Dynamic and Static Aging Precipitation of β -Mg₁₇Al₁₂ in the AZ80 Magnesium Alloy During Multi-directional Forging and Subsequent Aging

Qing-Feng Zhu^{1,2} · Gao-Song Wang^{1,2} · En-Ge Zhang^{1,2} · Fan-Zheng Liu^{1,2} · Zhi-Qiang Zhang^{1,2} · Jian-Zhong Cui^{1,2}

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Abstract Dynamic and static aging precipitation of Mg₁₇Al₁₂ phases in AZ80 magnesium alloy was studied by multi-directional forging (MDF) with decreasing temperatures from 410 to 300 °C and subsequent aging process. The results show that the morphology of the β -Mg₁₇Al₁₂ phases during forging process dynamically precipitates and aging process (statically precipitation) exhibited granular and laminar shapes, respectively. During the MDF, the inhomogeneous dynamic precipitation of the β -Mg₁₇Al₁₂ phases results in the uniformity on grain size, which is fine in the area with many granular Mg₁₇Al₁₂ phases but the grain is still coarse where there is no Mg₁₇Al₁₂ phases. During the aging process, the morphology of newly formed β -Mg₁₇Al₁₂ phases depends on the structural character of the forged sample. The newly precipitated β -Mg₁₇Al₁₂ phases are coarse laminar and needle-like shape in area with coarse grain. While, the fine newly precipitated β -Mg₁₇Al₁₂ phases are fine granular and needle-like in the area with fine grain.

KEY WORDS: Dynamic strain aging; Precipitation; AZ80 magnesium alloy; Multi-directional forging; Dynamic recrystallization

1 Introduction

Dynamic strain aging (DSA) is a phenomenon in metals and alloys resulted from the interaction between the diffusing solute atoms and the moving dislocation, and it has been reported in many alloys during uniaxial tension or compression process [1–13]. However, it is difficult to control the DSA process in traditional metal forming process (such as extrusion and rolling), because both deformation time and deformation reduction of the traditional

metal forming process are limited. By contrast, they can be effectively controlled during the severe plastic deformation (SPD) by constantly increasing accumulative strains. Recently, the DSA behavior during SPD process has been widely studied [14–20]. Roven et al. [15] found that the nano-scaled globular β'' phase (~ 4 nm) precipitated in 6063 aluminum alloy during equal-channel angular pressing (ECAP) at room temperature and 175 °C. Furthermore, DSA process also has been studied in magnesium alloys during the multi-directional forging (MDF) process [17–19] and it is reported that the granular Mg₁₇Al₁₂ phases can precipitate during MDF and distribute along the grains boundaries, restricting the growth of dynamic recrystallized grains and therefore refining the grains. Our previous paper has also found the dynamic precipitation behavior during MDF process in AZ80 magnesium alloy [20]. However, the effect of dynamic precipitated Mg₁₇Al₁₂ phases on the following static aging precipitation was rarely discussed.

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✉ Qing-Feng Zhu
zhuqingfeng@epm.neu.edu.cn

¹ Key Laboratory of Electromagnetic Processing of Materials, Ministry of Education, Northeastern University, Shenyang 110819, China

² School of Materials Science and Engineering, Northeastern University, Shenyang 110819, China

In the present work, the dynamic and static strain aging precipitation of $\text{Mg}_{17}\text{Al}_{12}$ phases in AZ80 magnesium alloy was studied by MDF in the temperature range from 410 to 300 °C and subsequent aging process.

2 Experimental

Three rectangular samples [61 mm (x axis) $\times$ 52 mm (y axis) $\times$ 45 mm (z axis)] were cut from a commercial direct-chill-casted AZ80 magnesium alloy (annealed at 410 °C for 16 h) with compositions of Mg –7.85 Al –0.43 Zn –0.21 Mn (wt%). The samples were first reheated to 410 °C and hold for 1.5 h and then forged by a 300 t hydraulic press. The MDF process was carried out with alternating loading axes (i.e., $x \rightarrow y \rightarrow z \rightarrow x \dots$). The strain rate and the true strain of each pass are 0.13 s^{-1} and 0.5, respectively. The experimental parameters are shown in Table 1, and more detailed information for MDF process was described in Ref. [20]. After the forging process, the samples were quenched into water at room temperature. Afterward, the specimens were cut from these samples and aged at 170 °C for different time (5, 10, 20, 30, 40, 50 h).

