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Letter

Microstructure evolution and precipitation of Al–12Mg alloy solidified under high pressure

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ABSTRACT

The microstructure evolution and precipitation process of Al–12Mg solidified under high pressure were investigated. The results show that the amount of β-Al_3Mg_2 phase decreases with increasing pressure and a supersaturated Al(Mg) solid solution is formed under 2 GPa. The distribution of Mg in the solid solution is inhomogeneous. The Mg concentration in the interdendritic region extends up to about 16 wt.%, which is higher than that in the dendrite. Microstructures of sample annealed up to different temperatures indicate that the precipitation process in Al–12Mg solid solution takes place in a non-uniform manner during the heating process, i.e. the β′and β phases are formed in the interdendritic region but no precipitates can be observed in the dendrite.

1. Introduction

Al–Mg alloys are one of the most familiar metallic materials used for commercial wrought and casting alloys due to its high corrosion resistance, mechanical properties and formability, which can transform to be supersaturated solid solutions through solution treatment [1], rapid solidification [2] and solid-state processing [3–5]. The ageing behavior of supersaturated solid solution has been studied extensively and a four-stage process is reported as following [6–11]:

$$\text{Solid solution} \rightarrow \text{GP zones} \rightarrow \beta'' \rightarrow \beta' \rightarrow \beta$$

where GP zones stand for Guinier–Preston zones, β″is an intermediate phase, sometimes described as an ordered GP zone; probably having an $L1_2$ structure (Al_3Mg) [12], The β′phase is a semi-coherent intermediate phase (approximate composition Al_3Mg_2) with a hexagonal unit cell ($a = 1.002$ nm, $c = 1.636$ nm) [13] and the β phase is a complex metallic alloy, i.e. an intermetallic compound with a large unit cell ($a = 2.824$ nm) [14].

Pressure is an essential thermodynamic parameter which can influence the solidification process. Solidification of metals under high pressure can change the distribution of second phase, uniformity of composition, decrease the grain size, and obtain non-equilibrium microstructures [15–19]. For example, Al(Mg) supersaturated solid solution with 21.6 at.% Mg can be prepared by solidified under 2 GPa [17]. It is believed that Al–12 wt.% Mg can transform to be a single solid solution solidified under high pressure and its precipitation process should be different compared with that of alloys prepared by other methods, such as solution treatment and rapid solidification.

In the present work, Al–12 wt.% Mg was chosen to study the microstructure evolution solidified under high pressures and a supersaturated Al(Mg) solid solution was obtained under 2 GPa. The aging behavior of the Al(Mg) solid solution was also investigated and discussed.

2. Experimental procedures

An Al–Mg alloy with 12 wt.% Mg was prepared by conventional casting from 99.99 wt.% pure Al and Mg. The samples for high pressure solidification were cylinders of 20 mm in diameter and 18 mm in length. The experiments were carried out by a tungsten-carbide six-anvil apparatus and the pressures were 1 GPa and 2 GPa. Details of high pressure solidification are described elsewhere [17]. The phases were characterized by a Rigaku D/max-RB X-ray diffractometer with monochromatic Cu-Kα radiation. Morphology and compositional homogeneity were examined on an Olympus optical microscopy and a scanning electron microscope (SEM) operated at 20 kV equipped with an energy dispersive X-ray spectrometer (EDX). Samples for calorimetric analysis were in a disk shape of 5 mm in diameter and 1 mm in thickness. The calorimetric measurements were performed with a Netch 449 differential scanning calorimeter. A protective atmosphere of pure argon was used. In order to study the phase evolution during heating process, samples were heated treated in the DSC by continuous heating at 10 K/min through the exothermic peaks and then cooled to room temperature at 100 K/min.

