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Production of Mg-Al-Zn magnesium alloy sheets with ultrafine-grain microstructure by accumulative roll-bonding

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Abstract: Accumulative roll-bonding (ARB) was applied to Mg-Al-Zn magnesium alloy sheets to prepare ultrafine-grain microstructure. Significant grain refinement is achieved after three cycles of ARB with average grain size of about 1.3 μm. The microstructure is characterized by nearly uniform ultrafine equiaxed microstructure without twins. The evolution of the misorientation distribution during ARB was measured by EBSD. Grain refinement can be contributed to the grain subdivision induced by severe accumulated strain, the accumulated strain enhanced concurrent dynamic recovery and recrystallization as well as the complicated distribution of interface and shear strain during ARB.

Key words: Mg-Al-Zn magnesium alloys; microstructure; grain refinement; dynamic recrystallization; misorientation

1 Introduction

Magnesium alloys are promising structural materials due to their excellent properties, such as low density weight, high specific strength and specific stiffness, as well as machinability and recyclability[1]. However, Mg alloys often exhibit poor ductility and workability, and this limitation reduces their application in engineering components[2]. In practice, useful improvements in ductility are observed at elevated temperatures, with finer grain sizes and solute additions[3]. Grain refinement is a promising technique for improving the strength and ductility of metal materials due to the strong influence of grain size on the mechanical properties. It is well established for wrought magnesium alloys that decreased grain size could increase the ductility[3].

Severe plastic deformation (SPD) techniques emerged in the last decade as effective methods for the production of bulk metallic materials with very fine grains[4]. Among these processes, accumulative roll-bonding (ARB) process developed by SAITO et al[5] is appropriate to manufacture nanocrystalline and ultrafine grained sheets and plates which are most widely used material shapes in the commercial and industrial fields[6]. The ARB process was proved to be very effective in refining grains and enhancing the strength of aluminum, steels and copper[7−8]. However, the ARB was used only for cubic materials and rarely for HCP structured metal. PEREZ-PRADO et al[9] used accumulative roll-bonding to produce AZ31 sheets with grain size of 3 μm. del VALLE et al[10] found that the ultimate grain size was achieved during ARB of AZ61 and the degree of bonding depended on the rolling temperature and the thickness reduction per pass. The information available in the literatures about the microstructural change of magnesium alloys during the ARB process is still very limited.

Therefore, it is necessary to conduct investigation on the microstructure evolution of magnesium alloys during ARB process. By using the ARB process, fine-grained AZ31 sheets with an average grain size of 2.4 μm and an elongation of 19.6% at room temperature were fabricated[11]. The objective of the present work is to obtain a better understanding of the microstructure development during the ARB process.

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2 Experimental

The material used in the current study was magnesium alloy AZ31 sheet (Mg-3%Al-1%Zn) with a thickness of 1.5 mm, prepared by hot rolling at 300 °C. In the accumulative roll bonding process, two pieces of the strip with dimensions of 200 mm×75 mm×1.5 mm were stacked and rolled with a 50% reduction ratio. Then, the ARB-processed sheet was cut and stacked to the initial thickness and the stacked pieces were rolled again with the same reduction ratio. Such procedure was repeated for up to 3 cycles. Before each ARB cycle, the stacked sheets were heated at 300 or 400 °C for 10 min in an electrical furnace. ARB was performed at the rolling machine furnished with two rolls with diameter of 400 mm and rotated at 23 r/min. The sheets were hereafter considered as ARB1, ARB2 and ARB3 after one, two and three ARB cycles, respectively.

Microstructural examination was conducted on as-rolled and as-ARB processed specimens by optical microscopy and electron backscattered diffraction (EBSD). The longitudinal cross sections normal to the transverse direction were observed. Samples for metallography were ground with 1200 grit SiC paper. Polishing was done with diamond polishing through 6, 3 and 1 μm and then etched with a solution of 6 g picric acid, 100 mL ethanol, 5 mL acetic acid and 10 mL water for 10−30 s. The specimen for EBSD over the sample thickness was prepared by mechanical polishing with 2000 grit silicon carbide abrasive paper followed by electro-polishing using a solution comprising 10% $HClO_4$+90% ethanol (volume fraction) at −35 °C to eliminate the residual stress of the surface layer. EBSD mappings were studied using scanning electron microscope (FEG-SEM, Nova Nano430) equipped with electron back scattered diffraction. Microstructure maps derived from EBSD show the distribution of grain size (based on misorientation angle, i.e. grains and sub-grains) which was estimated using intercept method.

