Microstructural Evolution during Partial Melting and Semisolid Forming Behaviors of Two Hot-Extruded Magnesium-Rare Earth Alloys

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Abstract. Reheating experiments and semisolid compression tests were conducted on two hot-extruded magnesium-rare earth (Mg-RE) alloys, Mg-8.20Gd-4.48Y-0.36Zr-3.34Zn and Mg-3.75Gd-5.15Y-0.75Zr-3.05Zn by using a multistage hot compression test machine. Dissolution of eutectic compounds, growth of grains, and partial melting took place during the reheating of these Mg-RE alloys and resulted in spherical semisolid slurries at certain temperatures (580 °C for Mg-8.20Gd-4.48Y-3.34Zn-0.36Zr, 560 °C for Mg-3.75Gd-5.15Y-0.75Zr-3.05Zn). Owing to the different alloying element contents of these two Mg-RE alloys, eutectic compounds with different morphologies were found inside them after reheating and rapid cooling processes. The forming characteristics of these two Mg-RE alloys in semisolid state were discussed based on the results of compression tests.

1 Introduction

Addition of Zr and rare earth (RE) elements was one of the most feasible precipitation strengthening methods for Mg alloys [1]. Compared with conventional Mg alloys, Mg-RE alloys exhibited more excellent high-temperature mechanical properties [2]. Thus, Mg-RE could be used to manufacture structural parts working in extreme environment [3]. However, casting was not an ideal forming process for Mg-RE alloys, because of the weaker mechanical properties of cast parts [4]. Moreover, the formalities of Mg-RE alloys for traditional forging were not good enough to meet the requirement of near-shape forming [5].

Owing to the characteristic microstructure and forming behaviors of different phases in semisolid slurries, semisolid forming technology was considered as one of the idea forming technology to realize the near-shape forming of Mg-RE alloys [6]. The feasibility of semisolid forming in manufacturing of conventional Mg alloy parts was verified by in the past decade by researchers [7-9]. However, the research on semisolid forming of Mg-RE alloys is quite limited. To establish a feasible strategy for semisolid forming of Mg-RE alloys, essential acknowledgements should be obtained and accumulated by basic experiments.

In this study, two hot-extruded Mg-RE alloys with different chemical compositions were selected as starting materials. A series of reheating-cooling experiments, semisolid compression tests were conducted in this study. On the basis of the experimental results, not only the microstructural evolution of those Mg-RE alloys during reheating, subsequent cooling, and semisolid forming, but

also the semisolid forming behaviors of those alloys were investigated and discussed.

2 Experiments

In this study, Mg-8.20Gd-4.48Y-0.36Zr-3.34Zn and Mg-3.75Gd-5.15Y-0.75Zr-3.05Zn were selected and named as alloy A and B, respectively. Their chemical compositions are listed in Table 1. First, raw material including high purity elemental Mg and Zn, Mg-30Gd (wt.%), Mg-30Y (wt.%) and Mg-25Zr (wt.%) with designed volume fractions were melted and casted in to two cylindrical billets, respectively. Then, these cast billets were held at 420 °C for 4 h isothermally and extruded into bars with a speed of 0.4 mm/s. The diameters of cylindrical billets and extruded bars were 155 and 30 mm, respectively. The micrographs of these hot-extruded Mg-RE alloys are shown in Fig. 1. To determine the semisolid temperature ranges of these alloys, differential scanning calorimetry (DSC) analysis was conducted using a LINSEIS PT1600 high temperature differential scanning calorimeter. Based on the result of DSC analysis, the liquid fractions of two Mg-RE alloys at different temperatures are shown in Fig. 2. The solidus and liquidus temperature of alloy A are 522 and 635 °C, respectively. Meanwhile, the solidus and liquidus temperature of alloy B are 503 and 595 °C, respectively.

Reheating experiments and compression tests of two Mg-RE alloys were conducted on a multistage hot compression test machine under the protection of a nitrogen atmosphere. Cylindrical specimens were cut from the extruded bars. The diameter and height of these cylindrical specimens were 8 and 12 mm, respectively. A

cylindrical induction coil and a k-type thermocouple were used to control and measure the temperature of specimens during experiments. A pair of cylindrical ceramic dies covered by lubricated mica pads were used to fix and compress the specimen in reheating experiment and compression test, respectively. To freeze the microstructure of specimen at high temperature, specimen was cooled rapidly by cold water in the end of reheating experiment and compression test.

Table 1. Chemical compositions of Mg-RE alloys, wt%.