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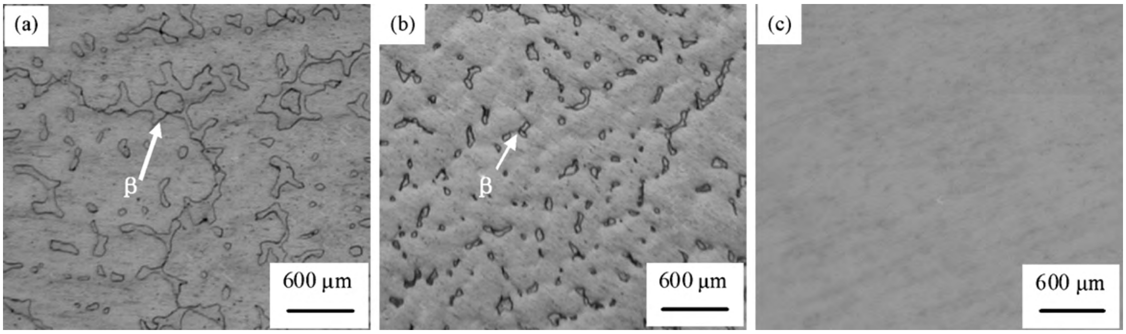

Fig. 1. Microstructures of Al–12Mg alloy solidified under different pressures: (a) normal pressure; (b) 1 GPa; (c) 2 GPa.

3. Results and discussion

3.1. Microstructure evolution and Mg distribution

Fig. 1 shows the microstructures of Al–12Mg alloy solidified under different pressures. The Al–12Mg alloy solidified under normal pressure contains α-Al and β-Al_3Mg_2 as shown in Fig. 1a. It can be seen that the amount of β phase decreases with increasing pressure, and no β phase is present in the alloy under 2 GPa as shown in Fig. 1c.

The X-ray diffraction (XRD) patterns of Al–12Mg solidified under different pressures are shown in Fig. 2a. The reflections of the β phase decrease with increasing pressure and disappear when the pressure is up to 2 GPa according with the observation of microstructure. So it can be concluded that a supersaturated Al(Mg) solid solution is prepared under 2 GPa. The peaks of the α-Al phase shift to the lower angles, indicating that the lattice parameter increases with pressure which is caused by an increase of the solubility of Mg in Al. In addition, peak asymmetry can be seen in the XRD patterns under high pressures. Fig. 2b shows the (3 1 1) and (2 2 2) diffraction peaks of α-Al. It can be seen that these peaks are skew to the left. The most common reasons of peak asymmetry can be given as: chemical heterogeneities, precipitates and inclusions, long-range internal stresses, twining, dislocations and stacking faults [20]. In order to investigate the reason for peak asymmetry, X-ray diffractions of pure Al solidified under normal pressure, 1 GPa, and 2 GPa were conducted and the results are shown in Fig. 2c. No peak asymmetry can be observed, indicating that the peak asymmetry of Al–12Mg solidified under high pressures is caused by the addition of Mg element. It can be concluded that large range of chemical heterogeneities, i.e. an inhomogeneous distribution of Mg element, causes the peak asymmetry of the XRD patterns. Fig. 3 shows the backscattered electron (BSE) image and the line scanning result of Al–12Mg alloy solidified under 2 GPa. The atomic number contrast afforded by the BSE image indicates that the Mg concentration in the interdendritic region is higher than that of the dendrite. The line scanning shows that the concentration of Mg in the interdendritic region increases gradually up to about 16 wt.%, but the dendrite contains only 7–9 wt.% Mg solute. Inhomogeneous distribution of Mg was also found in Al–21.6 at.% Mg alloy solidified under 2 GPa, i.e. the Mg concentration in the interdendritic region increased up to about 30 at.% but only 15 at.% in the dendrite [17]. It is considered that Al–12 wt.% Mg can transform to be a solid solution under 2 GPa due to its less Mg concentration than 15 at.%. But inhomogeneous distribution of Mg solute still exists in the alloy.

In order to study the reason of the peak asymmetry, the Al–12Mg solid solution solidified under 2 GPa was homogenized at 430 °C for