3 Results and discussion

3.1 Microstructure after ARB

Figure 1 shows optical microstructures of the AZ31 specimen before ARB (Figs.1(a) and (b)) and ARB-processed for various cycles (Figs.1(c)−(e)).

The as-rolled AZ31 samples contained small and equiaxed recrystallized grains with size of about 1 μm and elongated grains with length of 10−17 μm, as well as some fraction of twins (as shown in Fig.1(a)). Figure 1(b) shows the microstructure of annealed AZ31 sheet samples. The grain shape in this sample is almost spherical and the twins disappear.

The microstructures of the specimens that were produced by one to three cycles ARB are illustrated in Figs.1(c)−(e). As can be seen from Fig.1(c), fine and reasonably equiaxed grains are developed after one cycle ARB. The microstructure of the ARB processed AZ31 alloy after one cycle shows a bimodal grain size distribution consisting of a large fraction of the newly formed grains with very small grain size and a small fraction of the non-fully fragmented original grains. However, although the grain refinement is evident, the microstructure is far from homogeneous. As the number of ARB cycles increases, the microstructures become finer. The specimen processed by 3 cycles ARB is almost covered with ultra-fine grains of 1.317 μm in average diameter. Larger average grain size is obtained as the annealing temperature increases from 300 to 400 °C. Once a critical minimum grain size is achieved, the subsequent cycles do not have any important refining effects. This is attributed to the incomplete dynamic recrystallization followed by grain growth due to the residual accumulated strain energy. An interesting feature of the grain structure in Fig.1 is that there are no twins present in the ARB processed samples.

3.2 Grain size

Experimental results show that the crystallite size does not change significantly by further deformation after three cycles ARB. The histograms of average grain size distribution of AZ31 magnesium sheets during ARB are illustrated in Fig.2. The distribution of grain size for as-rolled samples is widespread due to the inhomogeneous plastic deformation at microscopic level (Figs.2(a) and (b)). The mean grain size $\overline{d}$ decreases from 4.127 μm of the initial material to 1.317 μm after 3 cycles of ARB at 300 °C. By increasing the ARB process strain, homogenous distribution of plastic strain in the bulk of samples is attained and the distribution of grain size becomes narrow (Figs.2(c)−(e)). It shows that the ARB process is a reproducible process for grain refinement and can successfully produce AZ31 sheets with ultra-fine grains. It is also apparent that ARB is especially effective in reducing the grain size when the rolling is conducted at low temperatures, which limits the occurrence of grain growth.

3.3 Grain boundary and misorientation

Figure 3 represents the boundary misorientation maps recorded on the longitudinal cross sections normal to the transverse direction of the samples deformed by ARB. In these figures, high angle grain boundaries (HAGB) with misorientations larger than 15° are drawn in bold lines, while low angle grain boundaries (LAGB) with misorientations between 2° and 15° are drawn in thin lines.

