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ARTICLE

Grain Refinement and Texture Evolution of Mg-Gd-Y-Zn-Zr Alloy Processed by Repetitive Usetting-extrusion at Decreasing Temperature

Du Yue , Zhang Zhimin, Zhang Guanshi , Yan Zhaoming, Yu Jianmin

North University of China, Taiyuan 030051, China

Abstract: A homogenized Mg-13Gd-4Y-2Zn-0.5Zr (wt%) alloy was subjected to a repetitive upsetting-extrusion (RUE) process at decreasing temperature from 480 ºC to 370 ºC. The microstructure and texture development of the alloy during the RUE process was investigated. The results show that average grain size decreases with increasing cumulative strain and decreasing temperature. Uniform ultrafine-grain structure with an average grain size of 3.4 μm is achieved after 6 passes, i.e., cumulative strain of 8.4. Grain refinement is induced by a complicated combination of discontinuous dynamic recrystallization(DDRX) and continuous dynamic recrystallization(CDRX). Kink of lamellar long period stacking order (LPSO) phases play an important role in the grain refinement through subdividing the coarse grains by inducing recrystallization evolved in the kink bands. After 1 pass of RUE, a strong basal texture is obtained. With increasing RUE passes, the texture is gradually weakened, which results from cooperative effect of dynamic recrystallization(DRX) and the alternative change of loading directions (axial and radial) during RUE processing.

Key words: Mg-Gd-Y-Zn-Zr alloy; repetitive upsetting-extrusion (RUE); grain refinement; texture

Magnesium (Mg) alloys have great potential applications for aircraft, automotive and electronic industries due to their low density and high specific strength, good damping and casting capability[1,2]. However, it is necessary to enhance the mechanical properties of the Mg alloys in order to expand their application[3,4]. It is reported that the addition of rare earth (RE) elements to Mg alloys can bring about remarkable mechanical properties at both ambient and elevated temperatures and thus RE containing Mg (Mg-RE) alloys have been attracting much more attention[5,6]. Kawamura et al.[7] developed a rapidly solidified powder metallurgy (RS P/M) alloy $Mg_{97}Y_2Zn_1$, which exhibited an ultrahigh strength of 600 MPa at room temperature. The excellent properties are considered to be due to the ultra-fine grains with average grain size of 200 nm with a novel long period stacking ordered (LPSO) structure. Zhang et al.[8] produced the as-cast Mg-14Gd-3Y-1.8Zn-0.5Zr (wt%) alloy with high tensile strength of 366 MPa and fracture elongation of 2.8% through solution-treatment at 793 K for 10 h and aging at 498 K for 16 h. Thus, by controlling the microstructure to achieve fine grain size and homogeneous dispersion of LPSO phase, the mechanical properties of the Mg alloys can be significantly improved.

On the other hand, it has been proved that microstructure refinement and texture control was an effective way to improve the ductility and strength of Mg alloys simultaneously. Recently, researches have shown that severe plastic deformation (SPD) provides a strong grain refinement effect, and the ultrafine-grained (UFG) materials produced by SPD exhibit exceptional mechanical properties, such as equal channel angular extrusion/pressing (ECAE/P)[9], high pressure torsion (HPT)[10] and cyclic extrusion and compression (CEC)[11]. Among the various SPD techniques, repetitive upsetting-extrusion (RUE) is an effective method to realize large accumulated deformation strains and can be suitable for fabricating relatively large bulk samples with maintaining the original shape. Hu et al. [12] have

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Corresponding author: Zhang Zhimin, Ph. D., Professor, School of Material Science and Engineering, North University of China, Taiyuan 030051, P. R. China, Tel: 0086-351-3921778, E-mail: nucforge@126.com

investigated grain refinement mechanisms for LY12 aluminum alloy during RUE process. They suggested that homogeneous fine structure can be obtained when the billet material was subjected to an accumulated strain large enough and the cellular structures and subgrains resulting from upsetting-extrusion alternating deformation processing will be equiaxed. Moreover, RUE process has been used for refinement of microstructure of Mg alloys. Xu et al. [13] investigated the microstructure and mechanical properties of AZ61 alloy processed by RUE. They found that the grain structure was refined to 3.5 μm after 3 RUE passes at 285 ºC with significant improvement on mechanical properties. Chen et al.[14] studied microstructural evolution of AZ80 magnesium alloy treated by RUE, and the results indicated that fine and humongous microstructure were obtained when the RUE passes reached to 8.