The samples for MDF were cut along middle plane parallel to the last compression axis for observation of the

macro- and microstructure. Traditional methods were used to prepare the sample for metallography, including grinding, polishing and etching with the mixture of liquid picric acid and nitric acid. The Al compositions in the Mg matrix were manually detected by the EPMA-1600/1610 electron probe microanalysis (EPMA). Small specimens with different forging and aging condition were analyzed by optical microscope (OM) and scanning electron microscope (SEM).

In order to determine the precipitation temperature of the $\text{Mg}_{17}\text{Al}_{12}$ phase, the differential scanning calorimetry (DSC) was applied to analyze the thermal behaviors of the AZ80 alloy. The heating rate was 5 °C/min. Scans were performed between room temperature and 400 °C with purified nitrogen flow at a rate of 50 ml min^{-1} .

3 Results and Discussion

Figure 1a shows the microstructure of homogenized AZ80 alloy. It can be seen that the matrix is composed of equiaxed grains with the size of about 210 μm (Fig. 1a). Some white phases are present along the grains boundaries, which have the laminar morphology (Fig. 1b). They are identified as from SEM-EDX analysis. As shown in Fig. 1c, these laminar $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases formed at the grain boundaries and were perpendicular to the grain boundaries.

Figure 2a, b shows the macrostructures of samples P1 and P2, respectively. The shape of two samples is basically the same. The flow lines are present on the cross section of the both two samples, but they become more obvious in P2 (Fig. 2b) with increasing the forging passes and decreasing

Table 1 Experimental parameters

Sample number	Forging passes	Final forging temperature (°C)	Accumulative true strain ε
P1	4	335	2
P2	14	300	7

Fig. 1 Optical micrograph of the homogenized AZ80 alloy specimen: **b** magnified micrographs from the rectangular frame in **a**, **c** magnified micrographs from the rectangular frame in **b**

Fig. 2 Macro- and microstructure of the MDF samples: **a** P1-sample, **b** P2-sample. **c, d** Magnified micrographs from the *rectangular frame* in as signed by *rectangular frames* in the **a, b**, respectively

temperature. To reveal the details, the microstructures corresponding to the zones enclosed by the rectangular frames in Fig. 2a–b are displayed in Fig. 2c–d, respectively. It can be found that the metal flow lines are actually composed of some mutual-mixed black and white structures. Furthermore, the area of the black structures increases with increasing accumulative strains and decreasing temperature. The enlarged image in Fig. 2d shows that the black structures are very fine grains with granular $\beta\text{-Mg}_{17}\text{Al}_{12}$ distributed along the grain boundaries while the white structures are relatively coarse grains without $\beta\text{-Mg}_{17}\text{Al}_{12}$ formed [20]. The calculated grain size in white structure in Fig. 2c, d is similar, indicating the minor influence of forging passes and temperature.

Figure 3 shows the enlarged SEM images of the transitional region between black and white structures in Fig. 2d. In contrast to the laminar morphology observed in the homogenized alloy (Fig. 1), the $\beta\text{-Mg}_{17}\text{Al}_{12}$ shows granular shape and they mainly distributed along the grain boundaries in the fine grain area but no $\beta\text{-Mg}_{17}\text{Al}_{12}$ present in the coarse grain area. The appearance of fine-grained area is always accompanied by the presence of granular $\beta\text{-Mg}_{17}\text{Al}_{12}$. In addition, it is also found that the Al content in the Mg matrix is also different from the fine- and coarse-grained area. EPMA analysis at the points indicated in

Fig. 3 Micro-area of P14-sample measured by EPMA

Fig. 3 reveals that the average Al content in the Mg matrix of the fine- and coarse-grained areas is 8.1 and 6.1 wt%, respectively, indicating that the inhomogeneous distribution of Al in the Mg matrix has directly led to unevenly distribution of the $\beta\text{-Mg}_{17}\text{Al}_{12}$ -phase precipitations. The $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases in Figs. 2 and 3 are dynamically precipitated during the progressive deformation. In addition, the pinning effect of the $\beta\text{-Mg}_{17}\text{Al}_{12}$

Fig. 4 Microstructures of the forged sample with different aging treatments: **a** P1-sample, **b** P1-sample after aged at 170 °C for 10 h, **c** P1-sample after aged at 170 °C for 30 h, **d** P2-sample, **e** P2-sample after aged at 170 °C for 10 h, **f** P2-sample after aged at 170 °C for 30 h

phases may play an important role in grain refinement of the AZ80 aluminum alloy.