	Gd	Y	Zn	Zr	Mg
Alloy A	8.20	4.48	3.34	0.36	Bal.
Alloy B	3.75	5.15	3.05	0.75	Bal.

Figure 1. Micrographs of different hot-extruded Mg-RE alloys at room temperature

Figure 2. Liquid fractions of different hot-extruded Mg-RE alloys at different temperatures

Different specimens processed under various experimental conditions were polished and etched in 4% nitric acid/alcohol solution. Their microstructures were observed using a field-emission scanning electron microscopy (FE-SEM) system. The qualitative and quantitative analyses of the alloying elements in the specimens were conducted using an energy-dispersive X-ray spectroscopy (EDS) system. The constituent phases and their volume fractions in these specimens were analyzed by X-ray diffraction (XRD) analysis using an X-ray diffractometer with a collimator of $\Phi 800 \mu\text{m}$. Data was collected using CuK α radiation in the 2θ range of 20–90 deg.

3 Results and discussion

3.1 Starting materials

As shown in Fig. 1a, plate-shaped and lamellar-shaped eutectic compounds were observed in alloy A. As shown in Fig. 1b, block-shaped eutectic compounds with a large size and string-shaped eutectic compounds with a small size were observed in alloy B. In both alloys A and B, eutectic compounds were distributed parallel to the extrusion direction. According to the results of XRD shown in Fig. 3, lamellar- and plate-shaped eutectic compounds in alloy A were identified as Mg₁₂(RE, Zn) and Mg₅(RE, Zn) respectively. The block- and string-shaped eutectic compounds in alloy B were both identified as Mg₅(RE, Zn). The inhomogeneous distribution and the morphologies of eutectic compounds in these alloys were attributed to the casting and subsequent hot extrusion processes [10]. During casting, the dendritic mode solidification of metal crystals resulted in the microsegregation of alloying elements between the interdendritic and dendritic regions. Eutectic compounds formed in the interdendritic regions with high contents of alloying elements. The dendritic structure was broken up by the subsequent hot extrusion. Thus, the eutectic compounds distributed parallel to the extrusion direction. The lamellar-shaped Mg₁₂(RE, Zn) eutectic compounds were only observed in alloy A, owing to the different chemical compositions of these two alloys.

Figure 3. XRD patterns of different hot-extruded Mg-RE alloys at room temperature

3.2 Reheating experiment

To investigate the microstructural evolution of two Mg-RE alloys during reheating to their semisolid temperatures, specimens were reheated to 520, 540, 560, and 580 °C with a heating rate of 20/s and held isothermally for 20 s. The microstructures of specimens rapidly cooled from different temperatures are observed in longitudinal cross sections and shown in Fig. 4. Volume fractions of different eutectic compounds in different specimens cooled from different temperatures are listed in Table 2. The investigated the morphological change of α -Mg grains during reheating quantitatively, the sizes and shape factors of α -Mg grains in different specimens cooled from different temperatures were measured by image analysis software, the results are shown in Fig. 5. size D and shape factor F, which describes the sphericity of the solid particles, are respectively defined as following [11]:

$$D = \frac{\sum 2(A_1/\pi)^{0.5}}{N} \quad (1)$$

$$F = \frac{\sum 4\pi A_1/P_1^2}{N} \quad (2)$$

Here A_i are the areas of the solid particles, P_i are the perimeters of the solid particles and N is the number of solid particles. In the case of perfectly spherical grains, F has a value of 1. As shown in Fig. 5, higher reheating temperature resulted in smaller size but higher shape factor of α -Mg grains in both alloy A and B.

Figure 4. Micrographs of different hot-extruded Mg-RE alloys cooled from different temperatures.

As shown in Fig. 4a, the lamellar-shaped $Mg_{12}(RE, Zn)$ eutectic compounds almost dissolved in the α -Mg matrix of alloy A at 520 °C. Meanwhile, part of $Mg_5(RE, Zn)$ eutectic compounds dissolved in the α -Mg matrix of alloy A at 520 °C, as shown in Fig. 4b. Owing to the high alloying elements content in these alloys, atom positions were not enough for sufficient dissolution of eutectic compounds. The dissolution of plate-shaped $Mg_5(RE, Zn)$ eutectic compounds mainly took place on their edges and resulted in the change of their geometric morphologies. As shown in Fig. 4c, extrusion direction could be identified by the directional distribution of eutectic compounds in alloy A. According to Table 2, the volume fraction of $Mg_5(RE, Zn)$ eutectic compounds in the specimen cooled from 540 °C is lower than that of the specimen cooled from 520 °C. The decreasing volume fraction of $Mg_5(RE, Zn)$ eutectic compounds was caused

