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Microstructure stability of cold drawn AZ31 magnesium alloy during annealing process

SUN Hong-fei, CHAO Hong-ying, WANG Er-de
School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, China

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Abstract: The microstructural evolution of heavily cold drawn AZ31 magnesium alloy wires was investigated during a wide range of annealing temperature from 200 to 450 °C. The results show that the mean grain size of the as-annealed material is sensitive to the annealing temperature and cold drawn area reduction. Upon annealing symmetrical grain growth takes place in the AZ31 wires of cold drawn area reduction 12.2% under all the investigated annealing conditions, while three different stages of grain size refinement, normal grain growth and abnormal grain coarsening occur gradually in the materials with cold drawn area reduction 23.0%−60.5% with annealing temperature increasing. The increase of cold drawn reduction leads to the decrease of critical annealing temperatures for the three periods, and also enhances the start of abnormal growing grains preferentially in the severely drawn materials from the outer surface where larger amount of shear deformation constitutes the driving force for growth.

Key words: AZ31 Mg alloy; cold drawing; annealing; static recrystallization; grain growth

1 Introduction

The process of annealing treatment is always applied on plastically deformed metals and alloys in order to soften and restore the ductility and formability of materials [1−2], hardened by low temperature deformation or by the uncompleted dynamic recrystallization at moderate temperatures. Numbers of influential factors act on the microstructure evolution and the consequent properties of annealed materials, such as the history of deformation and the annealing parameters. As recrystallization after deformation is the only method to create a fully new grain structure with a modified grain size and mean orientation/texture, investigations of annealing behaviors of severely deformed metals are of concern during largely extended annealing processes.

For Mg alloys, as a consequence of the limited formability at room temperature, studies on the recrystallization have mainly focused on the hot-deformed samples where recovery and recrystallization have been active [3−4]. To understand the recrystallization mechanism and obtain favorable annealing technique, alloys need to be deformed at sufficiently large strains and possibly low deformation temperatures. SU et al [5] recently studied the annealed microstructure evolution of AZ31 Mg alloy with a maximum deformation strain of 0.5 obtained via uniaxial compression test at room temperature. The pressed materials were subjected to relatively expanded annealing temperatures up to 400 °C, as a result, the kinetics of recrystallization and grain growth were described in detail. Texture evolution of AZ31 and AZ61 Mg alloys during severe annealing process at 450−520 °C was analyzed by means of EBSD by PÉREZ-PRADO and RUANO [6−7]. Abnormal grain growth was observed in both alloys which took place from the outer surface to the mid-layer of the alloy sheets, leading to a homogeneous $\{11\bar{2}0\}$ texture throughout the thickness. Among the above and other literatures [8−10], annealing behaviors of deformed Mg alloys were discussed on the moderately deformed materials at moderate or high temperatures. The thorough presentation on recrystallization for a sufficiently highly cold deformed Mg alloys are rarely found. In this work, various cold drawn area reductions from 12.2% to 60.5%, which were equivalent to true strains of 0.13−0.93, were developed in AZ31 Mg alloy during a traditional drawing processing at room temperature. On the basis of the severely drawn materials, better understanding of the deformed microstructure stability in the heating process becomes possible at broad annealing temperatures ranging from 200 to 450 °C.

Corresponding author: CHAO Hong-ying; Tel: +86-451-86417629; Fax: +86-451-86412064; E-mail: hychao@hit.edu.cn

2 Experimental

An Mg−3.28Al−0.46Zn−0.27Mn (mass fraction, %) (AZ31) alloy in a post-extrusion annealed state with an initial diameter of 1.75 mm was subjected to a traditional drawing processing at room temperature. In this procedure, the sample was drawn through seven passes with a diameter reduction of about 0.1 mm each pass. Several cumulative area reductions were therefore obtained such as 12.2%, 23.5%, 42.2%, 53.0% and 60.5%. It is equivalent to true strains of 0.130, 0.270, 0.550, 0.755 and 0.930, respectively. These samples were then annealed at a selected series of constant temperatures ranging from 200 to 450 °C during various holding time from 600 to 10 000 s. The heat treatments were conducted in an electric furnace with ±2 °C accuracy and then cooled in air.