In the present work, RUE process with decreasing temperature was introduced to as-homogenized Mg-13Gd-4Y-2Zn-0.5Zr (wt%) alloy. The microstructures and texture evolution after different RUE passes were studied systematically.

1 Experiment

The Mg-13Gd-4Y-2Zn-0.5Zr (wt%) alloy was used in this research. The alloy ingot was provided by Yinguang Magnesium Industry Co. Ltd. Cylinder billets for RUE with 50 mm in diameter and 210 mm in height were machined from the center of the ingot, and then homogenized at 520 ºC for 16 h followed by cooling in air.

The schematic of RUE process adopted in this study is illustrated in Fig.1. Each pass of RUE consisted of two basic steps: (1) upsetting and (2) extrusion, i.e., the billet was subjected to upsetting and extrusion repeatedly. The RUE temperature decreased from 480 ºC to 370 ºC by a drop of 10 ℃ every step. The accumulated equivalent strain is approximately given by:

$$\varepsilon = 4n\ln\left(\frac{D}{d}\right) \tag{1}$$

where D is the diameter of upsetting cavity, d is the diameter of extrusion cavity, and n is the number of process passes. In this study, the upsetting and extrusion rams were designed to be 70 and 50 mm in diameter, respectively, corresponding to extrusion ratio of 1.96. Therefore, a cumulative strain of 1 pass of RUE was calculated to be 1.35.

The RUE process was carried out on a 6300 kN hydraulic press at pressing speed of 5 mm/s. Before each process pass, the billets and dies were heated at deformation temperature and isothermally held for 1 h, and then lubricated with an oil-based graphite lubricant.

Specimens for microstructure analysis were cut from the central region of RUEed samples and prepared by the conventional mechanical polishing and etched with a solution of 100 mL ethanol, 6 g picric acid, 5 mL acetic and 10 mL water. Microstructure of the alloys was observed by Zeiss Axio Imager A1m optical microscope (OM), Hitachi SU5000 scanning electron microscopy (SEM) equipped with EDAX energy dispersive X-ray spectrometer (EDS), JEOL JEM-2100F transmission electron microscope (TEM) operating at 200 kV. The EBSD measurement was conducted by EDAX-TSL EBSD system, equipped with OIM analysis software.

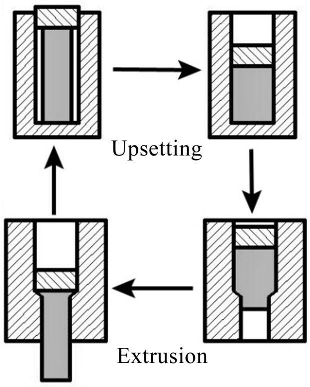

Fig.1 Schematic of the repetitive upsetting-extrusion at decreasing temperature

2 Results and Discussion

2.1 Initial microstructure

Fig.2 shows the optical microstructures of the as-cast and homogenized alloy samples. The as-cast alloy consists of α-Mg matrix with the average grain size of 61 μm and eutectic compounds mainly distributed at the grain boundaries. Furthermore, a small amount of fine lamellar-shaped phases can be observed inside grains, which should be solute segregated stacking faults (SFs) or the short lamellar-shaped long period stacking ordered (LPSO) structure[15]. After homogenization treatment (Fig.2b), the eutectic compounds are dissolved into α-Mg matrix and block-shaped LPSO phases are formed at grain boundaries. Meanwhile, the majority of α-Mg grains are full of the lamellar-shaped phases. The average grain size of as-homogenized alloy increases slightly to 64 μm, due to the pinning effect of these phases on the motion of grain boundaries.