by increasing atom positions for dissolution in alloy A reheated to 540 °C. As shown in Fig. 4d, some α -Mg grains in alloy B were surrounded by newly formed eutectic compounds, when the reheating temperature was increased to 540 °C. According to their morphology, the newly formed eutectic compounds were presumed to be $(Mg, Zn)_3RE$. This presumption was confirmed by the increased volume fraction of $(Mg, Zn)_3RE$ eutectic compounds in the extruded specimen when the reheating temperature was increased from 520 to 530 °C, as shown in Table 2.

Table 2. Volume fractions of different eutectic compounds in different specimens cooled from different temperatures.

	Temperature (°C)	Volume fraction (%)		
		$Mg_{12}(RE, Zn)$	$Mg_5(RE, Zn)$	$(Mg, Zn)_3RE$
A	Room temperature	2.8 ± 2.0	22.4 ± 1.0	0.0
	520	0.0	20.2 ± 1.0	0.0
	540	0.0	16.8 ± 1.0	2.8 ± 2.0
	560	0.0	1.8 ± 1.0	12.6 ± 2.0
	580	0.0	2.0 ± 1.0	17.5 ± 2.0
B	Room temperature	0.0	18.4 ± 2.0	0.0
	520	0.0	4.3 ± 1.0	6.6 ± 2.0
	540	0.0	0.0	13.2 ± 3.0
	560	0.0	0.0	23.6 ± 3.0
	580	0.0	0.0	43.3 ± 5.0

According to Fig. 2, the liquid fraction of alloys A and B increased to 18% and 32% at 560 °C, respectively. More eutectic compounds were observed in the rapidly cooled specimens as shown in Figs. 4e and 4f. It means that when the reheating temperature increased to 560 °C, more partial melting occurred on grain boundaries and resulted in more liquid films in these regions. The increasing liquid phase connected and formed liquid network surrounding the discrete solid particles. The distribution of eutectic compounds in these specimens no longer exhibited any directionality. XRD patterns of alloys A and B cooled from 560 °C are shown in Fig. 6. According to Fig. 6 and Table 2, the eutectic compound networks in these specimens were mainly composed of $(Mg, Zn)_3RE$. The volume fraction of $(Mg, Zn)_3RE$ in alloys A and B increased with the increasing reheating temperature. Moreover, when the reheating temperature increased from 540 to 560 °C the volume fraction of $Mg_5(RE, Zn)$ eutectic compounds in alloy A decreased rapidly. The decrease of $Mg_5(RE, Zn)$ eutectic compounds is estimated as a dynamic process. During reheating, $Mg_5(RE, Zn)$ kept dissolving in adjacent Mg grains and made the edge regions of Mg grains rich in alloying elements. These regions of Mg grains melted and resulted in increasing volume fraction of liquid phase at elevated temperature. Part of $Mg_5(RE, Zn)$ also dissolved in the neighboring liquid network simultaneously. During subsequent rapid cooling process, liquid phase mainly transformed to $Mg_3(RE, Zn)$ eutectic compounds and α -Mg. In compared with $Mg_5(RE, Zn)$, $Mg_3(RE, Zn)$ contains less Mg element but more alloying elements. More Mg atoms were released during reheating and subsequent cooling processes. Above description could explain why the measured volume fraction of total

eutectic compounds in alloy A decreased rapidly when the reheating temperature increased from 540 to 560 °C.

Figure 5. Sizes and shape factors of α -Mg grains in different specimens cooled from different temperatures

When reheating temperature was increased to 580 °C, the liquid fraction of alloys A and B increased to 25% and 84%, respectively. Homogenous microstructures containing an equiaxed spherical Mg grains surrounded by uniform eutectic compound networks were observed in rapidly cooled specimen of alloy A, as shown in Fig. 4g. According to Table 2, $Mg_3(RE, Zn)$ and $Mg_5(RE, Zn)$ were the main eutectic compounds in the alloy A cooled from 580 °C. Both the volume fractions of these eutectic compounds increased when the reheating temperature increased from 560 to 580 °C. This phenomenon was attributed to the continuing partial melting of the specimen. During the rapid cooling, part of the liquid phase solidified as the edge regions of Mg grains. A systematic investigation on the partial solidification of liquid phase during cooling was done by researchers from Germany [12, 13], solidified parts in the grains cannot be distinguished from former solid particles by normal methods. However, the dendritic casting structures dominated in the alloy B cooled from 580 °C, as shown in Fig. 4h. According to Table 2, $(Mg, Zn)_3RE$ with the largest volume fraction were measured in alloy B cooled from 580 °C. On the basis of the description above, the illustration of microstructural evolution of alloys A and B during reheating and rapid cooling processes are illustrated as Figs. 7 and 8, respectively.