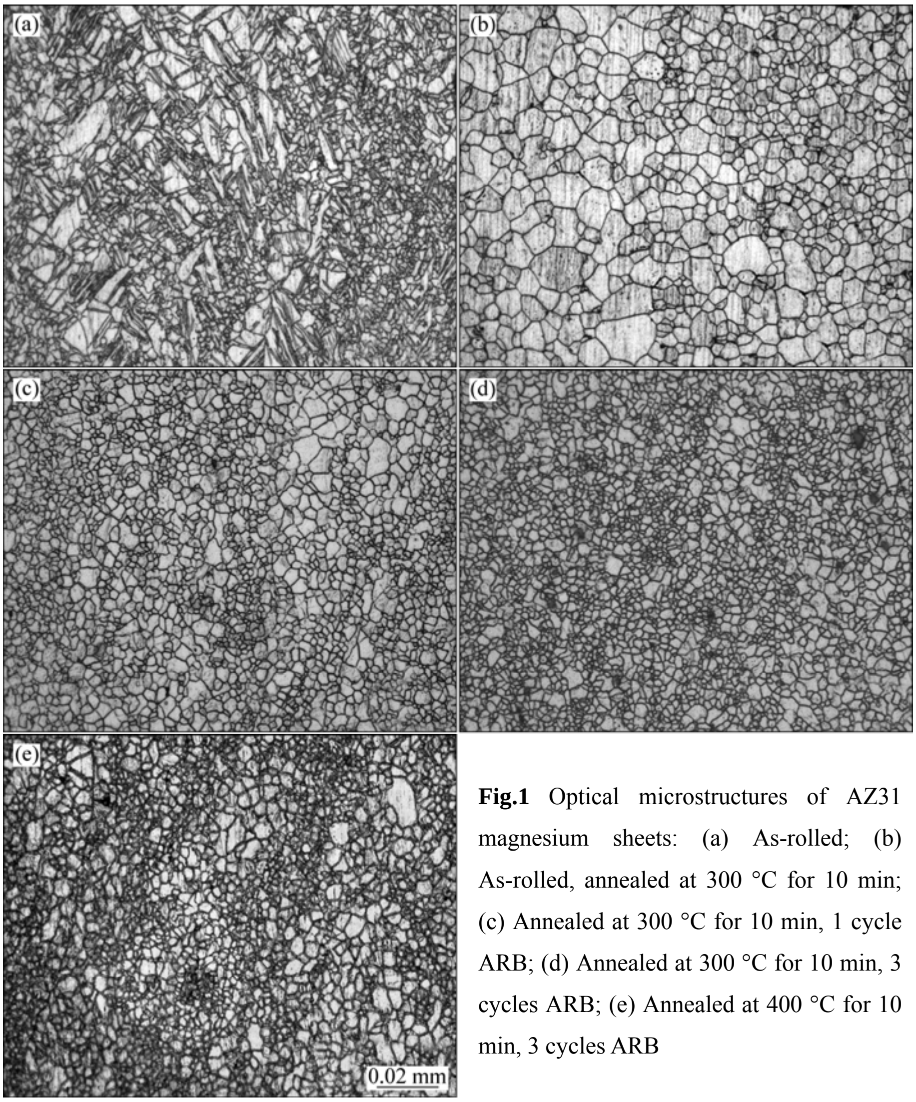


Fig.1 Optical microstructures of AZ31 magnesium sheets: (a) As-rolled; (b) As-rolled, annealed at 300 °C for 10 min; (c) Annealed at 300 °C for 10 min, 1 cycle ARB; (d) Annealed at 300 °C for 10 min, 3 cycles ARB; (e) Annealed at 400 °C for 10 min, 3 cycles ARB

