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Effect of RE elements on the microstructural evolution of as cast and SIMA processed Mg–4Al alloy

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ABSTRACT

In the present article, the effect of Rare Earth elements on the microstructural development of as cast and semisolid Mg–4Al alloy produced by SIMA process is studied. Investigation conducted by metallographic observation, scanning electron microscope and quantitative metallographic methods. Results showed that alloy's dendrites turn into larger fully dendritic shape with sharp and narrow arms from equiaxed rosette type as the amount of RE elements increased from 0 through 4%. The effect of RE elements on the microstructure of the treated-alloys was detectable through elimination of vast number of intragranular liquid droplets as well as by decreasing kinetic of microstructural changes. It was shown that the trend of grain coarsening decreased drastically by addition of RE elements to the Mg–4Al alloy during partial remelting at 610 °C. Moreover, the effect of REs on the other parameters such as fraction of liquid, shape factor and particle size was studied.

1. Introduction

Magnesium alloys as the lightest structural metals are attractive for many applications such as automobile, aerospace and railway industries. During recent years, semisolid processes have become popular for industrial and commercial production because of their excellent advantages such as low temperature, low forming force, good quality of parts and so forth, over conventional casting processes [1–3]. Preparation of semisolid slurry is so important since the globular structure has an essential role on the final properties of the product [4]. Strain induced melt activated (SIMA) technique introduces significant commercial advantages of simplicity and low cost. Generally, SIMA process consists of 3 discrete steps [5]: casting of the alloy to achieve a typical dendritic structure, a mechanical work to store an internal energy due to the distortion and partial remelting of the alloy at semisolid temperatures [6]. This process is based on the scientific understanding that high angle grain boundaries induced by plastic deformation and recrystallization will be wetted by liquid metal at the semisolid temperature, resulting in a fine and globular structure [7]. The purpose of the first stage is to obtain billets with suitable shape for further deformation, so the casting temperature is not strictly controlled and there may be many casting defects in the billets. But after the isothermal treatment these defects will decrease and the suitable slurry with a fine and globular structure will be acquired [8].

During recent years, some studies investigated the effect of different process parameters like compression ratio, semisolid temperature and initial cast structure on the final structure and mechanical properties of variety of alloys [1,4,6,9,10]. Also some works has been conducted on the effect of rare earth metals on the microstructure of magnesium alloys [11–14]. However, because of limited semisolid range of the Mg–4Al alloy, a few studies were carried out on the structure of this alloy during semisolid treatment.

The aim of this paper is to investigate the effect of RE elements on the microstructural evolution of as cast and semisolid treated Mg–4Al alloy. In addition, the coarsening kinetics of deformed Mg–4Al and Mg–4Al–2RE alloys in the semisolid state was also studied.

2. Experimental procedures

Commercial-purity magnesium (+99.95%) and aluminum (99.99%) ingots were melted in a mild steel teapot at 720 °C in an electric resistance furnace (3 kW) under protection of a flux (50% $MgCl_2$, 20% KCl, 15% MgO and 15% CaF_2). Different weights of Rare earth metals (0, 2 and 4 wt%) were added to molten metal as mischmetal at 720 °C. The melt was held at 720 °C for 20 min in a furnace to ensure that the rare earth metals were completely dissolved. The composition of mischmetal as presented by the Table 1 contains 88% rare earth, 11.5% zinc and 0.5% other elements.

The alloy was poured into a 250 °C preheated cylindrical permanent mold with internal diameter of 30 mm and height of 250 mm. As cast specimens were machined and cut into samples with the dimensions of 20 mm height and 20 mm diameter. Samples (Mg–4Al and Mg–4Al–2RE) were pressed by a hydraulic press at 250 °C followed by quenching in a water tank. The compression ratio was defined by relative reduction of sample's height ($(H_0 - H)/H_0$) and the induced strain was 0.3. Contact surfaces were lubricated by graphite powder to minimize friction and dead zone. The deformed specimens were held at 610 °C for various times (10, 20, 30 and 40 min) in a salt bath (63.5% KCl and 36.5% $MgCl_2$) where the temperature was controlled

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Table 1
The composition of mischmetal (wt%).