After annealing, samples were cut for optical microscopy on Olympus PMG3 microscope and the microstructure observation planes were aligned along the drawing direction. The grain sizes of all examined specimens were measured using a mean linear intercept method. Method for the metallographic examination is everywhere [3, 10].

3 Results

As shown in Fig. 1, the microstructure of the as-received AZ31 alloy is formed by equiaxed, recrystallized grains with a mean grain size of 3.7 μm. Twinning is absent in the microstructure observation.

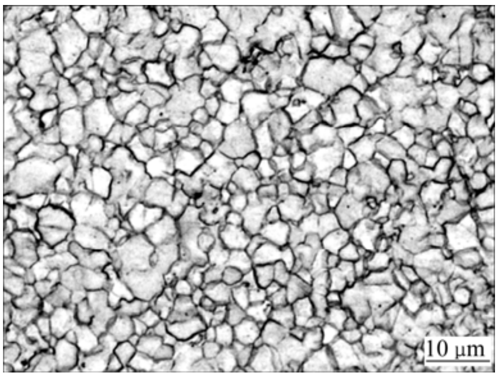

Fig. 1 Microstructure of as-received AZ31 Mg alloy wire

Annealing at the wide temperature range results in different effects on the microstructure evolution in the cold drawn AZ31 samples, including grain size refinement, normal grain growth and abnormal grain growth. Figure 2 shows the recrystallization characteristic of the 60.5% drawn sample during the isothermal annealing processing at 250 °C. Upon moderate annealing for only 10 min (Fig. 2(a)), a significant amount of small grains are formed and complete recrystallization takes place. The mean grain size is refined to 2.9 μm. As the annealing time is increased up to 60 min (Fig. 2(b)) and 120 min (Fig. 2(c)), normal grain growth occurs. However, the long annealing time is not as significant as the annealing temperature in the grain size coarsening. The mean grain sizes measured are 3.6 μm and 3.7 μm under both conditions of 60 min and 120 min, respectively, similar to the initial grain size of 3.7 μm. This also suggests that the recrystallization temperature for the 60.5% drawn AZ31 is lower than 250 °C during 30−60 min. Here, the usual definition of recrystallization temperature is the lowest annealing temperature when 95% and above

