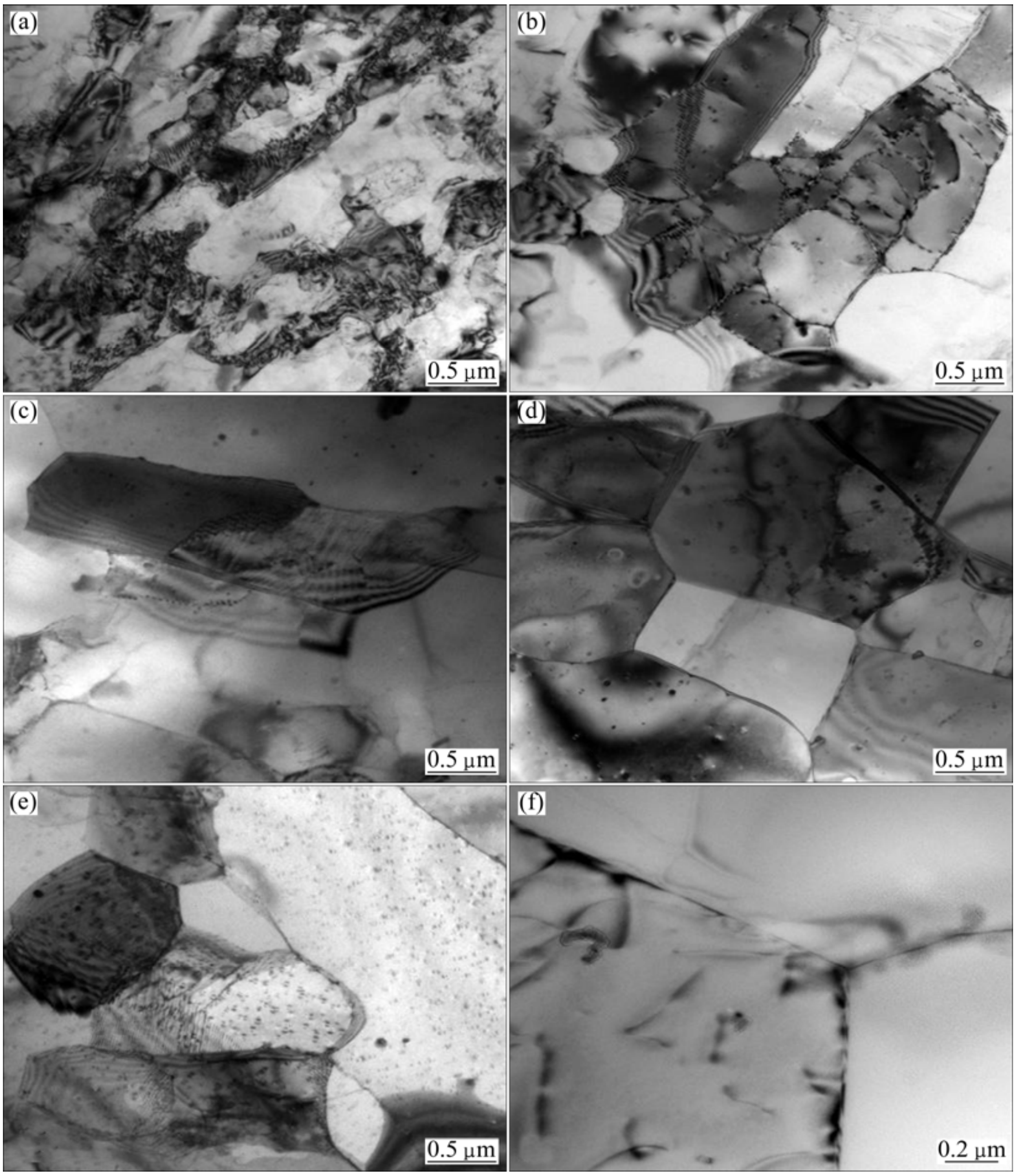

Fig. 4 TEM images of Al−Mg−Mn−Sc−Zr alloy annealed at various annealing temperatures for 1 h: (a) 200 °C; (b) 350 °C; (c), (d) 450 °C; (e), (f) 550 °C

mechanism of recrystallization involves not only sub-grain coalescence but also sub-grain growth. Furthermore, the sub-grain boundaries may move with the annihilation and incorporation of dislocations from the matrix. Additionally, the increase in misorientation between neighboring sub-grains leads to the disappearance of low-angle grain boundaries [20,21]. Although this is a relatively slow process depending on thermally activated dislocation climb and cross slip, the nucleation of the recrystallization is complete. As a result, the high-angle grain boundaries move quickly, leaving a distortionless fully recrystallized microstructure behind, and subsequently, a group of sub-grains are aggregated to form new recrystallization grains.

5 Conclusions

1) The recrystallization temperature can be increased from 150 °C to 450 °C by adding Sc and Zr to Al−Mg−Mn alloy.