Fig.3 shows the SEM image of the as-homogenized alloy. It can be seen that the block-shaped LPSO phases are discontinuously distributed at the grain boundaries and some of them grow in the grains with a specific orientation relationship with α-Mg matrix. By EDS analysis, the composition of the block-shaped LPSO phase is determined

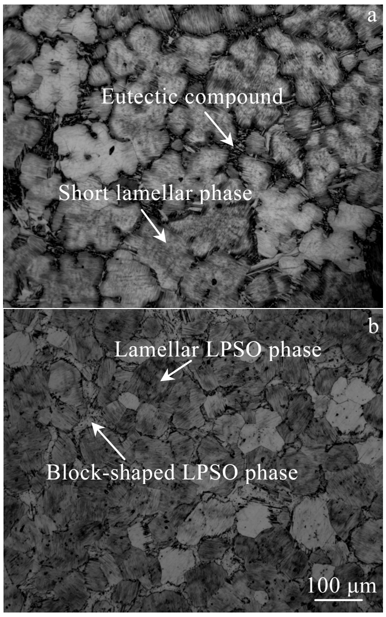

Fig.2 Optical micrographs of as-cast (a) and as-homogenized alloy (b)

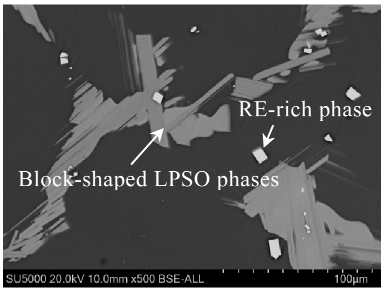

Fig.3 SEM image of the secondary phases in as-homogenized alloy

to be 88.12Mg-3.77Gd-2.86Y-4.83Zn-0.42Zr (at%), which is close to $Mg_{12}RE_1Zn_1$, same as the composition reported for 14H LPSO phase in the Mg-10Gd-3Y-1.2Zn-0.4Zr alloy[16]. Besides LPSO phases, fine cuboidal particles can be observed and the composition is determined to be RE-rich phases with 31.74Mg-21.16Gd-40.9Y-0.97Zn-5.52Zr (at%) by EDS, which was reported commonly in Mg-RE alloys[17].

2.2 Microstructural and texture evolution

Fig.4 shows the microstructure of alloys with various RUE passes taken from the longitudinal section. After 1 RUE pass, many fine grains are formed along the grain boundaries of coarse grains resulting in a ‘necklace’ structure due to DRX, as shown in Fig.4a. It can be also observed that lamellar LPSO phases in coarse grains bend or kink to some degree. The kink bands can induce the nucleation of DRXed grains, as indicated by the circles in Fig.4a. As the RUE passes increases to 3 (Fig.4b), unDRXed regions with coarse grains still remain although the number of recrystallized grains is obviously increased. A large amount of lamellar phases are also observed throughout the whole coarse deformed grains. Moreover, it is found that the coarse grain boundaries are serrated, which indicates that the DRX mainly take place at grain boundaries and may be inhibited by lamellar LPSO phases. After 5 passes, the microstructures are clearly refined and become homogeneous, and the grain boundaries in the DRXed region are illegible. With further deformation, the microstructure is occupied by fine DRXed grains after RUE 6 passes, as shown in Fig.4d. In addition, particles can be recognized in the DRXed region during the RUE process, which are β phase ($Mg_5(Gd,Y)$, $F\bar{4}3m$ a = 2.23 nm) [18] according to TEM observation (Fig.5). The β phases should inhibit the grain boundary mobility and restrict the grain growth. After 6 pass RUE (Fig.4d), volume fraction of the β