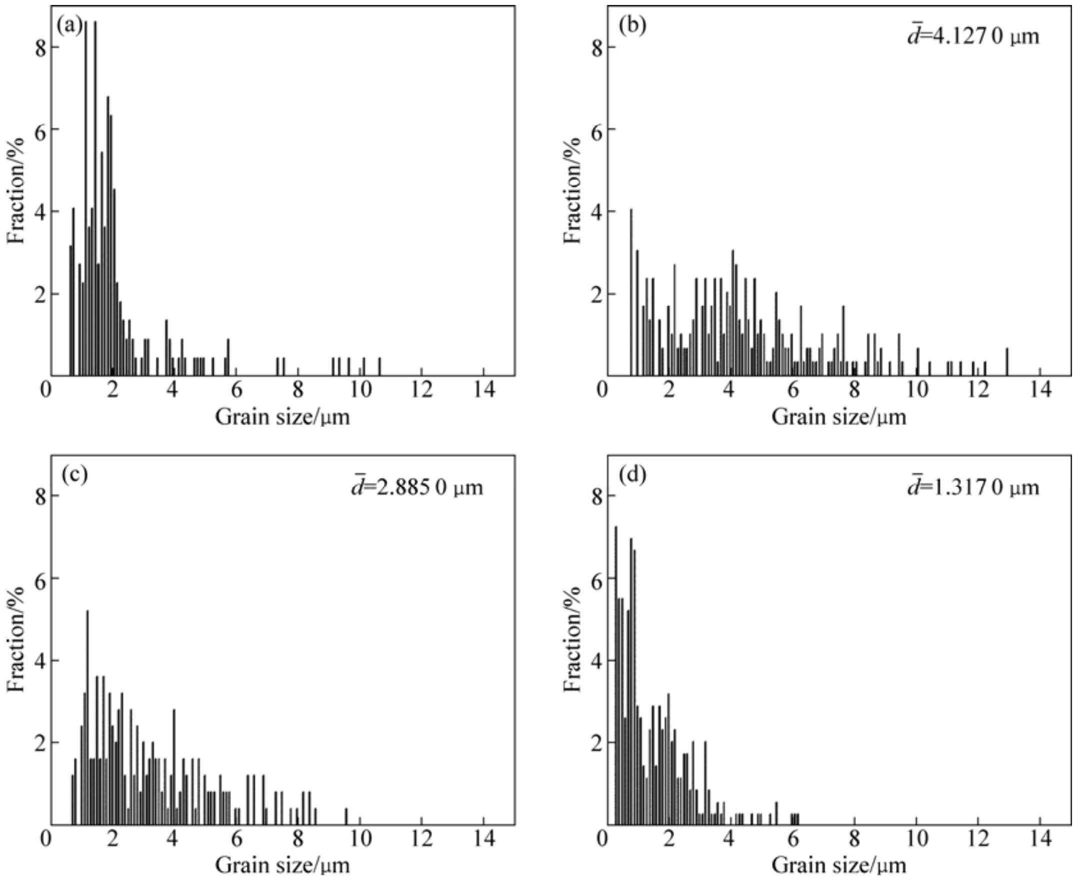
As seen from Fig.3(a), grain boundaries for as-rolled sample after annealing at 300 °C for 10 min are almost high angle grain boundaries. The ARB deformation introduces severe plastic deformation into the AZ31 sheets, therefore, grains with low angle grain boundaries are observed as the typical expected microstructure after one cycle of ARB. The number of ultra fine grains (UFGs) with high angle grain boundaries increases with increasing ARB cycles up to three. The microstructure after three cycles of ARB consists of the following two components: 1) grains surrounded by clear high angle boundaries with misorientations larger than 15° and 2) sub-grains bounded by low angle boundaries with misorientations smaller than 15°.

3.4 Microstructure evolution

The ARB process has been applied mainly to metals and alloys with cubic crystal structure, but the study on the number of ARB processes to hexagonal materials is limited. It is well known that the deformation behaviors of hexagonal metals are significantly different from those of cubic metals. The number of active slip systems is limited, and sometimes the deformation twining plays an important role in plastic deformation[12]. Thus, it is expected that the microstructure evolution during ARB is different between cubic and hexagonal metals.

In the ARB process of this work, the evolution of microstructure in the longitudinal cross sections normal to the transverse direction of AZ31 sheets evolved in the following way with increasing number of cycles:

1) Fine and reasonably equiaxed grains are developed after one cycle of ARB, whereas the microstructure of sample ARB1 is far from homogeneous. A large fraction of subgrains network with low angle grain boundaries are created; 2) The grain structure is gradually refined with increasing strain and the misorientation of the early low angle boundaries



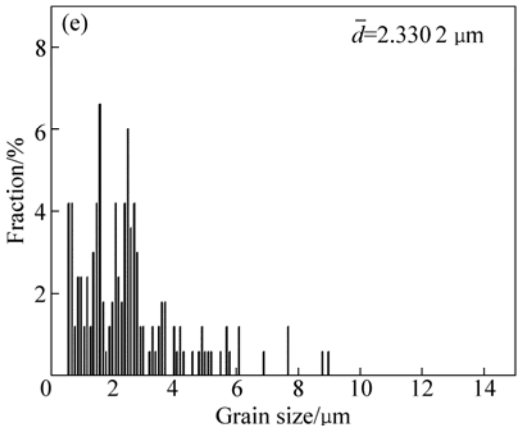


Fig.2 Histograms showing distribution of grain sizes of AZ31 magnesium sheets: (a) As-rolled; (b) As-rolled, annealed at 300 °C for 10 min; (c) Annealed at 300 °C for 10 min, 1 cycle ARB; (d) Annealed at 300 °C for 10 min, 3 cycles ARB; (e) Annealed at 400 °C for 10 min, 3 cycles ARB

increases; 3) Well-defined grain structure with sharp high-angle boundaries are formed. The microstructure of ARB processed AZ31 sheets are characterized by equiaxed grains without twins.