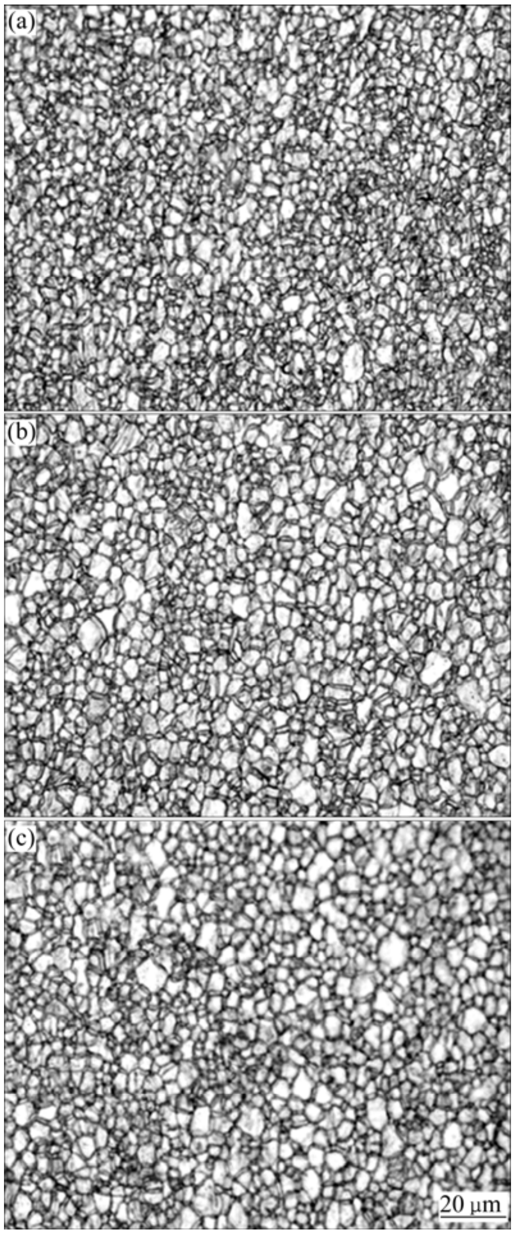

Fig. 2 Microstructure evolution of 60.5% drawn AZ31 sample at isothermal annealing temperature of 250 °C for various holding times: (a) 10 min; (b) 60 min; (c) 120 min

deformed microstructure is replaced by formation and migration of high angle grain boundaries. As the difficulty in accurately determining the frequency of high angle grain boundaries except using EBSD, a feasible and commonly used method is hardness measurement and/or microstructure observation. The latter is the choice in this work.

The effect of static annealing depends not only critically upon the annealing temperature but also on the conditions of the deformed materials such as the amount of cold deformation. Figure 3 illustrates the microstructure evolution of both 12.2% and 53.0% drawn samples during the isochronal annealing process at selective temperatures of 225, 400 and 450 °C for 10 or 30 min. At the lowest temperature of 225 °C for short holding time of 10 min (Figs. 3(a) and 3(b)), both the mean grain sizes are slightly coarser than that of the as-received material, together with the appearance of both equiaxed and elongated grains. This is attributed to the uncompleted recrystallization under such annealing conditions. It also suggests that the recrystallization temperature is around or above 225 °C for both drawn samples. However, the 53.0% drawn sample shows obviously refined microstructure in comparison with the 12.2% one, exhibiting greater potential in grain refinement than the slightly deformed one. It demonstrates that more nucleation has developed in the heavily drawn material at the same annealing state due to more plastic stored energy.

At higher temperature of 400 °C for 10 min, normal grain growth takes place in the 12.2% drawn sample, as shown in Fig. 3(c). However, there is a marked change in the microstructure evolution of the 53.0% deformed one, which is clearly divisible into areas of abnormal growing

