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Microstructures and mechanical properties of Mg–Al–Zn–Ca alloys fabricated by high frequency electromagnetic casting method

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Abstract The microstructures and mechanical properties of AZ31 and 1 wt%Ca-containing AZ31 billets fabricated using EMC (Electromagnetic Casting) and EMS (Electromagnetic Stirring) were examined. The results show a great potential of producing high-quality surface magnesium billets with fine-grained microstructure at a relatively high casting speed. Application of EMC + EMS for production of the 1 wt%Ca-AZ31 alloy billet with a diameter of 150 mm produced a reasonably homogeneous microstructure composed of fine grains with an average size of 45 μm. Attainment of the fine-grained and homogeneous microstructure by EMC + EMS was attributed to reduction of temperature gradient and fragmentation of dendrite structure under electromagnetic force. Strength of the EMC and EMC + EMS 1 wt%Ca-AZ31 billets was higher than that of the EMC AZ31 billet due to the grain size and particle strengthening effects.

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Introduction

Magnesium alloys have wide applications in weight-saving applications in transportation vehicles and parts of electronic, information, and communication fields. The majority of magnesium components are currently produced using die casting technique. To improve productivity and quality, however, products production by plastic working on rods or sheets is highly desired. Compared with an aluminum billet, however, the cost of a Mg billet is higher and extruding the Mg billet is more expensive. Therefore, Mg billets with good extrusion ability need to be developed to reduce the cost of Mg products. Continuous casting technology for high-quality surface billets with fine-grained and homogeneous microstructure can be a solution for accomplishing this purpose.

The latent heat of fusion per weight (J/g) of magnesium is similar to that of aluminum. When the latent heat of fusion per volume is considered, however, it is recognized that magnesium is more rapidly solidified in the mold during casting and thus probability of forming surface defects is higher. Defects such as oscillation marks and surface cracks seriously affect the productivity because they should be removed by scarping or grinding before the billets are passed on to the next process for extrusion or forging.

Electromagnetic force can be used to control the solidification process for improving the surface quality and microstructure of the billets. There are two electromagnetic casting (EMC) methods. One is to use high-frequency magnetic field of tens of kHz [1, 2], and the other is to use low frequency magnetic field of 60–200 Hz [3]. In the former method, the meniscus stability is good but the EMC apparatus needs special molds and power devices. On the other hand, the latter method has a major challenge in stabilizing the meniscus. Up to now, there are several reports on the effect of low frequency electromagnetic force on microstructure of magnesium castings [4, 5], but few reports on the effect of high-frequency electromagnetic force are available.

The addition of calcium is known to be effective for the ignition prevention of magnesium alloys during casting [6]. This is because it retards the oxidation rate during melting process by forming thin and dense CaO film on the surface of the molten alloy [5]. The Ca addition is also one of the effective ways of refining the microstructure of magnesium casting. It was shown that with increasing amount of added Ca, the grain refinement tendency of AZ91D alloy became intensified [7].

The aim of the present work is to examine the effect of high-frequency electromagnetic force on the microstructure and mechanical properties of the 1 wt%Ca-AZ31 alloy castings. The solidification structure and mechanical properties of the EMC billets with or without EMS were investigated and compared.