Rare earth				Other elements				
La	Ce	Nd	Pr	Zn	Al	Mg	Si	Fe
55	32	10	3	11.5	0.11	0.16	0.9	0.14

by a K-type thermocouple, placed in the center of the bath near to the specimens. The specimens then rapidly quenched after being isothermally heated at 610 °C. The microstructures and phase distribution were characterized by Olympus BX51M optical microscopy and VEGA II TESCAN scanning electron microscopy (SEM). The observed samples for metallographic and SEM investigations were ground, polished and etched by nital 1% solution. Quantitative analysis was carried out by using Clemex Image Analyzer (version 3.5). 10 representative areas for each sample were studied for microstructural evolutions. Sphericity, particles size and fraction of liquid were measured. The chemical compositions of phases and RE-rich particles were determined with an energy dispersive spectrometer (EDS).

3. Results and discussion

SEM and Optical micrographs of the as cast Mg–4Al alloy containing 0, 2 and 4 wt% of RE are represented in Figs. 1 and 2. As it can be seen in Fig. 1(a), the microstructure of the Mg–4Al alloy consists of α-Mg and β ($Mg_{17}Al_{12}$) phase. α-Mg dendrites are in form of equiaxed rosette (Fig. 2(a and c)). By increasing of RE elements, $Al_{11}RE_3$ intermetallic compounds started to form by depletion of β phase and the shape of the dendrites converted from rosette to fully dendritic with narrow and sharp arms. The grain size became larger (95, 160 and 185 μm for Mg–4Al, Mg–4Al–2RE and Mg–4Al–4RE respectively) as the RE elements increased in the alloy.

Although shape and size of dendrites were changed by increasing of the RE elements, inter-dendritic arm spacing is identical for all alloys (20 ± 1 μm). RE elements show a high tendency to combine with aluminum rather than magnesium [12,15]. This combination led to a large latent heat [10] which reduces the rate of temperature decrease during solidification. Therefore α-Mg dendrites obtain an opportunity to grow into coarse dendritic grains, followed by decreasing of concentration of aluminum in the melt. Therefore, when concentration of aluminum in the liquid diminishes, the difference between the actual and liquidus temperature increases and constitutional undercooling is achieved. In other word, solute aluminum, decreases as the amount of RE increases, i.e.