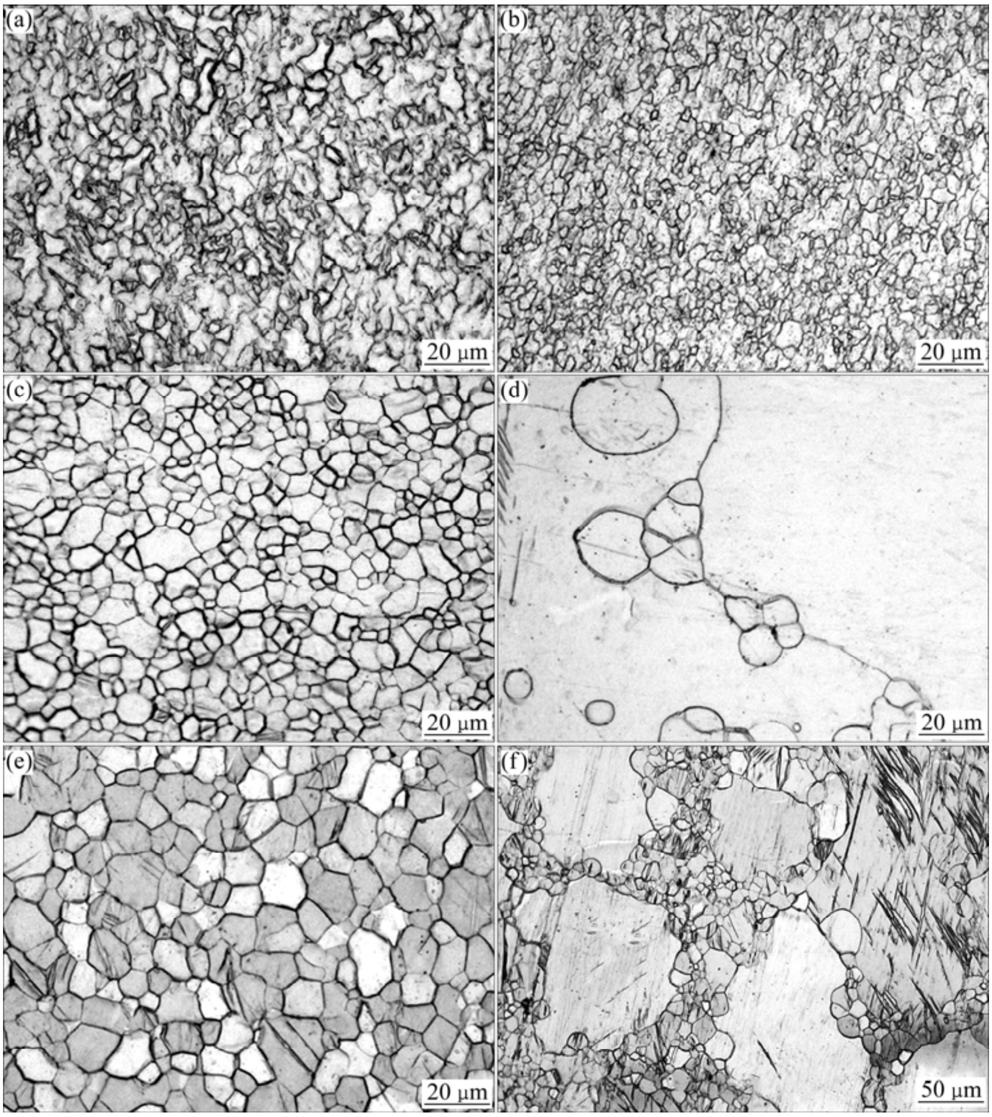

Fig. 3 Representative microstructures showing progress of three different recrystallization stages of drawn AZ31 Mg alloys during isochronal annealing process: (a) 225 °C for 10 min at 12.2% drawn area reduction; (b) 225 °C for 10 min at 53.0% drawn area reduction; (c) 400 °C for 10 min at 12.2% drawn area reduction; (d) 400 °C for 10 min at 53.0% drawn area reduction; (e) 450 °C for 30 min at 12.2% drawn area reduction; (f) 450 °C for 30 min at 53.0% drawn area reduction

grains and areas of refined recrystallized grains. An example of the abnormal grain with a size of about 200 μm is shown in Fig. 3(d). After a more severe heat treatment at 450 °C for 30 min (Figs. 3(e) and 3(f)), further grain coarsening is developed in the 12.2% deformed alloy but with a normal growing size, whereas largely coarse grains are observed to immerge into the matrix of fine grains in the highly drawn sample. A significant amount of twinning is seen in these abnormal growing grains but relatively less in the normal growing grains, which is in accordance with the observation in Ref. [6]. Microstructure analysis is also pointed out that abnormal grain growth is primarily developed in the outer surface of the highly deformed Mg alloys. As annealing exceeds, more large grains are formed at the expense of more neighboring small grains. Ultimately, large growing grains are seen in the mid layer of the entire sample. Such a process is shown in Fig. 4 in the 60.5% drawn material at high annealing temperature of 450 °C. PÉREZ-PRADO and RUANO [6] also reported that secondary recrystallization in the as-extruded AZ61 took place preferentially in the outer surface after a heat treatment at 450 °C for a relatively long holding time of 3 h and longer time of 22 h in the mid-layer. In their study, the initial material was in a hot-deformed state which was fully recrystallized with rather equiaxed grains, while the as-received materials in this study were subjected to cold drawing process. Consequently, the