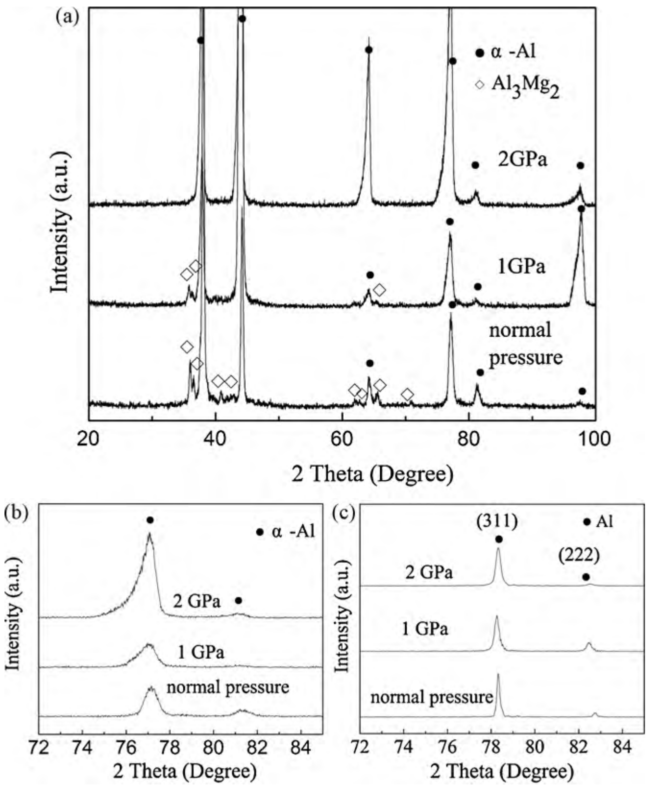

Fig. 2. X-ray diffraction patterns of Al–12Mg and pure Al (a) under different pressures, (b) (3 1 1) and (2 2 2) diffraction peaks of Al–12Mg, and (c) (3 1 1) and (2 2 2) diffraction peaks of pure Al.

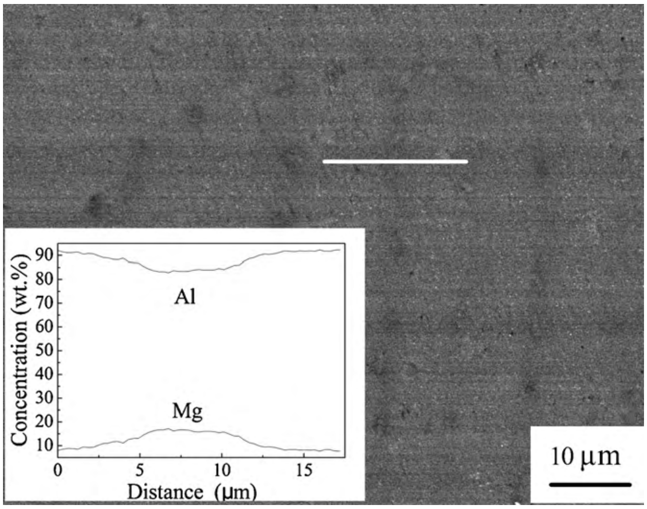

Fig. 3. BSE image and line scanning result of Al–12Mg solidified under 2 GPa.

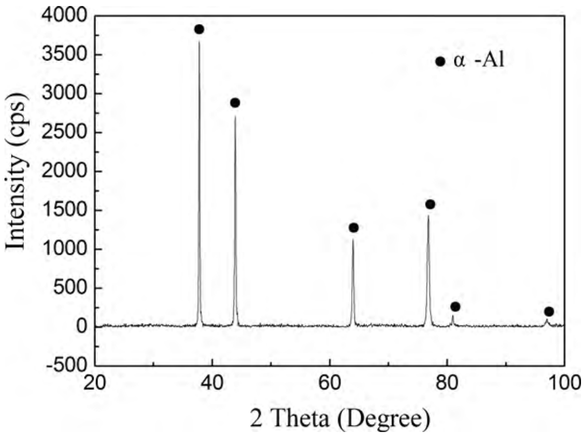

Fig. 4. XRD pattern of Al–12Mg solidified under 2 GPa and then homogenized at 430 °C for 17 h.

17 h and then quenched in ice water. Fig. 4 shows the corresponding XRD pattern and no peak asymmetry can be seen. It further proves that the peak asymmetry is caused by the inhomogeneous distribution of Mg element between the interdendritic region and dendrite.