Figure 4 shows the microstructures of the forged and subsequently aged AZ80 alloy. Both the black and white structure can be observed in all samples. However, the area fraction of the black structure is greatly depended on the aging condition, which generally increases with prolonging aging time. A further analysis showed that the new precipitated β - $\text{Mg}_{17}\text{Al}_{12}$ phases during aging at 170 °C result in the enlargement of the black structure area. So, the total area of black structure is the area with $\text{Mg}_{17}\text{Al}_{12}$ precipitation, and the increment area of the black structure with prolonging aging time is the area of newly precipitated $\text{Mg}_{17}\text{Al}_{12}$ phases.

Image analysis was employed to measure the area fraction of the black structure of the samples in Fig. 4, and the results are shown in Table 2. It is shown that, for the same aging condition, the area fraction of the black structure in the sample P2 is much higher than that of the sample P1, indicating that the higher deformation stored energy in P2 can promote the formation of $\text{Mg}_{17}\text{Al}_{12}$ phase.

In addition, the precipitation process of the $\text{Mg}_{17}\text{Al}_{12}$ phase in the different samples is related to the condition of final forging temperature and forging passes. With the aging time prolonging from 0 to 10 h, the fraction of the black structure in P1 and P2 increases 30.9 and 10.7%, respectively. With the aging time prolong from 10 to 30 h, the fraction of the black structure in P1 and P2 increases 12.2 and 20.8%, respectively.

Figure 5 shows the detail morphologies of the new precipitated phases in the coarse- and fine-grained areas in sample P1 aged at 170 °C for 10 h. As shown in Fig. 5a, laminar β - $\text{Mg}_{17}\text{Al}_{12}$ phases precipitated in both coarse and fine grain areas. The pre-precipitated (with granular shape) and newly precipitated (with laminar shape) β - $\text{Mg}_{17}\text{Al}_{12}$ phases almost covered the whole fine grain area. And the new precipitated β - $\text{Mg}_{17}\text{Al}_{12}$ phases are surrounded by the pre-precipitated β - $\text{Mg}_{17}\text{Al}_{12}$ phases, as shown in Fig. 5c. This clearly indicates that the pre-precipitated β - $\text{Mg}_{17}\text{Al}_{12}$ phases can inhibit the growth of new precipitated β - $\text{Mg}_{17}\text{Al}_{12}$ phases during the aging process. In the coarse grain area, the laminar β -phases also nucleated at the grain boundaries and growth perpendicular to the grain boundaries during the aging process, as shown in Fig. 5b. With further aging, the newly precipitated laminar β -phases at the grain boundaries grew and contacted with the adjacent new formed phases, and then, the area of the β -phases constantly expanded. Figure 6 displays the precipitates in sample P1 after 30 h at 170 °C. Compared with aging 10 h at 170 °C (Fig. 5a), the area fraction of β -phases is much bigger after 30 h (Fig. 6a). In addition, some very fine

Table 2 Fraction of the black structure in forged sample after aged at 170 °C for different time

Aging time (h)	P1	P2
0	21.7	55.5
10	52.6	66.2
30	64.8	87.0

Fig. 5 Microstructures of P1 with aged at 170 °C for 10 h: **b, c** magnified micrographs from the *rectangular frame* in as signed by *rectangular frames* in the **a**

Fig. 6 Microstructures of P1 with aged at 170 °C for 30 h: **b** magnified micrographs from the *rectangular frame* in as signed by *rectangular frames* in the **a**

white phases appeared among the granular β -phases in the fine grains area, as shown in Fig. 6b. It is clear that the inhomogeneous distribution of Al element in the matrix affected the precipitation of the β - $\text{Mg}_{17}\text{Al}_{12}$ phases. During the aging treatment, the β -phases easily precipitated in the area with high Al content, but the β - $\text{Mg}_{17}\text{Al}_{12}$ phases only

precipitated along the grain boundaries in coarse grain area (with lower Al content).