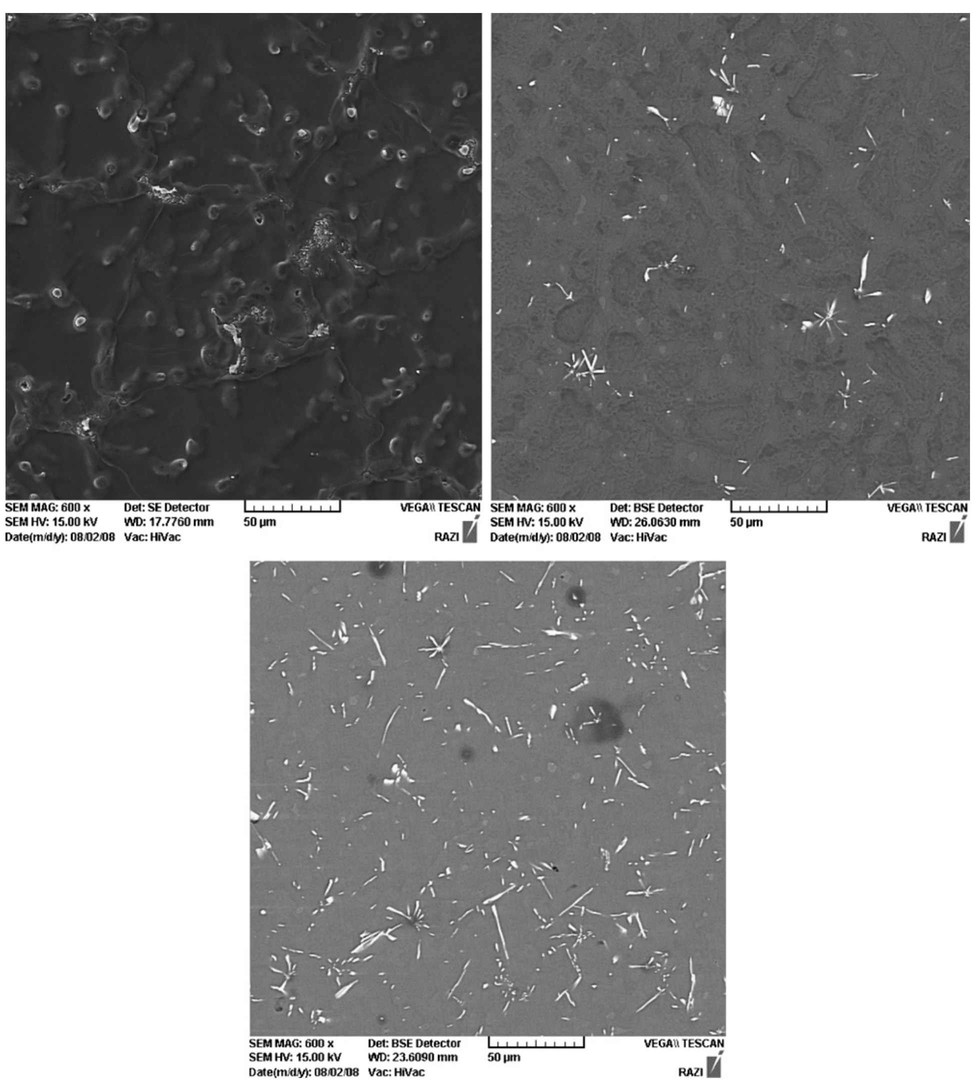

Fig. 1. SEM micrograph of (a) Mg–4Al, (b) Mg–4Al–2RE and (c) Mg–4Al–4RE alloys.

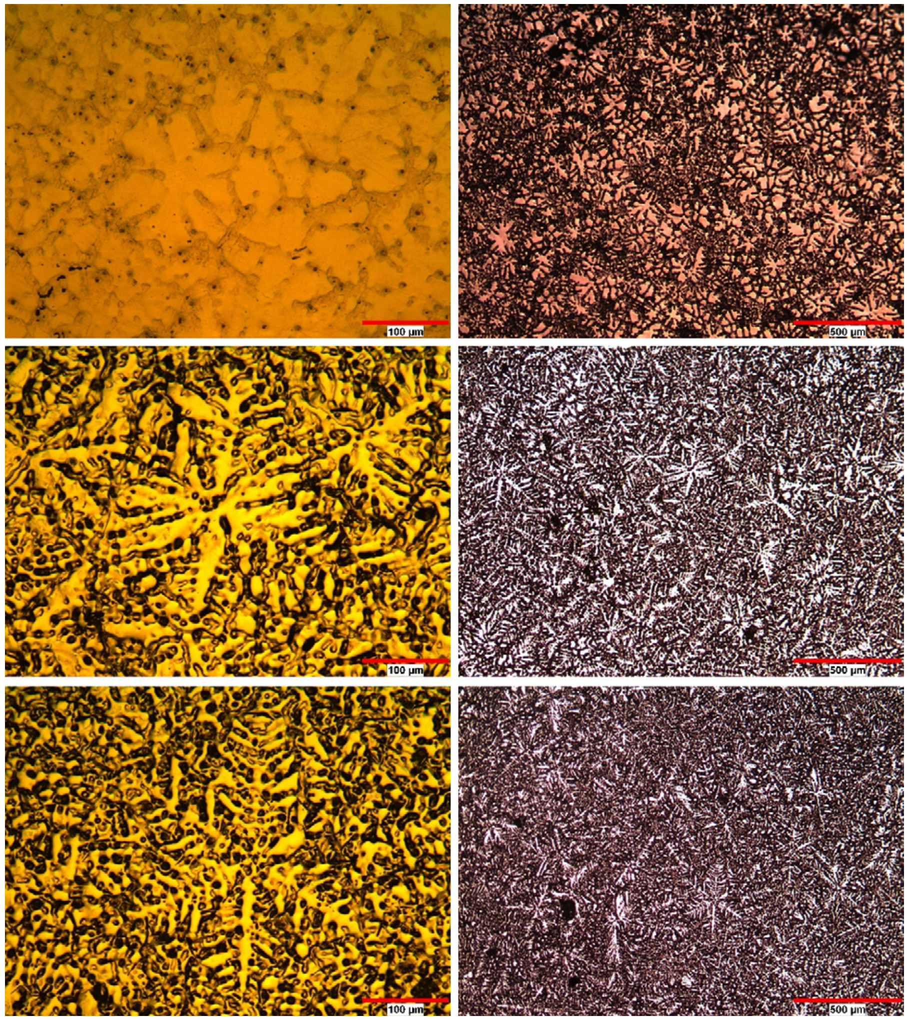

Fig. 2. The microstructure of as cast Mg–4Al with different amount of RE as alloying elements: (a) 0%, (b) 2%, (c) 4%. (d), (e) and (f) are enlarged micrograph of (a), (b) and (c) respectively.

constitutional undercooling increases, and equiaxed morphology of dendrites is replaced with sharp and narrow dendritic arms.