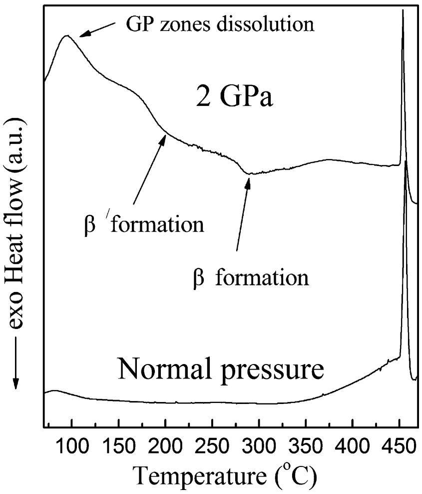

Fig. 5. DSC curves of Al–12Mg solidified under normal pressure and 2 GPa.

3.2. *DSC results of Al–12Mg under different pressures*

Fig. 5 shows the DSC curves of the Al–12Mg solid solution prepared under normal pressure and 2 GPa. The endothermic peak

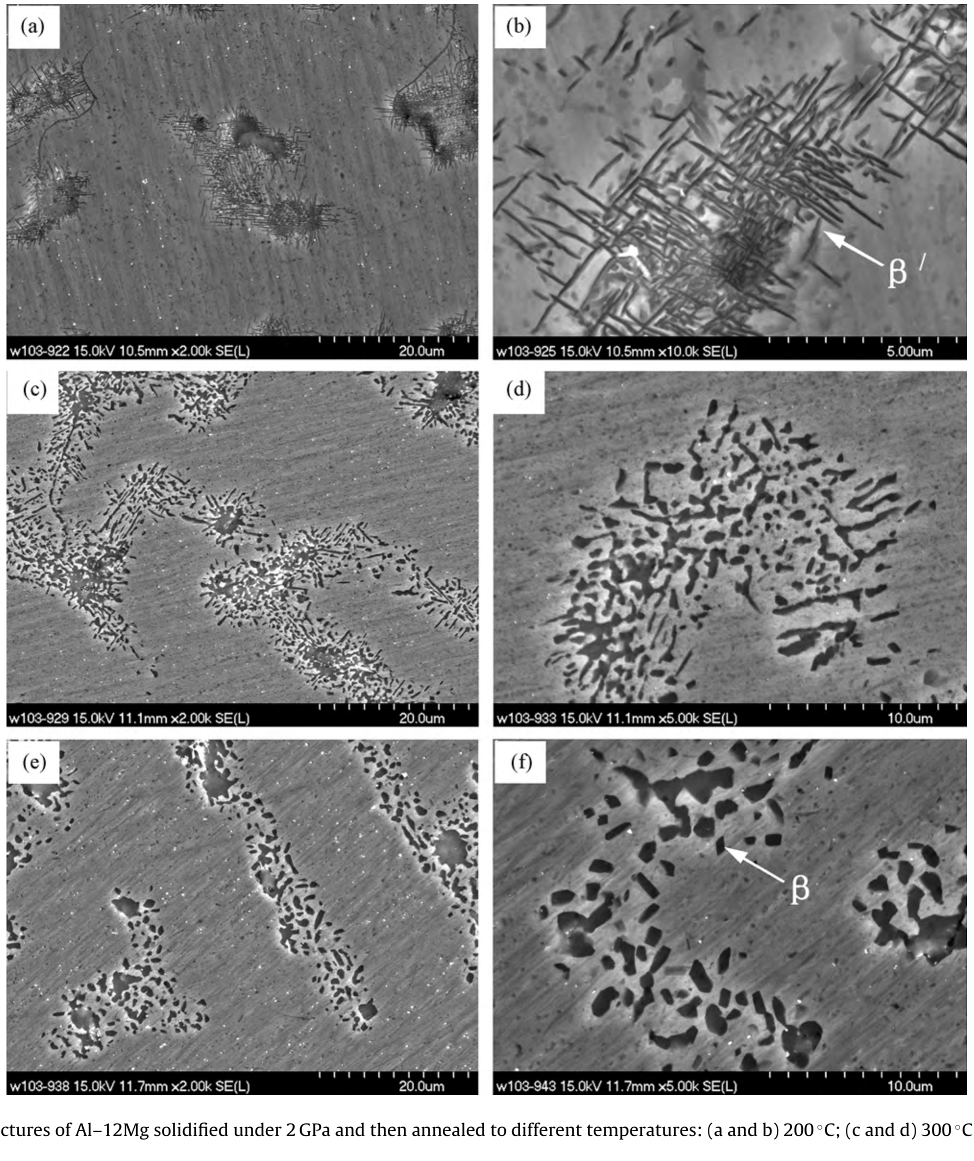

Fig. 6. Microstructures of Al–12Mg solidified under 2 GPa and then annealed to different temperatures: (a and b) 200 °C; (c and d) 300 °C; (e and f) 400 °C.