The typical enlarged micrographs of sample P2 aged at 170 °C for 10 h are shown in Fig. 7. It can be seen that newly precipitated small-sized laminar β - $\text{Mg}_{17}\text{Al}_{12}$ phases mainly distributed on the periphery of the fine-grained area,

Fig. 7 Microstructures of P1 with aged at 170 °C for 10 h: **b** magnified micrographs from the *rectangular frame* in as signed by *rectangular frames* in the **a**

as shown in Fig. 7a. In the coarse-grained area, the β - $\text{Mg}_{17}\text{Al}_{12}$ phases only formed at the grain boundaries. In the fine-grained area, some small-sized β - $\text{Mg}_{17}\text{Al}_{12}$ phases only precipitated among the granular β - $\text{Mg}_{17}\text{Al}_{12}$ phases, as shown in Fig. 7b. After aged at 170 °C for 10 h, the volume fraction of newly formed β - $\text{Mg}_{17}\text{Al}_{12}$ phases in P2 is less than that in P1. The reason for this phenomenon

should attribute to more Al element has precipitated to granular β - $\text{Mg}_{17}\text{Al}_{12}$ phases in P2 during forged at lower temperature. With further aging to 30 h (Fig. 8), the newly formed lamellar β -phases expanded to the coarse grain area, as shown in Fig. 8a. Many of the newly formed β -phases grew to the lamellar shape. At the same time, many fine needle-like β -phases formed in some coarse grains, as

Fig. 8 Microstructures of P2 with aged at 170 °C for 30 h: **b, c** magnified micrographs from the *rectangular frame* in as signed by *rectangular frames* in the **a**

shown in Fig. 8b. This phenomenon suggests that the Al content in some grains is not enough to support the formation of the coarse laminar shape $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases. In addition, many newly formed fine granular and needle-like white phases appear among the granular β -phases in the fine-grained area, as shown in Fig. 8c.

Figure 9 presents the DSC curve of the as-cast AZ80, which shows an endothermic peak between 300 and 350 °C. This reaction can be related to the dissolution of $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases, meaning that the $\beta\text{-Mg}_{17}\text{Al}_{12}$ phase will precipitate from the Mg matrix as the temperature lower than 330 °C for this alloy. Therefore, the dynamic precipitation of granular $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases occurs when the forging temperature constantly decreases from 410 to 335 °C, as shown in Fig. 2c. When the final forging temperature further decreased to 300 °C, more granular $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases dynamically precipitated from the Mg matrix, as shown in Fig. 2d. These Al atoms remained in solid solution after forging would precipitate during the following aging process. Under the same aging condition, the precipitation of the $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases was determined by the Al content and local stored energy of Mg matrix. In the coarse grain area, the high Al content (around the pre-precipitation and fine grain areas) could provide more driving force for precipitation of $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases. Meanwhile, the grain boundaries in the coarse grain area could provide ideal places for the nucleation of the $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases. Hence, the newly formed $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases firstly nucleated on the grain boundaries and grew to coarse laminar shape with increasing aging time. And the newly precipitated $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases expanded from the periphery to the center of the coarse-grained area.

During the aging process, the conditions in the fine-grained area were different from the coarse-grained area.

Fig. 9 Typical DSC scan at a heating rate of 5 °C/min of the as-cast AZ80

The pre-precipitated granular $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases formed on the grain boundaries have consumed the local Al element, leading to that the newly formed $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases could only nucleate in the area far from the grain boundaries. Moreover, the growth of these newly formed $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases was limited by pre-precipitation granular $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases, and then, these newly formed $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases cannot grow up to developed laminar shape.

4 Conclusions

- (1) In the present multi-directional forging process, the coexistence of the fine-grained (with many granular $\text{Mg}_{17}\text{Al}_{12}$ phases) and coarse-grained (without $\text{Mg}_{17}\text{Al}_{12}$ phases) area is the main character of the structure. The fraction of fine-grained area and the fraction of granular $\text{Mg}_{17}\text{Al}_{12}$ phases increase with the increasing forging pass and decreasing temperature.
- (2) In the subsequent aging process, the morphology of the newly formed $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases is greatly related to the grain size that coarse laminar $\beta\text{-Mg}_{17}\text{Al}_{12}$ precipitated in the coarse-grained area while they are fine needle-like in fine-grained area.
- (3) The morphology of the dynamically precipitated $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases (formed during forging process) and statically precipitated $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases (formed during aging process) is quite different, which is granular and laminar in dynamic precipitation and laminar in static precipitation. The dynamic precipitation $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases formed during forging process can affect the static precipitation process of the newly formed $\beta\text{-Mg}_{17}\text{Al}_{12}$ phases.

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