It was assumed in this study that coarsening was controlled by diffusion in the liquid. Volume diffusion controlled coarsening, described by classical LSW equation in Ostwald ripening [16]:

$$D^3 - D_0^3 = Kt \quad (1)$$

where D and D_0 are final and initial particle size respectively, k is coarsening rate and t is the holding time. Experimental results and linear regressions are depicted in Fig. 3. It is clear that experimental values are well fitted to LSW equation, since the R^2 values are close to 1 for both alloys. The values of k obtained from the lines slope which is 48 $\mu m^3/s$, 70 $\mu m^3/s$ and 114 $\mu m^3/s$ for Mg–4Al–4RE, Mg–4Al–2RE and Mg–4Al alloys respectively. It is seen that k value for Mg–4Al–4RE is approximately 2.4 times less than what Mg–4Al is. In other words, rate of coarsening becomes slower by addition of RE elements. Particle growth depends on diffusion or flow of solute atoms between particles of different size [17]. Chen et al. [18] reported that Ce, is enriched at the solid–liquid interface and hinders the atom diffusion rate. Also rare earths are surface-active elements and they can decrease the surface energy of the alloy's melt [19,20].

The microstructure of Mg–4Al, Mg–4Al–2RE and Mg–4Al–4RE alloys after being isothermally treated at 610 °C are shown in Figs. 4–6 respectively. Comparing the microstructures of the reference vs. RE added samples (Figs. 4–6), it is clarified that a vast number of intragranular liquid droplets exist in the reference samples and their sizes are increased by further holding at semisolid temperature. By increasing the holding time, the fine droplets which are formed due to the segregation in the reference samples begin to move within the globules and combine together to form larger droplets in order to decrease their surface energy. As a con-

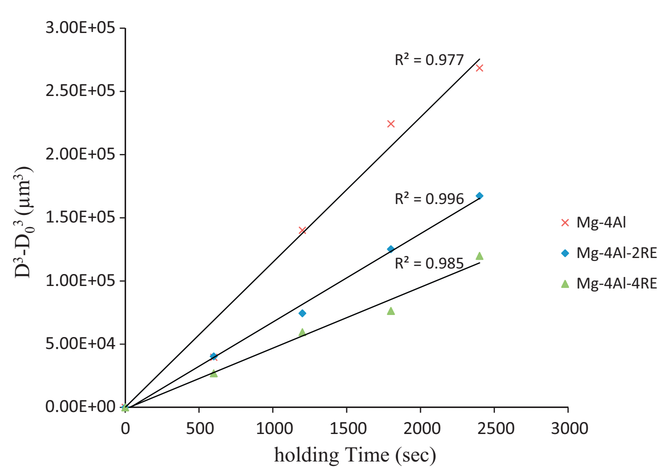

Fig. 3. Effect of holding time at semisolid temperature on the particles spherocity of Mg–4Al–2RE and Mg–4Al alloys.