in the range of 451–461 °C is caused by the dissolution of the β phase in the interdendritic region which is formed during the solidification under normal pressure. No obvious thermal peaks corresponding to the formation of the metastable precipitation can be found in the sample solidified under normal pressure. The DSC curve of Al–12Mg solidified under 2 GPa presents four peaks: two endothermic peak in the range of 80–140 °C and 450–458 °C, and two exothermic peaks in the range of 170–250 °C and 290–350 °C, respectively. DSC curve of the sample presents a first endothermic peak in the temperature range of 80–140 °C because of the dissolution of GP zones formed during the nature ageing of the sample, which is higher than that of the quenched Al–12Mg solid solution (50–80 °C). The kinetics of formation and dissolution of GP zones can be interpreted in terms of nucleation and excess-vacancy enhanced diffusion of magnesium [8]. The reason is probably that less vacancy exists in the alloy solidified under high pressure than that of the quenched alloy. The sharp endothermic peak with measured onset temperature 450.8 °C agrees well with the eutectic temperature for Al-rich Al–Mg alloys [21]. The two exothermic peaks represent the formation of the β′and β phases, respectively [1,9,10].

3.3. Microstructure evolution during heating process

The microstructures of the samples heated to different temperatures are shown in Fig. 6. It is interesting that the β′phase is formed in the interdendritic region at 200 °C which arranges in the case of a Widmannsttäten structure, but no β′ phase can be observed in the dendrite (Fig. 6a and b). It indicates that the precipitation of the β′phase takes place in a non-uniform manner. The β phase is formed when the temperature increases up to 300 °C which also distributes in the interdendritic region (Fig. 6c and d). Only equilibrium β phase particles disperse in the interdendritic region at 400 °C, which is responsible for the eutectic melting at about 450 °C (Fig. 6e and f).

The concentration of Mg in the interdendritic region is about 16 wt.% which is higher than that of the dendrite, i.e. the supersaturation of the interdendritic region is higher than that of the dendrite. The formation of the β′and β phases is a diffusion controlled process. The transformed volume V_p can be defined as the volume of a fully depleted area around a precipitate (with the rest of the matrix undepleted) needed to give a precipitate size equal to the real case with a diffusion zone as following [1,22]:

$$V_p = A_1[G(t - z)]^m \tag{1}$$

where G is the average growth rate, A_1 is a constant which is related to the initial supersaturation, the dimensionality of the growth and the mode of transformation, m is a constant related to the dimensionality of the growth and the mode of transformation, t is time, z is the time that the precipitate nucleated [22]. So the β′and β phases are first formed in the interdendritic region during the heating process due to its higher supersaturation. The distribution of Mg element in the Al–12Mg alloy by solution treatment is nearly homogeneous, resulting in the uniform precipitation during the heating process [9,10]. In liquid-quenched Al–Mg alloys, non-uniform precipitation occurs and bulk and grain-boundary precipitation proceed at different rates due to the occurrence of significant compositional differences in the aluminum matrix [6]. In the present study, the β′and β phases precipitate in the interdendritic region but no precipitation can be observed in the dendrite. A full understanding of the non-uniform precipitation mechanism of Al–Mg solid solution prepared under high pressure requires more theoretical and experimental studies.

4. Conclusions

During the solidification process under high pressure, the amount of β-Al_3Mg_2 phase decreases with increasing pressure and a supersaturated Al(Mg) solid solution is obtained under 2 GPa. The Mg concentration in the interdendritic region is about 16 wt.%, which is higher than that in the dendrite. The precipitation process in Al–12Mg solid solution takes place in a non-uniform manner during DSC heating, i.e. the β′and β phases precipitate in the interdendritic region but no precipitation can be observed in the dendrite due to the inhomogeneous distribution of Mg element.

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