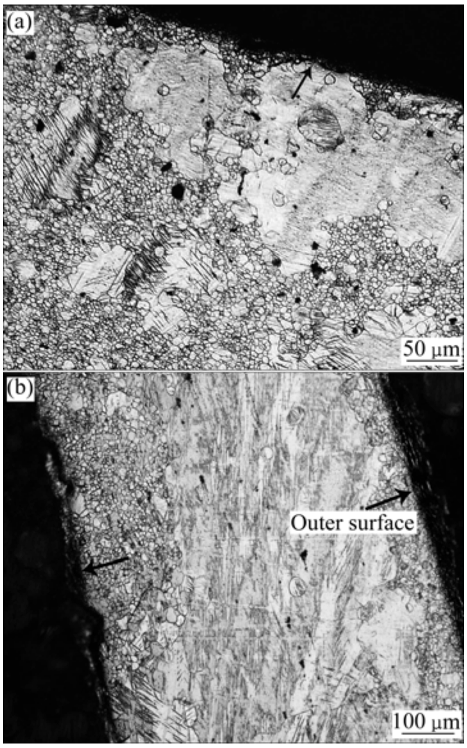

Fig. 4 Representative abnormal growing grains immerging from outer surface to mid-layer in 60.5% drawn AZ31 Mg alloy: (a) 450 °C for 10 min; (b) 450 °C for 2.8 h

significant amount of stored energy in the cold drawn AZ31 Mg alloys resulted in the decrease of annealing temperature and holding time for the subsequent abnormal grain growth. Although the stored deformation energy mainly promotes the onset of recrystallization and grain coarsening is driven by the reduction in grain boundary areas, here, the severely deformed samples still show higher tendency to grow quickly and largely than the slightly deformed ones as well as the hot-deformed ones.

The average grain size values as a function of cold drawn area reduction during the heat treatment are calculated and shown in Fig. 5. After the isochronal annealing of 10 min at temperatures lower than 300 °C (Fig. 5(a)), the mean grain sizes decrease gradually with increasing the drawn area reduction. However, only those heavily deformed samples with drawn area reductions over 40% exhibit refined recrystallized grain size below the initial value of 3.7 μm at the investigated annealing temperatures during 10 min. Such a drawn reduction of 40% seems higher than the so-called critical strain [11], below which only grain coarsening takes place while above which grain refinement is examined

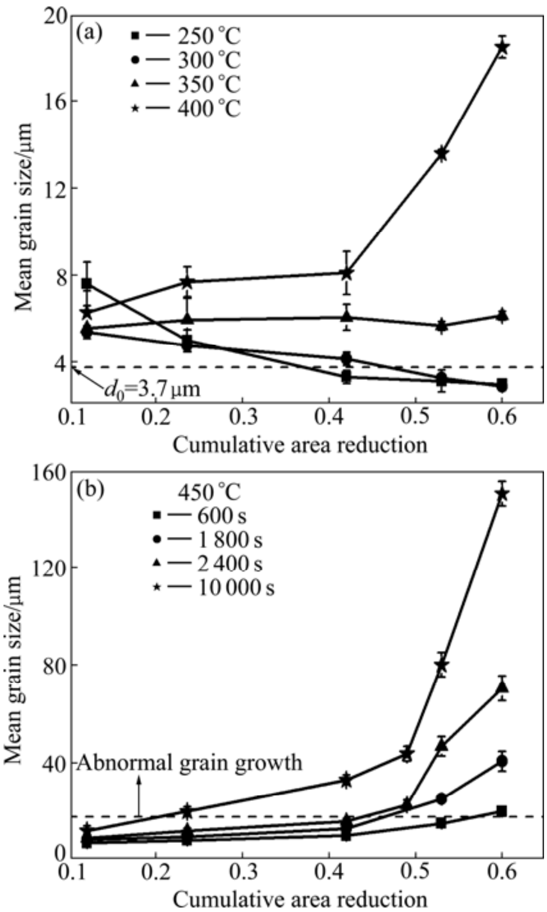

Fig. 5 Variation of average grain size as function of cold drawn area reduction in annealing process: (a) At temperatures ranging from 200 to 400 °C during 10 min; (b) At 450 °C in holding time range of 600−10 000 s