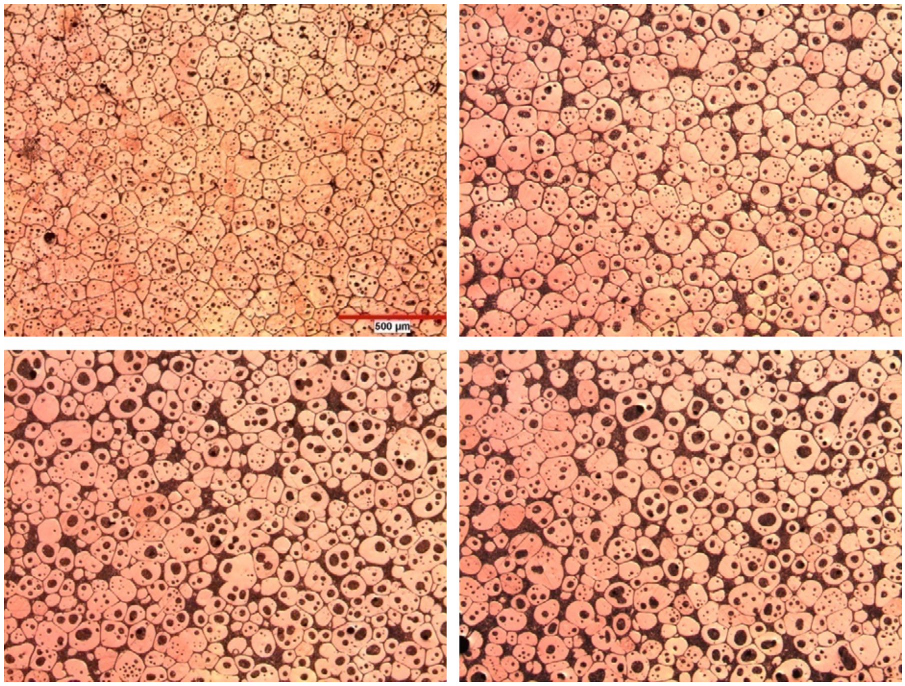

Fig. 4. Microstructure of 30% deformed Mg–4Al alloy after holding at 610 °C for (a) 10, (b) 20, (c) 30 and (d) 40 min.

sequence of reduction of the solid–liquid surface energy, it can be seen in Fig. 4(d).

Addition of RE elements into Mg–4Al alloy led to a drastically decrease in number and size of liquid droplets within the globules (Figs. 5 and 6). Two reasons may be suggested for these observations. The first one might be due to the origin of droplets. Since the droplets are basically formed because of inhomogenity and chemical segregation [21], RE elements reduce probability of chemical segregation through combining with aluminum during the solidification process and as a result decrease the number of droplets remarkably. In addition, this microstructural modification may be related to spherodization process. In other word, since RE containing alloys dendrites have wide variety of curvatures, then ripening would be the dominant spherodization process. Thus the entrapped liquids within globules are fewer than Mg–4Al which because of its rosette-dendrites has less diversity of curvatures which benefits from coalescence as the main spherodization process.

As it was discussed, addition of RE elements in to the melt, results in formation of larger as cast grains in Mg–4Al alloy. As cast grain and isothermally semisolid treated particle size are shown in Fig. 7. It can be seen that after pressing and isothermally holding at semisolid temperature, the size of the new-particles decreases

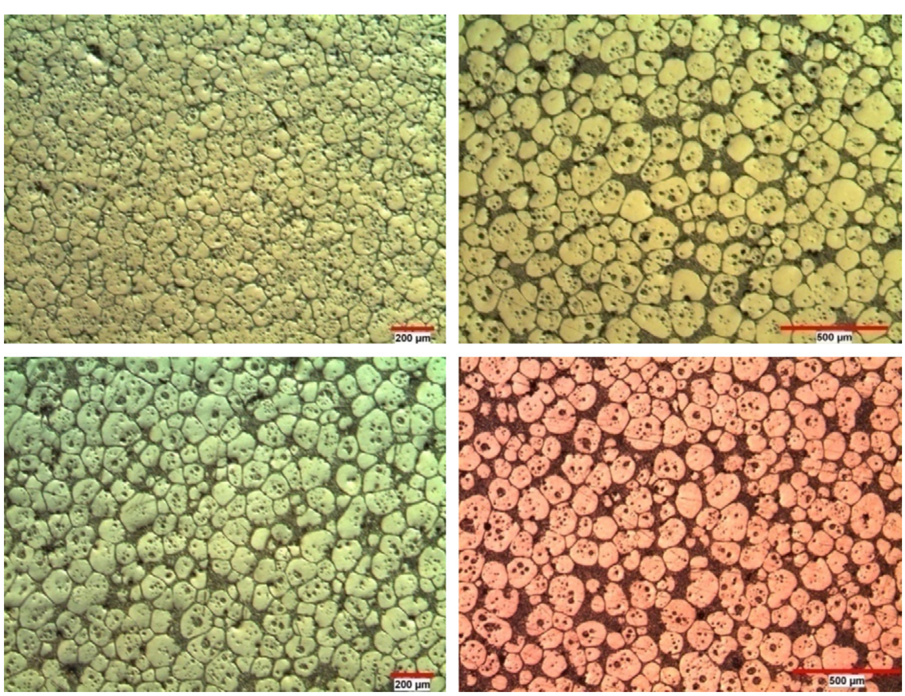

Fig. 5. Microstructure of 30% deformed Mg–4Al–2RE alloy after holding at 610 °C for (a) 10, (b) 20, (c) 30 and (d) 40 min.