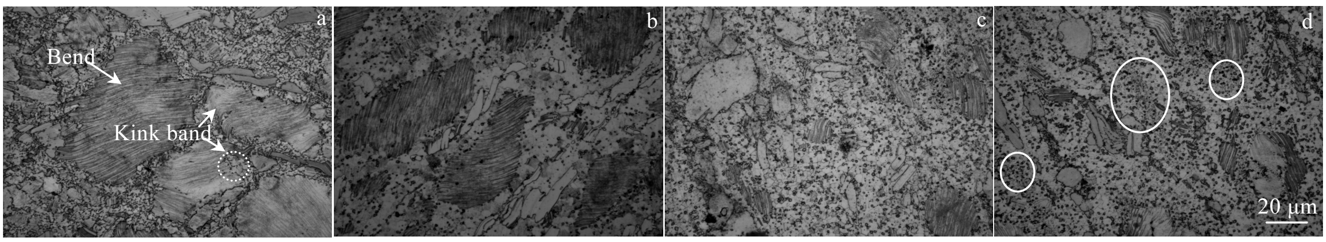

Fig.4 Optical micrographs of the RUEed alloys with different passes: (a) 1 pass, (b) 3 passes, (c) 5 passes, and (d) 6 passes

phases is significantly increased, which may improve the pinning effect on the DRXed gain growth. Therefore, the ultrafine grain (≤2 μm) areas are found among the DRXed grains formed in previous passes, as indicated by the circles in Fig.4d.

Fig.6 shows the inverse pole figure maps (IPF) of the alloy after different RUE processes. The black regions are corresponded to the secondary phases without sufficient confidence index (CI) values for obtaining clear Kikuchi diffraction patterns in EDAX EBSD system[19]. It can be seen that fine grains with relatively random crystal orientation are located around the coarse grains after 1 RUE pass. It should be the characteristic of a DDRX. Moreover, the obvious color variation caused by dense dislocations and

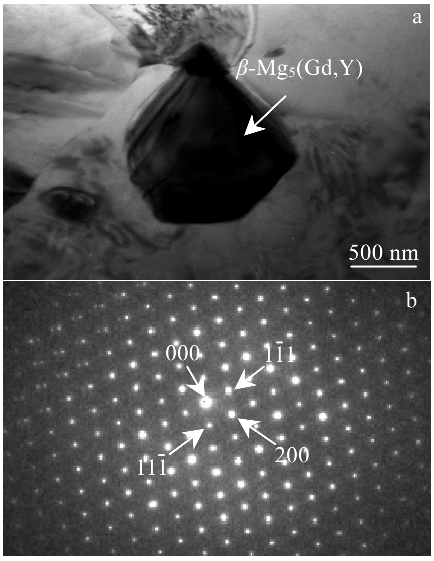

Fig.5 TEM BF image (a) and SAED pattern (b) of the precipitated particles with the incident electron beam//$[011]_{\beta}$

subgrain boundaries can be observed in some residual coarse grains, marked by the white frame in Fig.6a. Meanwhile, as indicated by the white arrows in Fig.6a, the kink bands (KBs) form in the lamellar LPSO phases, and these KBs can transfer to the adjacent α-Mg matrix leading to the formation of kink grain boundaries (GBs). Furthermore, the large misorientation of the KBs and local plastic strain around them may create favorable conditions for DRX to operate[20]. As a result, a small amount of fine grains form at the kink GBs of α-Mg matrix as indicated by the white circle in Fig. 6a. With increasing RUE passes, areas of the DRXed grains obviously increase. The much more randomly distributed grains colors indicate that the grain orientations are changed. Meanwhile, the DRXed grains are much finer than those after one RUE pass, which indicates that the former DRXed grains could further become sites for the formation of new DRXed grains. It should note that numerous ultrafine grains form at DRXed region in the alloy after 6 passes, as shown in Fig. 6c.