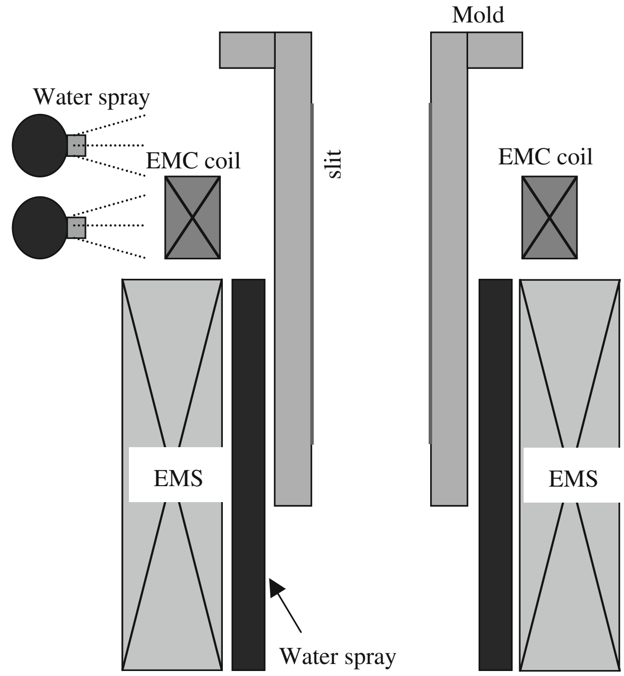

Fig. 1 A schematic view for the experimental devices

Experimental methods

The AZ31 alloy billets with and without Ca (3.02%Al–1.07%Ca–0.90%Zn–0.33%Mn–bal Mg and 3.04%Al–0.89%Zn–0.34%Mn–bal Mg in wt%) were fabricated using the following procedures. The AZ31 alloy ingots were melted in an electrical resistance furnace using a mild steel crucible under protection of the mixed gas of CO_2/0.5 wt%SF_6. When Ca was added, Mg–30 wt%Ca master alloys were used, which were melted at 1073 K. After holding for 10 min. at 1003 K, the melt was poured into a copper cylindrical mold with an inside diameter of 150 mm and a length of 200 mm. A schematic view for the experimental devices is shown in Fig. 1. The slit was machined in the mold along the longitudinal direction. Under the inductor coil, the electromagnetic stirring (EMS) device was installed. The current-frequencies for the EMC and EMS used in the study were 1000 A–20 kHz and 150 A–20 ~ 30 Hz, respectively. Cooling water was sprayed from the mold outside for cooling of the mold part, while the billet was directly cooled by water spray below the mold bottom (=30 L/min).

Samples for optical microscopy were etched with a solution of 4 vol% nitric acid + ethyl alcohol for revealing structure and 5 g picric acid + 5 g acetic acid + 100 mL ethyl alcohol. For TEM observation, a JEOL 2010 transmission electron microscope (200 keV) was used. Electron Probe X-ray Micro-Analysis (EPMA) was used to identify the second phase. The fraction of equilibrium phases in the 1 wt%Ca-AZ31 was calculated by using the thermodynamic phase diagram software (Pandat) linked with thermodynamic database for Mg alloys. The grain sizes of the billets were determined using the image analyzer. Tensile tests were conducted at room temperature on the tensile specimens of dog-bone geometry with the 5-mm gage length, 4-mm width, 2-mm thickness, and 2-mm shoulder radius, which were extracted from different positions on cross-sectional area of the billets.

Results and discussion

Following is a brief explanation about the concept of soft contact under the high-frequency magnetic field. As shown in Fig. 2, when electric current is applied to the coil, magnetic field is induced in the mold and electric current is induced by the magnetic field. This not only heats (Joule heating) the molten metal but also generates the electromagnetic force (Lorentz force) in the molten metal. The electromagnetic force enlarges the meniscus curvature of the molten metal in contact with the mold, thereby improving inflow of the mold flux and reduces contact pressure between the shell and mold. As the meniscus is heated by Joule heating, a thin solidified shell forms in the lower part of the meniscus and thus a hook, which is the root of the oscillation mark, can be prevented from taking place. In EMC, stable maintenance of the molten metal at a high casting speed is possible because of formation of the uniformly solidified shell without surface crack. Fine grained microstructure can be produced through

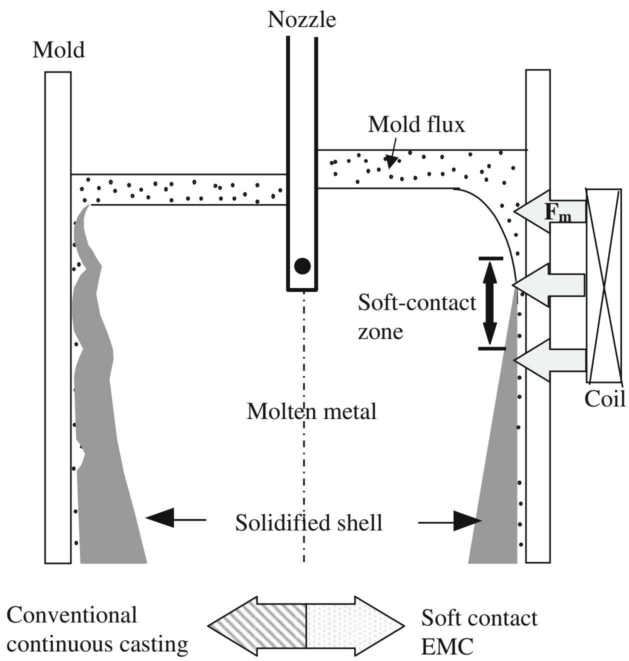

Fig. 2 Concept of soft contact EMC using the high-frequency magnetic field

electromagnetic stirring of the molten metal when the EMS device is used. Figure 3 shows the temperature change of the molten magnesium during casting. When EMC was used, the temperature between the center and periphery positions in the molten magnesium was 15 K. When EMS was additionally activated, the temperature difference was reduced to 10 K by forced convection.