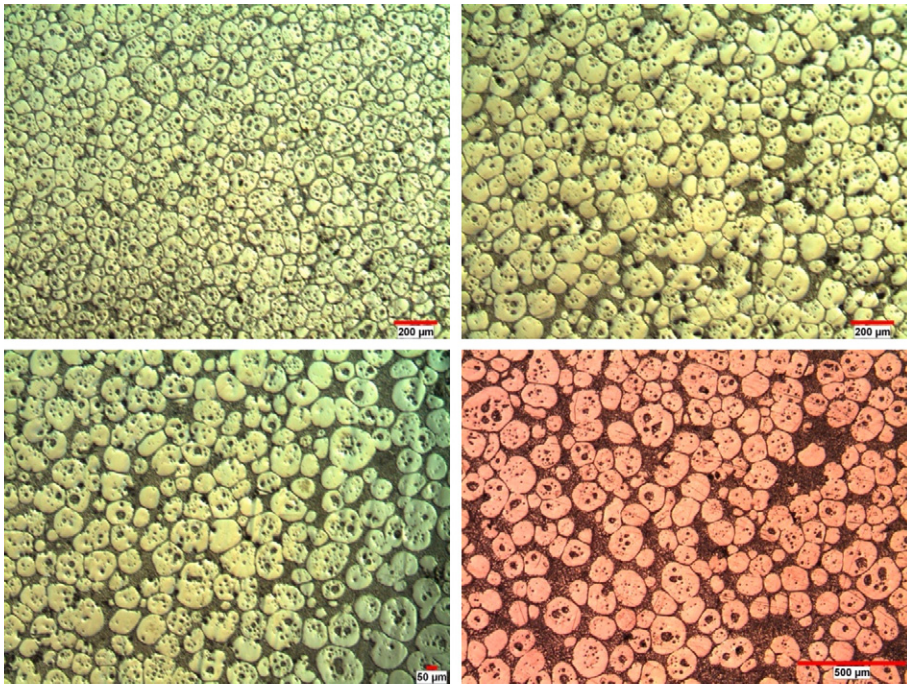

Fig. 6. Microstructure of 30% deformed Mg–4Al–4RE alloy after holding at 610 °C for (a) 10, (b) 20, (c) 30 and (d) 40 min.

rapidly and then increases gradually by prolonging the holding time. Since the Ostwald ripening is a diffusional phenomenon, diffused mass is increased which is a consequence of melting and disappearance of finer particles or detached dendrite's arms. Therefore, size of the larger particles gradually increases by absorbing the dissolved elements. It has to be noticed that isothermally holding of the samples at semisolid temperature led to formation of smaller grains in Mg–4Al–2RE alloy, in spite of their larger size in the as cast condition comparing the Mg–4Al alloy. Formation of finer particles can be related to the total stored energy in alloys during compression. Dendritic structure of RE containing alloy was easy to deform due to its narrow dendrite arm. As a result, more energy is stored in the alloy. This internal energy provides driving force for recrystallization; so by increasing of stored energy, the finer particles are formed. In other word, increasing of internal energy, results in large nucleation and growth rate [10] which cause smaller new-formed particles.

Increasing of the stored energy, results in acceleration of diffusional phenomena, by increasing dislocations and vacancies which act as diffusional channels for alloying elements [9,10]. Concentration of alloying elements boosts along these channels and as a result melting temperature decreases locally which results in more liquid formation for the more compressed specimens. For the same holding time, it is expected that the amount of liquid fraction (include both the liquid inside the globules and in between the particles) to be higher in the alloy with more stored energy (Fig. 8).

Shape factor of globules gradually enhance in both alloys during holding at semisolid range, whereas level of shape factor for RE containing alloys are always lower than Mg–4Al (Fig. 9). This may be affected by forming of initial as cast dendrites. In addition, the initial rosette shape microstructure of the Mg–4Al needs less time to change to a spheroidal structure comparing the required time to form dendritic structure [17].