It is of interest to note that the twins in the initial sample disappear in the ARB processed AZ31 sheets (see Fig.1). It is well known that five independent slips are necessary for homogeneous plastic deformation in a polycrystalline material[13], which is called the von Mises criterion. The main slip mode in magnesium and its alloys is basal slip. Only two independent slip systems are available in the basal plane. The role of twinning in the deformation of magnesium alloys is vital as a complementary deformation mechanism to provide additional independent slip systems. Twin nucleation decreases with decreasing grain size because the grain size dependence of the twinning stress is greater than that of the slip stress[14]. It is suggested that non-basal slips are activated for a fine-grained Mg, resulting in suppression of twin generation and reduction in mechanical anisotropy by grain refinement[15]. Grain size could be critical for twinning in magnesium alloy. As the grains become finer, twinning is increasingly inhibited. Deformation twins disappear when the average grain size reaches 2−3 μm[16]. As seen from Fig.2, the average grain size of AZ31 in our research is 1–3 μm after ARB. It is grain refining that inhibits twinning during ARB.

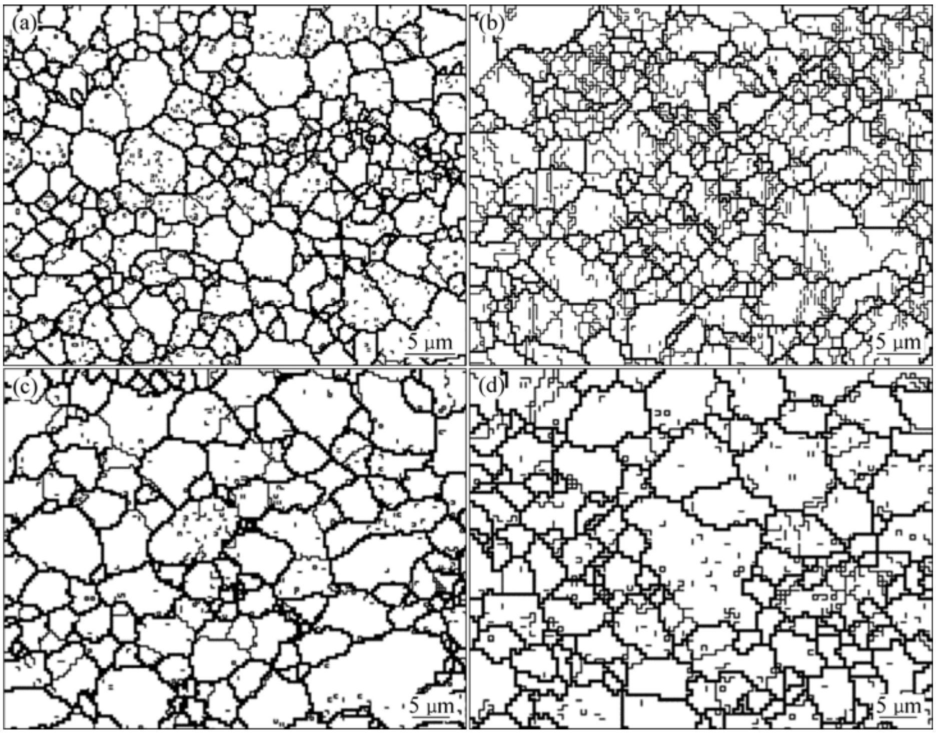


Fig.3 Grain boundary maps of AZ31 magnesium sheets: (a) As-rolled; (b) As-rolled, annealed at 300 °C for 10 min; (c) Annealed at 300 °C for 10 min, 1 cycle ARB; (d) Annealed at 300 °C for 10 min, 3 cycles ARB; (e) Annealed at 400 °C for 10 min, 3 cycles ARB