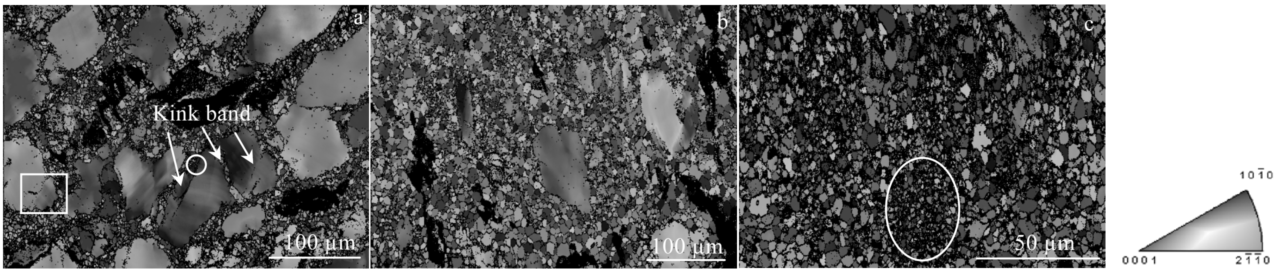

Fig.6 Inverse pole figure maps of the alloy after RUE with different passes: (a) 1 pass, (b) 3 passes, and (c) 6 passes

In order to describe the evolution of grain refinement, it is essential to quantify the grain size based on either the size of individual grain, d, or the average grain size, d_{ave}, because of the nonuniform grain distribution resulting from incomplete DRX at lower degree of deformation. The 'fine' grains are defined as grains having an diameter $d \leq 10$ μm and the volume fraction of fine grains V_f is defined as follows[21]:

$$V_f = \left(\frac{\sum A_f}{\sum A_i} \right) \tag{2}$$

where $\sum A_f$ and $\sum A_i$ are the total area of individual fine grains and total sample area, respectively. It can be seen that the distribution range of grain size decreases continuously.

Fig.7 shows typical distribution histograms of grain size for alloys processed by RUE. After 1 RUE pass, microstructure is characterized by a bimodal distribution of the grain size that the volume fraction of fine grains is merely 23.6% while the remaining 76.4% is larger than 10 μm, as shown in Fig.7a. This is typical microstructure of partially DRXed structure. With increasing the RUE passes, both the coarse and the fine grains are effectively refined and the volume fraction of fine grains increases, the grains become more homogenous and smaller, as show in Fig.7b~7c. After 6 RUE passes (Fig.7d), the percentage of fine grains increases to 99% with a peak distribution ranging from 1 μm to 4 μm, which could be concluded that 'complete' recrystallization is attained. And the average grain size reaches 3.4 μm.

Fig.8 presents misorientation angle distribution of RUEed alloys with different passes. After 1 pass, the misorientation distributions are mainly characterized by a sharp peak at low angles boundaries (LABs) less than 5° and a number of high angle boundaries (HABs) with misorientation above 15°. With increasing RUE pass, the fraction of LABs decreases sharply and that of HABs conversely increases. It demonstrates that new grains originate the subgrains with the continuous transformation of low angle boundaries (LABs) into high angle boundaries (HABs) during RUE process. This process is classified as CDRX.