2) Co-addition of small amounts of Sc and Zr can retard the recrystallization because the $Al_3(Sc,Zr)$ particles exert a strong pinning effect on dislocations and sub-grain boundaries, which inhibits the formation and merging of sub-grains.

3) The $Al_3(Sc,Zr)$ particles are coherent with the matrix, and the orientation relationship is $[110]_{Al_3(Sc,Zr)}//[110]_{Al}$ and $(002)_{Al_3(Sc,Zr)}//(002)_{Al}$.

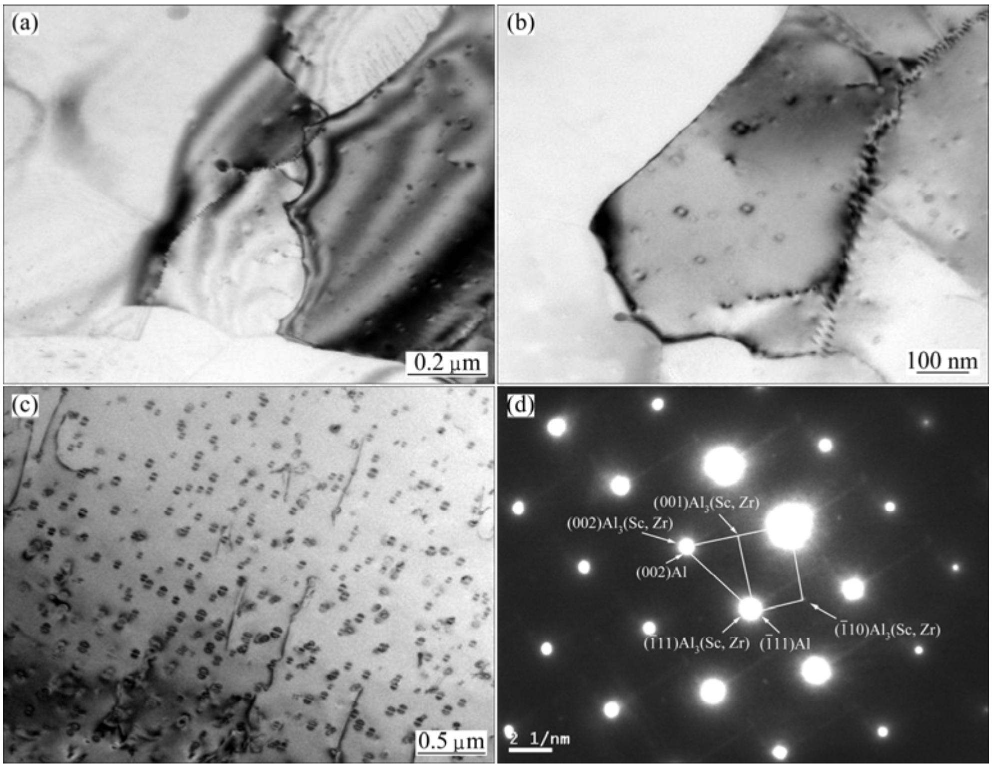

Fig. 5 TEM images of Al_3(Sc, Zr) particles annealed at various annealing temperatures: (a), (b) 350 °C for 1 h; (c) 580 °C for 1 h; (d) $[110]_{Al_3(Sc,Zr)}//[110]_{Al}$ zone axis SAD pattern corresponding to interface

4) The nucleation mechanism of recrystallization of Al−5.8Mg−Mn−Sc−Zr alloy is sub-grain coalescence and sub-grain growth.

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Al−5.8Mg−Mn−Sc−Zr 合金的再结晶行为

王 迎[1]，潘清林[1,2]，宋艳芳[1]，李 晨[1]，李智凤[1]，陈 琴[1]，尹志民[1]

1. 中南大学 材料科学与工程学院，长沙 410083；
2. 中南大学 有色金属材料科学与工程教育部重点实验室，长沙 410083

摘 要：采用活性熔剂保护熔炼，水冷铜模激冷铸造技术制备了 Al−5.8Mg−0.4Mn−0.25Sc−0.1Zr(质量分数，%)合金板材。通过显微硬度测试、光学显微镜和透射电镜观察等分析手段研究了该合金的再结晶温度和再结晶形核机制。结果表明：通过添加微量的 Sc 和 Zr 元素使 Al−Mg−Mn 合金的抗再结晶能力得到显著提高，即添加了 Sc 和 Zr 的 Al−Mg−Mn 合金再结晶温度(450 °C)比没有添加的合金(150 °C)的高。Al−Mg−Mn−Sc−Zr 合金的再结晶温度较高的主要原因是细小、弥散的 $Al_3(Sc,Zr)$粒子对位错和亚晶界的钉扎作用。合金的再结晶形核机制为亚晶合并和亚晶长大的双重机制。

关键词：Al−Mg−Mn−Sc−Zr 合金；再结晶；$Al_3(Sc,Zr)$粒子；形核

(Edited by Hua YANG)