Figure 6. XRD patterns of different hot-extruded Mg-RE alloys cooled from 560 °C.

Figure 7. Illustration of microstructural evolution of alloy A during reheating and rapid cooling processes

Figure 8. Illustration of microstructural evolution of alloy B during reheating and rapid cooling processes.

3.2. Semisolid compression

Specimens of two Mg-RE alloys were heated to 520, 530, 540, 550, 560, 570, and 580 °C with a heating rate of 20/s and compressed with a strain rate of 1/s to 70% height reduction after isothermal holding for 20 s. The time-load-stroke data were recorded and calculated using following equations by a computer system.

$$\sigma = \frac{F(H_0 - \Delta H)}{\pi \cdot R^2 \cdot H_0} \quad (3)$$

$$\varepsilon = \ln \frac{H_0 - \Delta H}{H_0} \quad (4)$$

where H_0 and R_0 are the original height and radius of the specimen, respectively, and F and ΔH are the measured

load and stroke, respectively. Owing to the liquid fraction of alloy B was too high when forming temperature exceeded 560 °C, the measured values of the load were too low to calculate the flow stress. The calculated results for the cast and extruded specimens are shown in Fig. 9.

The thixoforming behaviors of the semisolid slurries were affected by their liquid fractions and the morphologies of the solid phases [14]. When thixo-compression tests were conducted at lower semisolid temperatures, the outflow of the liquid phase with a lower volume fraction was difficult. Plastic deformation of the solid particles dominated during thixo-compression and resulted in higher values of the flow stress. As shown in Figs. 4 and 5, the semisolid slurries containing solid particles more spherical profiles and liquid phases with higher volume fraction were obtained at higher temperatures. Because liquid phase with a higher volume fractions exhibited excellent fluidity, liquid phase flowed outwards under forming load. Thus, outflow of the liquid phase and sliding and rotation of the spherical solid particles occurred during semisolid compression and resulted in lower values for the flow stress. Because the liquid fraction of alloy A was higher than that of alloy B at same temperature, lower value of flow stress was obtained in semisolid compression of alloy B at each forming temperature.

In order to express the yield strength (σ_y) of a semisolid slurry with a specific solid fraction (f_s) during thixoforming, following equation was employed [15].

$$\sigma_y = A \exp(Bf_s) \quad (5)$$

Here, A and B are the material constants. The relationship between the yield strength and liquid fraction (f_l) can be described by the following equation:

$$\sigma_y = A \exp[B(100 - f_l)] \quad (6)$$

Figure 9. Flow stress-true strain curves of different hot-extruded Mg-RE alloys compressed at different temperatures.

To obtain the values of A and B, Equation 6 was transformed to Equation 7.

$$\ln \sigma_y = \ln A + B(100 - f_l) \quad (7)$$

Based on the flow stress-true strain curves shown in Fig. 9, the values of the material constants for the two Mg-RE alloys were calculated by a linear fitting method. The yield strengths for alloys A and B in the temperature range from 520 to 580 °C are described by Equations 8 and 9, respectively. The yield strengths obtained from the experiments and calculations are shown in Fig. 10.

$$\sigma_y = 0.03660 \exp[0.07213(100 - f_l)] \quad (8)$$

$$\sigma_y = 0.02256 \exp[0.08686(100 - f_l)] \quad (9)$$

Figure 10. Yield strengths of different hot-extruded Mg-RE alloys with different liquid fractions during thixoforming

4 Conclusion

The results obtained in this study are concluded summarized as follows:

The partial melting behaviors of two hot-extruded Mg-RE alloys were investigated experimentally. Homogenous spherical semisolid slurries of alloys A and B were obtained when these alloys were reheated to 580 °C and 560 °C, respectively.

Phase transformation and arrangement of alloying elements took place during reheating and subsequent cooling of two hot-extruded Mg-RE alloys. These microstructural behaviors were affected by the chemical compositions of these alloys.

The thixoforming properties of the semisolid slurries were not only affected by the liquid fraction, but also depended on the distribution and morphologies of solid particles. Semisolid slurry with higher liquid fractions and more globular solid particles exhibited better formability.

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