Figure 4 compares the appearance of the 1 wt%Ca AZ31 billets processed with or without electromagnetic field. When the EMC was powered off, the billet cooled too

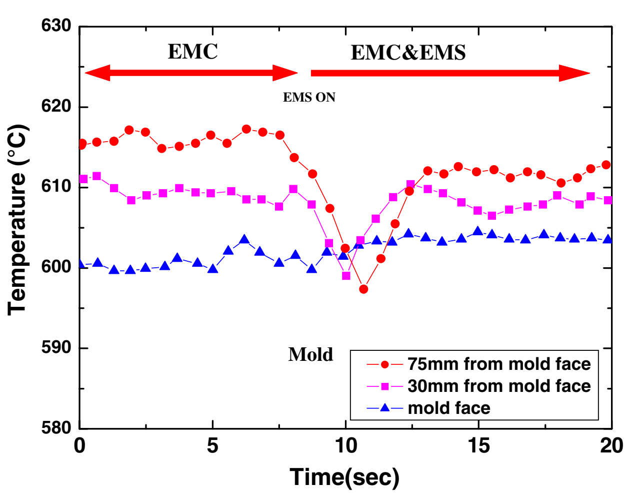

Fig. 3 Temperature distribution on the molten magnesium during EMC and EMC + EMS casting

fast and failed in producing high surface quality. When the power supply to EMC was turned on, the initial solidified shell cooled slowly in meniscus region such that oscillation marks could be totally suppressed. The homogeneity of the initial solidified shell was also greatly improved by EMC. Under the optimum condition, the casting speed could be as high as 0.4 m/min. When the billet size with 150 mm in diameter was used.

The uniform distribution of grain size within the cross section of a billet is an important factor for production of large-scale billets. The microstructures of the AZ31 casting billets processed with and without EMC are compared at different positions on the cross section plane in Fig. 5. In the casting process without EMC, dendritic structures developed and the overall microstructure was inhomogeneous. In contrast, when EMC was applied, the dendritic arms were fragmented to globular grains and microstructure was homogeneous over the entire cross-sectional area. The measured grain size differed depending on the position but was in narrow range between 153 and 170 μm (the average grain size = 163 μm). This improvement in microstructure has come from suppression of dendrite structure by reduction of temperature gradient in the molten metal by Joule heating during EMC.

The microstructures of the 1 wt%Ca-AZ31 billet processed by EMC and EMC + EMS are shown in Fig. 6. The Ca addition of 1.0 wt% resulted in significant grain refinement. In the case of EMC, the microstructure is composed of fine globular grains homogeneously distributed over the entire cross section of the billet. The average grain size is 56 μm, which is considerably smaller than that of the EMC AZ31 alloy without Ca addition (163 μm), indicating that there exists the influence of Ca on nucleation and/or growth rate of Mg grains. Application of EMC + EMS further refined the microstructure. There are the nugget zones characterized by agglomerates of small grains among the relatively coarse globular grains (marked as dotted circles), which is the result of grain refinement by enhanced fluid motion driven by EMS. Its population tends to increase as the distance from the center increases, such that the average grain size decreases towards the periphery. This microstructural gradient is resulted since electromagnetic force that drives the rotational flow in the molten pool increases as the distance from the center of a billet increases [8]. Higher cooling effect at the surface of the molten pool also contributes to formation of finer grained microstructure at the outer surface of the billet. The grain sizes anywhere within the cross section of the billet are less than 50 μm and the grain size is 45 μm. It is worthy to note that this grain size is significantly smaller than that observed in the 1 wt%Ca-AZ91 alloy (=107 μm) fabricated using conventional casting method [7].