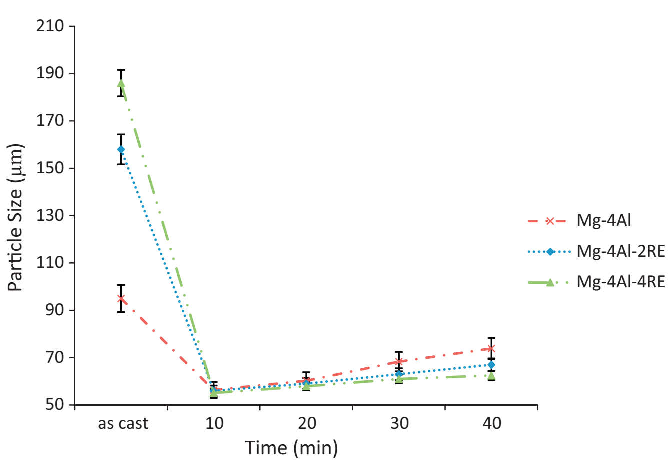

Fig. 7. Effect of holding time at semisolid temperature on the particle size of Mg–4Al–2RE and Mg–4Al alloy.

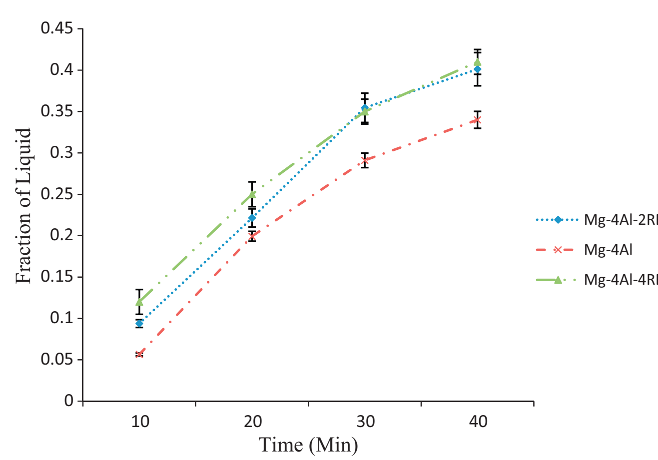

Fig. 8. Effect of holding time at semisolid temperature on the fraction of liquid for Mg–4Al–2RE and Mg–4Al alloys.

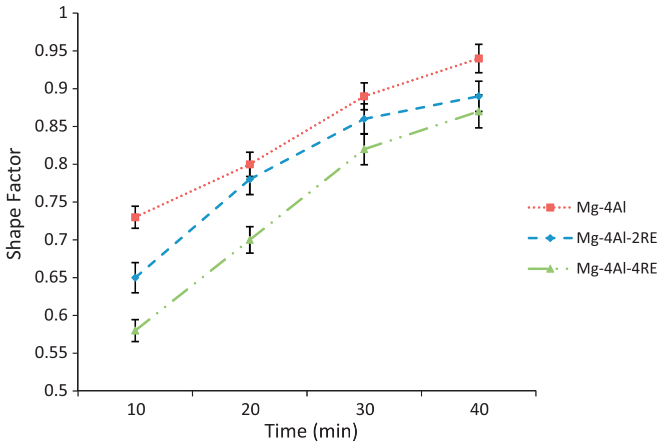

Fig. 9. Effect of holding time at semisolid temperature on the particles spherocity in Mg–4Al–2RE and Mg–4Al alloys.

4. Conclusion

The cast microstructures varied remarkably by addition of rare earth elements, from equiaxed rosette dendrite to fully dendritic structure with sharp and narrow arms even though the arm spacing was identical. Also, they cause an augmentation of dendrites. The elimination of intragranular liquid droplets is a result of REs addition. It was shown that the coarsening rate of α-Mg grains decreased drastically by addition of Rare Earth elements to the Mg–4Al alloys during semisolid holding. In addition, more liquid formed in the RE containing alloy during partial remelting because of the effect of RE elements on the solidification behavior of the alloy. Prolonging of the time has an identical effect on each alloy and brings about grain coarsening and increasing of shape factor.

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