3.5 Grain refinement

The mechanism of the UFGs formation during ARB is still an issue under discussion. Refining grain size is seen during ARB in various materials[17]. One of the ideas to explain the microstructure evolution during ARB is grain subdivision[12]. The grain subdivision is a process in which geometrically necessary boundaries induced by deformation sub-divide the original crystals. A detailed EBSD analysis was performed to gain information on the orientation of the ARB processed AZ31 sheets. The misorientation distributions across individual grains in the samples shown in Fig.3 are depicted in Fig.4. The results show that the AZ31 sheets processed by ARB have severely strained structures with large local misorientations. In the first cycle, a considerable fraction of grain boundaries have misorientation angle less than 15°. As the cycles increase, the continuous changes in misorientation are converted into the planer boundaries by rearrangement of the geometrically necessary boundaries with short-range diffusion[18]. The short-range diffusion of AZ31 sheets during ARB is possibly enhanced due to the temperature rise by severe plastic strain and the friction between roll and surfaces of the samples. The fraction of high angle grain boundaries increases with increasing strain in the specimen processed by three ARB cycles. The misorientation profiles plotted in Fig.4(a) confirm the increase of the HAGBs in expense of the LAGBs. In the end, a number of high angle grain boundaries are

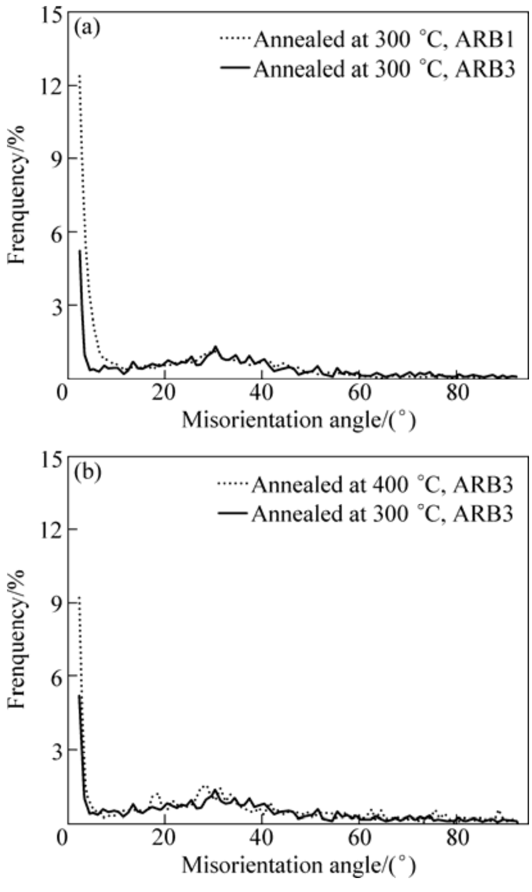


Fig.4 Misorientation distributions in samples processed by different ARB cycles

introduced to form UFG microstructures. Hence it could be considered that the grain subdivision by transition of low angle to high angle grain boundaries induced by severe accumulated strain is one of the mechanisms which result in fine equiaxed fine structure.

Lamellar structure with elongated grains is observed as the typical expected microstructure for AA1100 aluminum sheet processed by ARB[19]. However, Fig.1 shows that the grains are fine and reasonably equiaxed and homogeneously distributed for the ARB processed AZ31 samples, which suggests that recrystallization occurs during ARB. The micrographs in Fig.1 show clear evidences of dynamic recrystallization (DRX). One reason is the nearly spherical grain shape, which should be very much elongated due to large shear during ARB. Also, the forms of the grains are not always convex, which is a clear indication of the occurrence of DRX.

During hot rolling, AZ31 alloy is liable to undergo recrystallization, i.e. dynamic recrystallization (DRX) affects the microstructure, crystallographic texture and thus the material anisotropy. The onset of dynamic recrystallization for magnesium alloys is about 200 °C[20]. As for AZ31 magnesium alloy, if enough slip systems can be activated in a particular grain during rolling, as the case in the present study, the original grains subdivide homogeneously, and with increasing deformation, they become fully divided in smaller grains. This is one of the grain refinement mechanisms during ARB, i.e. accumulated strain induces grain refinement. On the other hand, in grains that are not favorably oriented for slip, large stresses accumulate along grain boundaries during ARB and thus small grains appear near grain boundary areas by another continuous process, rotational recrystallization (RRX). It is the accumulated strain that promotes and enhances the dynamic recrystallization in AZ31 alloy during ARB. The ratios of recrystallized, substructured and deformed microstructures for samples of as-rolled, ARB processed at 300 and 400 °C were measured by EBSD and shown in Fig.5, respectively. As can be seen, the fraction of recrystallized microstructure increases greatly after deformation by ARB. Concurrent dynamic recovery and recrystallization lead to the formation of ultra-fine microstructure. Our investigations suggest that the formation process of the UFGs is continuous recrystallization. Concurrent recovery and recrystallization processes transform subgrains with low angle boundaries into grains with high angle boundaries.