during the subsequent annealing process. Actually, the strain is still around 20%, since the 12.2% drawn sample shows a normal grain growth during all the studied annealing conditions and the coarsened grain size in the 23.5% drawn sample is ascribed to the mixture of both un-recrystallized and recrystallized grains during the short holding time of 10 min. If annealed at 350 °C for 10 min, about $0.7T_m$ (T_m=903 K, the absolute melting temperature of pure Mg), the grain size slightly increased at drawn reductions up to 42.2% followed by a decrease at 53.0% and then an increase at 60.5%. Such an unstable change is attributed to the competition of normal grain growth and abnormal growth, which are strongly dependent on the cold deformation level. For the samples annealed at 400 °C for 10 min, only increase in the mean grain size is observed, especially for the 53.0% and 60.5% drawn specimens, rapid increase is measured due to the widespread occurrence of abnormal grain growth.

Figure 5(b) shows the variation of mean grain size in the severely isothermal annealing process at 450 °C. Different from the observation in Fig. 5(a), no decrease in the grain size is measured. Instead, the mean grain size gently increase at short holding times up to 1 800 s and then increases rapidly. As the annealing time is further increased up to 10 000 s, the grain growth of the 12.2% drawn sample nearly ceases at a low level of approximately 20 μm irrespective of annealing time, whereas obvious grain coarsening is well developed in other samples. The ultimate grain size increases to 150 μm for the 60.5% drawn sample with some abnormal grains growing to 1−2 mm. That means nearly only one grain is present in some areas along the transverse section of the specimen since its diameter is 1.1 mm. The great increase of the mean grain size demonstrated that the deformed structure is very unstable at high temperatures around 450 °C, ultimately the deformed structure is fully removed and replaced by a mixture of equiaxed fine grain configuration and coarse grain configuration.

4 Discussion

4.1 Recrystallization temperature

Three different stages are observed in the microstructure evolution of the cold drawn AZ31 Mg alloys wires during the annealing treatments investigated. They are: the first stage of recrystallization at lower annealing temperatures with the recrystallized nuclei growing into the matrix of drawn materials, the second stage of normal grain growth at moderate temperatures which results in grain size coarsening with the annealing temperature, and the third stage of abnormal grain growth where a small number of grains grow abnormally at the expense of other small recrystallized grains. Both microstructure analysis and calculated mean grain size in Figs. 1−5 support to determine the recrystallization temperature as well as the critical grain coarsening temperatures since grain coarsening especially grains abnormally growing leads to significant decrease in the mechanical properties [4]. SU et al [5] studied the static recrystallization of cold compressed AZ31 Mg alloy and divided the annealing temperatures into three zones with upper and lower bounds to determine the recrystallization temperature. Below the lower bound recovery takes place in the pressed AZ31 Mg alloy, while above the upper bound the deformed materials undergo grain growth. The zone between the bounds represents the complete recrystallization area. Using the same classification method here, three different temperature zones related to the cold drawn area reductions of 12.2%−60.5% are developed according to the microstructure analysis in the present study, as illustrated in Fig. 6.

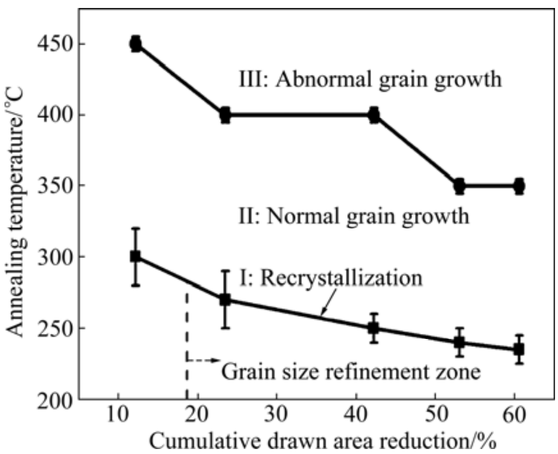

Fig. 6 Division of annealing temperatures during isochronal annealing (30−60 min) as function of cold drawn area reduction