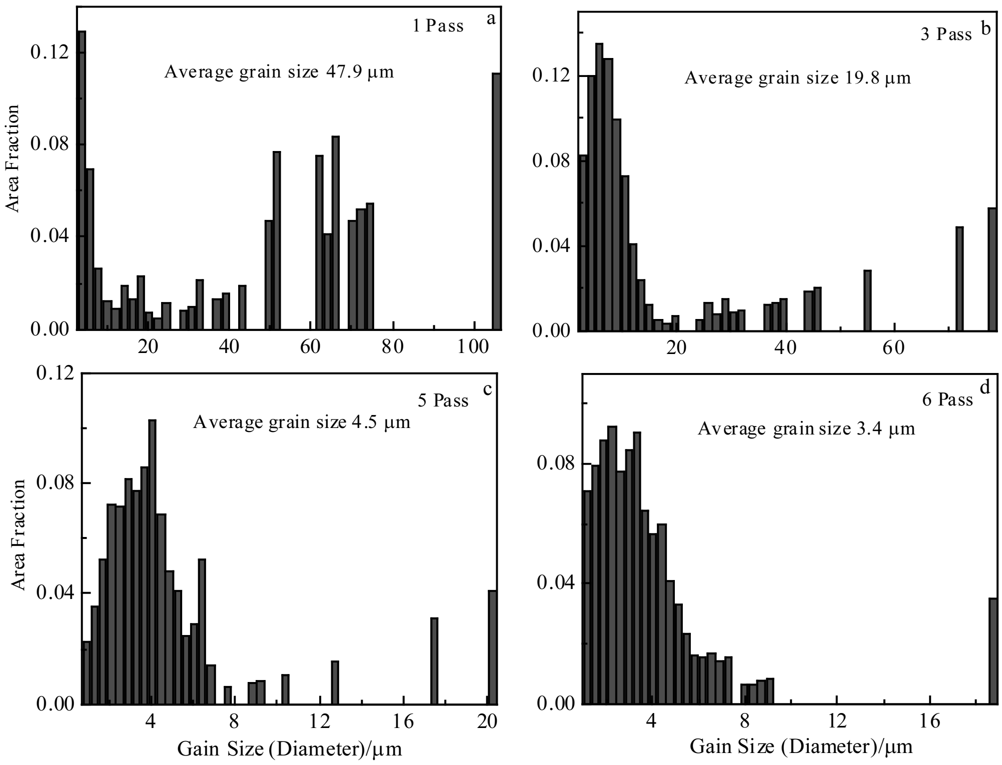

Fig.7 Grain size distribution of the alloy after RUE with different passes: (a) 1 pass, (b) 3 passes, (c) 5 passes, and (d) 6 passes