The redundant shear strain due to friction between the rolls and the specimen plays an important role in grain refinement by ARB process. Figure 6 shows the microstructure near the surface regions for sample ARB1, showing grain refining induced by shear strain. It is evident that the grains near surface are smaller than those far away from the surface. Deformation during rolling is strongly affected by the friction between the rolls and the work piece, which induces redundant shear strains. The ARB process is usually conducted without lubricant to achieve bonding, so that large shear strains are introduced into the surface regions at each pass. When the 50% rolled sheet is cut and stacked between cycles, one of the surface which has undergone the severe shear deformation comes into the center. This would cause the latter undergo a plain strain deformation state in the next pass and results in complicated distribution of accumulated strains through the sample thickness[21−22]. Such a change in deformation mode (or strain path) may play an important role in the formation of the uniform fine grains during ARB process.

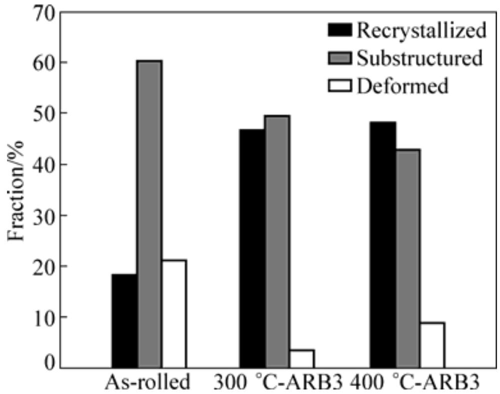


Fig.5 Areas fraction of recrystallized region for as-rolled and ARB processed samples

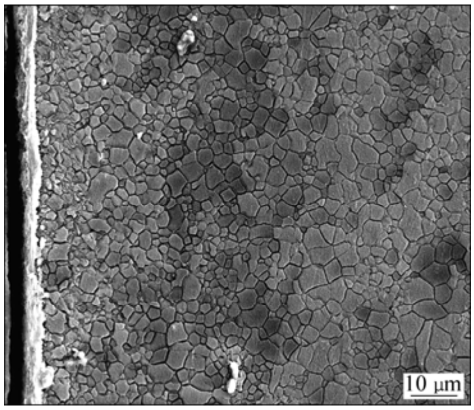


Fig.6 Microstructure near surface regions of ARB1 sample

4 Conclusions

1) ARB is an effective grain refinement method for producing AZ31 sheets with ultra-fine grain structure. Significant grain refinement is achieved after three ARB cycles with average size of about 1.3 μm.

2) The grain subdivision by transition of low angle

to high angle grain boundaries induced by severe accumulated strain is one of the mechanisms that result in the great grain refinement during ARB.

3) Concurrent dynamic recovery and recrystallization lead to the formation of ultra-fine microstructure. It is the accumulated strain that enhances the dynamic recrystallization in AZ31 during ARB.

4) The complicated distribution of interface and shear strain due to friction between the rolls and the specimens plays an important role in the formation of the uniform fine grains during ARB.

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累积叠轧焊制备超细晶组织 Mg-Al-Zn 合金薄板

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摘 要：采用累积叠轧焊(ARB)工艺制备超细晶组织 AZ31 镁合金薄板。实验结果表明，进行 3 道次 ARB 变形后，AZ31 板材晶粒显著细化，平均晶粒尺寸约 1.3 μm，呈等轴状，材料组织均匀，没有发现孪晶。采用 EBSD 技术观察组织演变和晶粒的取向差。ARB 变形过程中的晶粒细化可归因于累积应变诱导的晶粒细化、累积应变强化回复和再结晶以及 ARB 变形过程中复杂的界面和剪切应变分布。

关键词：Mg-Al-Zn 镁合金；组织；晶粒细化；动态再结晶；取向差

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