To estimate the critical temperatures accurately together with the obtained results, another annealing treatments at 225 and 240 °C for 60 min were applied on the 60.5% deformed sample. The results show essentially the same duplex structure of both elongated and recrystallized areas as the 12.2% and 42.2% drawn materials at 225 °C but full recrystallization at 240 °C. It turns easy to conclude that 230−240 °C is appropriately the optimum recrystallization temperature range for the 60.5% drawn sample to develop refined microstructure. Combined with the present results, it is clear that the critical recrystallization temperature increases with the amount of cold deformation up to the maximum value of 300 °C. However, it is noted that grain refinement only occurs in the severely drawn alloys above the critical strain. When annealing at 350 °C for 10 min, the first abnormal grains are observed in the 53.0% and 60.5%

cold drawn samples, above 400 °C for 10 min abnormal growing grains appear in samples with a drawn reduction of 42.2%. Similarly, the temperature increases with the decrease of the cold drawn deformation. For the 12.2% drawn sample, just normal grain coarsening, with one or two abnormal growing grains in small size, is observed during 40 min at 450 °C as shown in Fig. 5(b). Such temperature division is made to help draw up the annealing schedule and control the annealing processing of severely cold deformed AZ31 Mg alloys especially in the industrial application.

4.2 Effects of drawn deformation and surface on grain growth

Effects of annealing temperature and annealing time on the microstructure evolution and mean grain size of hot-deformed Mg alloys have been investigated everywhere [3−5, 8]. Kinetics of recrystallization and normal grain growth has also been studied and well understood. However, relatively few studies have systemically investigated the effects of heat treatments on the highly cold deformed Mg alloys during expanded annealing temperatures and annealing time. PÉREZ-PRADO and RUANO [6−7] found that abnormal grain growth was developed under severe annealing conditions of (520 °C, 19 h) and (520 °C, 3 h) in hot-extruded AZ31 and AZ61 Mg alloys, respectively. Compared with the observed results in the present study, the enhanced annealing temperature and holding time in their studies are due to the dynamic recrystallized microstructures, which release the stored energy and lead to the low mobility of the grain boundaries. In other words, the deformed materials in cold drawn states store more plasticity deformation energy, particularly those heavily drawn samples with highly misoriented regions. The presence of large amount of stored plastic energy could constitute the driving force for the abnormal grain growing, since this deformation energy is released in three main process including recovery, recrystallization and grain coarsening [1]. The highly misoriented regions induced by the severe deformation supply the favorable boundaries to grow abnormally because the high angle boundaries have higher energy and mobility than the lower angle boundaries [7]. Therefore, both the recrystallization temperature and the abnormal grain growing temperature is lower in highly drawn samples than in slightly drawn alloys as well as in hot-deformed alloys.

Then here comes a question, naturally, why these grains are inclined to develop from the outer surface to the mid-layer in samples with the same drawn reductions. Some researchers [12−13] reported that during the secondary recrystallization process, the abnormal growing grains must compete with the grain growth, otherwise these grains require a mobility or energy advantage. RIOS and GOTTSTEIN [14] reported that the abnormal growing grains must be reasonably large or it will be consumed; or its boundary must be a fast moving boundary and it must be in a favorable environment. As a consequence, the surface is the best region for the small fraction of grains to develop preferentially and successfully. It should be noted that, in the drawing process, the surface is the severest deformed region where the defects and dislocation density are higher than that in the mid-layer. During recrystallization, sufficient thermal energy is supplied to allow the dislocations to rearrange themselves into lower-energy configurations. After that, the residual distortion energy stored in the deformed grains in the outer surface provides the driving force for grain growth. So, given the grain sizes of two adjacent grains in the surface and mid-layer are i and j, respectively, then the velocity of a boundary between grains of type i and j can be given by the following equation [14]:

$$V_{i-j}=k_{i-j}(M_i-M_j)(\gamma_i-\gamma_j) \tag{1}$$

where k_{i-j} is the grain boundary curvature; γ_i and γ_j are the interfacial free energy per unit of area in i and j grains. Since the residual distortion energy stored in the grains close to the surface is more than that in the mid-layer, then $\gamma_i>\gamma_j$. M_i and M_j are the boundary mobility of grains i and j. So, only when $M_i>M_j$ can $V_{i-j}>0$ come true. Then the grain i is possible to immerge into grain j.

Here, it has been confirmed that the texture component $\{11\bar{2}0\}$ parallel to the deformed orientation predominates in the abnormal grain growth of Mg−Al−Zn alloys with a strong basal texture. In particular, the intensity strength of $\{11\bar{2}0\}$ component in the outer surface is higher than that in the mid-layer where more random texture is present [6−7]. KIM et al [15] verified that in the grain growth of severely annealed Al alloy the abnormal growing grains were clustered in a pronounced cube recrystallization texture, while a random recrystallization texture prevailed in the normal growing grains. That is to say, the discrepancy of texture components between the grains in the outer surface and in the mid-layer partly determines the abnormal or normal growth in these grains, together with the driving force partly provided by the presence of large amounts of deformation in the outer surfaces. Therefore, the abnormal growing grains in a favorable environment like the outer surfaces would grow more quickly.

5 Conclusions

1) The annealing behavior was studied at temperatures ranging from 200 to 450 °C in cold drawn

AZ31 Mg alloys at area reductions of 12.2%−60.5%, which were equivalent to the true strains of 0.13−0.93. It was found that the mean grain sizes of the annealed alloys were refined with the drawn area reduction increasing and the annealing temperature decreasing in the range of 200−300 °C.

2) Upon moderate annealing at temperatures of 300−400 °C normal grain growth developed in all the cold drawn samples. When annealing up to 400 °C, abnormal grain growth started in the outer surfaces of the severely deformed material firstly and then in the slightly deformed materials with the extension of annealing time and temperature. However, no abnormal growing grains were observed in the 12.2% drawn AZ31 until severely annealing at 450 °C for 2 400 s.

3) The annealing process is divided into three different stages in the form of heated temperature: recrystallization at 200−300 °C, normal grain growth at 300−400 °C and abnormal grain growth over 400 °C, which is significantly dependent on the prior cold drawn deformation. It is the anisotropy of strain energy and texture effect that results in the abnormal growing grains emerging and then growing from the outer surface into the mid-layer in the deformed materials.

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冷拉拔 AZ31 镁合金在退火过程中显微组织的稳定性

孙宏飞，晁红颖，王尔德

哈尔滨工业大学 材料科学与工程学院，哈尔滨 150001

摘　要：研究冷拉拔 AZ31 镁合金线材在退火温度为 200～450 °C 范围内显微组织的演变。研究结果表明：退火态材料的晶粒尺寸对退火温度和冷拉拔面积的减少敏感。在所有退火条件下，冷拉拔面积减少 12.2%时的 AZ31 镁合金丝材晶粒尺寸均匀长大，而晶粒的 3 个不同变化过程：晶粒细化、正常长大和异常粗化，会随着退火温度的升高和冷拉拔面积减少(23.0%～60.5%)时逐步发生。随着拉拔面积的逐渐减少，3 个阶段的临界退火温度降低，同时优先提高材料外表面剧烈拉拔部位异常长大晶粒的起始温度，在材料外表面大量的剪切应变构成了晶粒长大的驱动力。

关键词：AZ31 镁合金；冷拉拔；退火；静态再结晶；晶粒长大

(Edited by YANG You-ping)