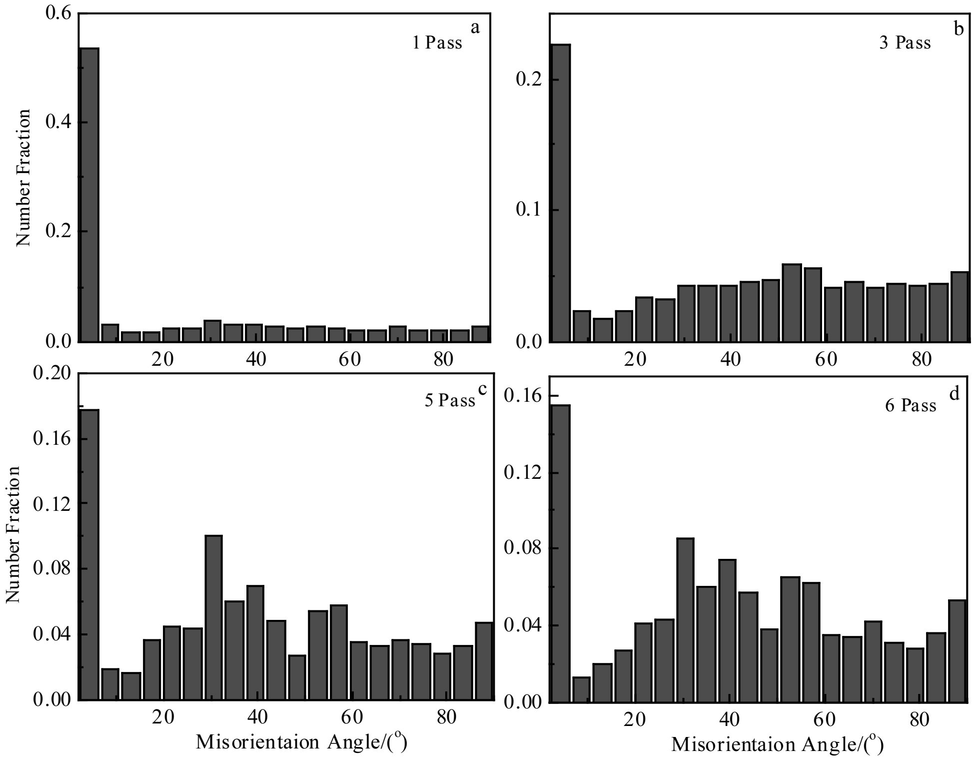

Fig.8 Misorientation angle distribution of the alloy after RUE with different passes: (a) 1 pass, (b) 3 passes, (c) 5 passes, and (d) 6 passes

According to discussion above, it can be concluded that during the RUE process, with increase in the number of RUE passes, the average grain size of the RUE formed samples decreases continuously. Moreover, the efficiency of grain refinement is gradually weakened, because strain-induced grain refinement ceases as a dynamic balance between recrystallization refinement and grain growth when DRX process is complete. The microstructure featured by a bimodal grain structure is composed of fine grains of several microns and unRDXed regions come into being during the RUE process due to the presence of the lamellar LPSO phases and fine Mg_5(Gd,Y) precipitates. The fine grains area expands gradually with increasing accumulative strains and decreasing temperature. Therefore, it can be proposed that DDRX and CDRX mechanisms may simultaneously work in grain refinement during RUE in present study. Local stress concentration can be formed easily at the interfaces between the LPSO phase, grain boundary and Mg matrix during the hot deformation, which cause the motion, piling-up, rearrangement and emergence of dislocations, and in turn to form subgrains and subgrain boundaries, as further deformation, DRX grains are formed by the in situ evolution of the subgrains with the growth of low angle boundaries to high angle grain boundaries. In addition, kink of lamellar LPSO phase plays an important role in refining grains through subdividing the grain by inducing recrystallization evolved at the kink bands.

Fig.9 shows the textures of the samples with different RUE passes. For easy analysis, {0001} and $\{10\bar{1}0\}$ pole figures after different RUE process were illustrated. It can be seen that there is a strong basal texture in the alloy after RUE 1 pass according to pole intensity. The texture is different from typical basal textures in both the as-compressed and as-extruded Mg alloys, i.e., with {0001} plane perpendicular to the compression direction (CD) and parallel to the extrusion direction ED, respectively[22, 23]. Apparently, basal planes are mainly not only perpendicular to extrusion direction but also 45° with the extrusion direction, which may be resulted from the rotation of the original basal plane by 45° from the extrusion direction since the loading direction is turned from radial to axial direction alternatively during RUE processing. The new texture is similar to that of CECed alloys[20], but the texture intensity of the RUEed alloy is much higher. With increasing the RUE passes, the basal planes have the tendency to periodically change. Therefore, the initial intense texture becomes so disintegrated that the basal poles are distributed more randomly and the maximum texture intensity declines gradually from 13.8 to 8.7 as the RUE passes increase from 3 to 6, as shown in Fig.10b~10d. The $\{10\bar{1}0\}$ pole intensity is also distributed randomly following the same tendency. In addition, Xu et al[24]. reported that the DRXed grains play an important role in weakening the overall texture by counteracting the strong deformation texture of the unDRXed grains in hot deformation Mg alloys. Xia et al. [25] reported that the precipitation particles of Mg-Gd-Y alloy have a strong ability to inhibit the grain rotation, which is helpful for the texture weakening. Thus, it can be concluded the RUE process with decreasing temperature provide a more effective method featured by randomization for basal texture weakening of the alloy.

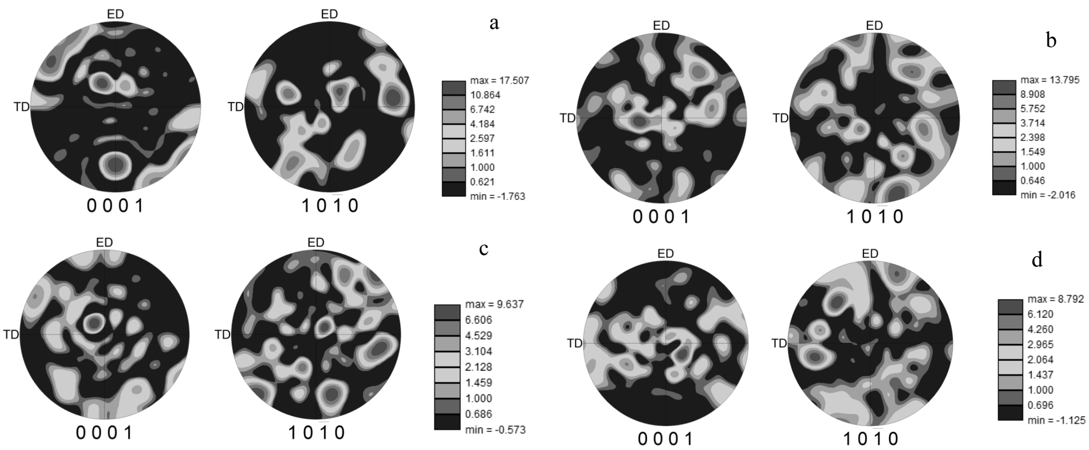

Fig.9 {0001} and $\{10\bar{1}0\}$ pole figures of the alloy after multidirectional forging with different passes: (a) 1 pass, (b) 3 passes, (c) 5 passes, and (d) 6 passes

3 Conclusions

1) The microstructure of Mg-13Gd-4Y-2Zn-0.5Zr (wt%) alloy can be effectively refined by RUE at decreasing temperature. A homogeneous ultrafine-grain structure with an average grain size of 3.4 μm is obtained after 6 passes RUE processing, i.e., accumulative strain of 8.4.

2) The basal texture is gradually weakened with

increasing the RUE passes, which results from cooperative effect of dynamic recrystallization and alternative change in loading direction during RUE process.

3) Grain refinement during RUE is dominated by a complicated combination of discontinuous dynamic recrystallization and continuous dynamic recrystallization. Kink of lamellar LPSO phase plays an important role in refining grain in low strain regions through subdividing the grain by inducing recrystallization evolved at the kink bands.

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降温往复镦粗-挤压变形对 Mg-Gd-Y-Zn-Zr 合金晶粒细化及织构变化的研究

杜 玥，张治民，张冠世，闫钊鸣，于建民

(中北大学，山西 太原 030051)

摘 要：在 480 °C 降温至 370 °C 条件下，采用循环镦−挤工艺对均匀化后的 Mg-Gd-Y-Zn-Zr 合金进行变形，对循环镦−挤变形过程中的合金微观组织和织构变化进行研究。结果表明，随着循环镦−挤变形道次的增加，晶粒尺寸逐渐减小。在变形 6 道次后，累积应变达到 8.4，得到了平均晶粒尺寸为 3.4 μm 的细小均匀的微观组织。晶粒细化是由非连续动态再结晶和连续动态再结晶复杂共同作用引起的。另外，变形过程中，原始粗大晶粒内的片层状长程有序相（LPSO）发生扭折变形产生扭折带，并在扭折带上发生动态再结晶，分割原始粗晶，起到晶粒细化作用。结果还表明，1 道次镦−挤变形后，合金产生强的基面织构，随着变形道次的增加，织构强度有所减弱。织构弱化的原因是动态再结晶和加载力在轴向和径向交替变化共同作用。

关键词：Mg-Gd-Y-Zn-Zr合金；往复镦−挤；晶粒细化；织构

作者简介：杜 玥，男，1988 年生，博士，中北大学材料科学与工程学院，山西 太原 030051，电话： 0351-3921778，E-mail: nuc_duyue@126.com