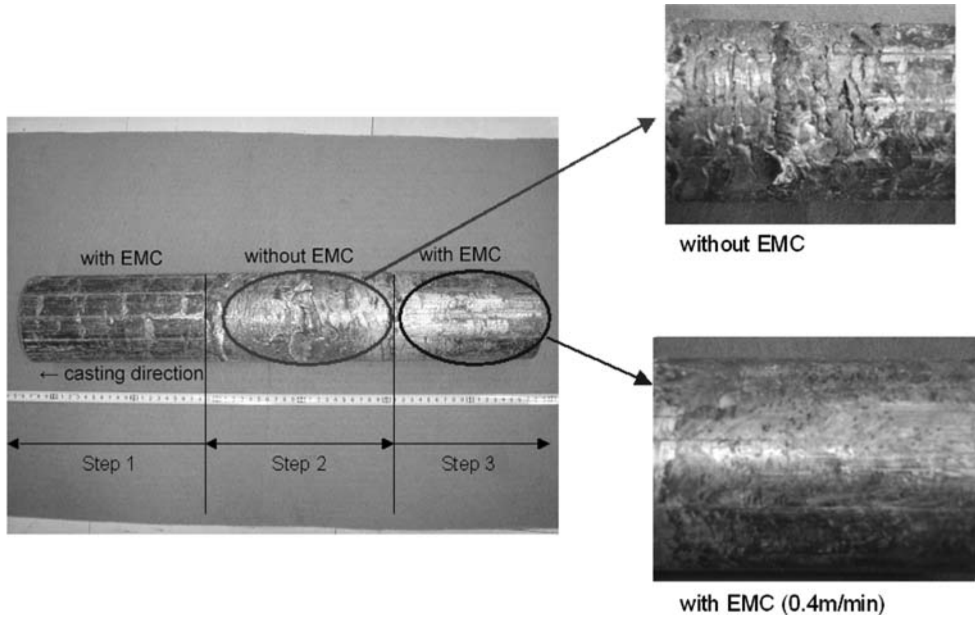

Fig. 4 Appearance of AZ31 billets processed with and without electromagnetic field

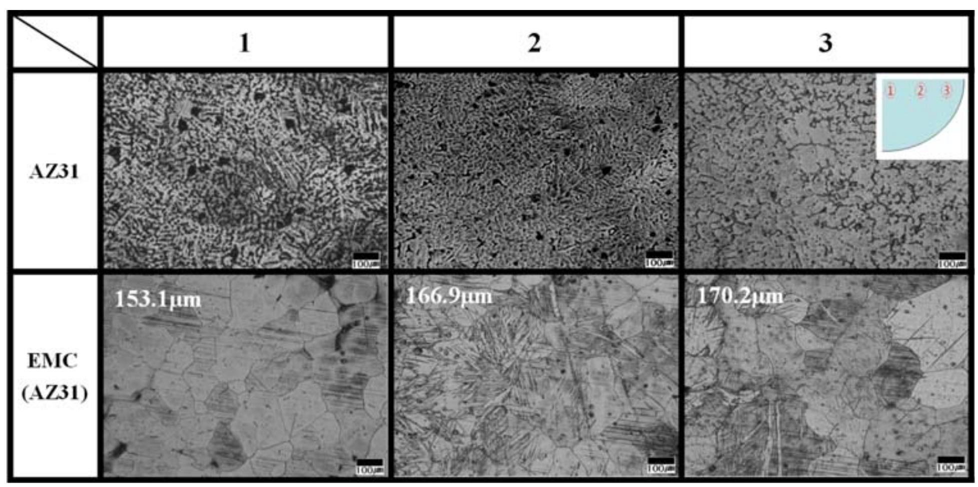

Fig. 5 Microstructures of the AZ31 casting billets processed with and without EMC

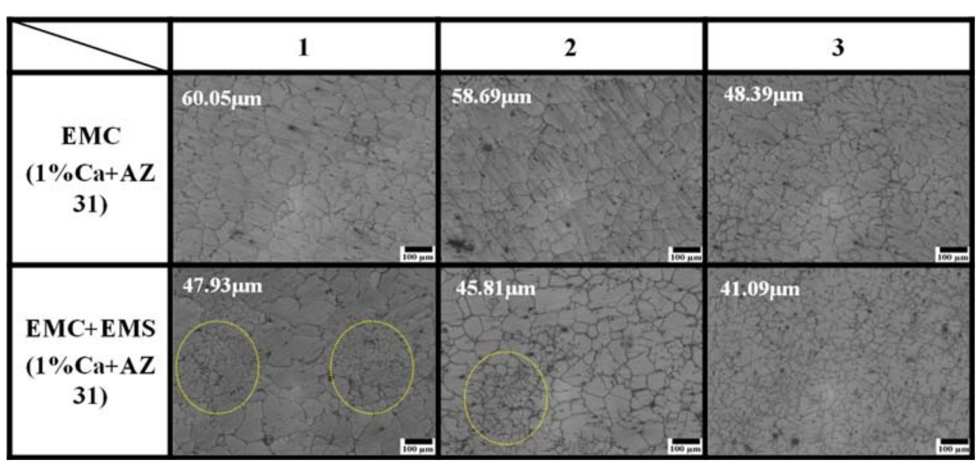

Fig. 6 Microstructures of the 1 wt%Ca-AZ31 billet processed by EMC and EMC + EMS

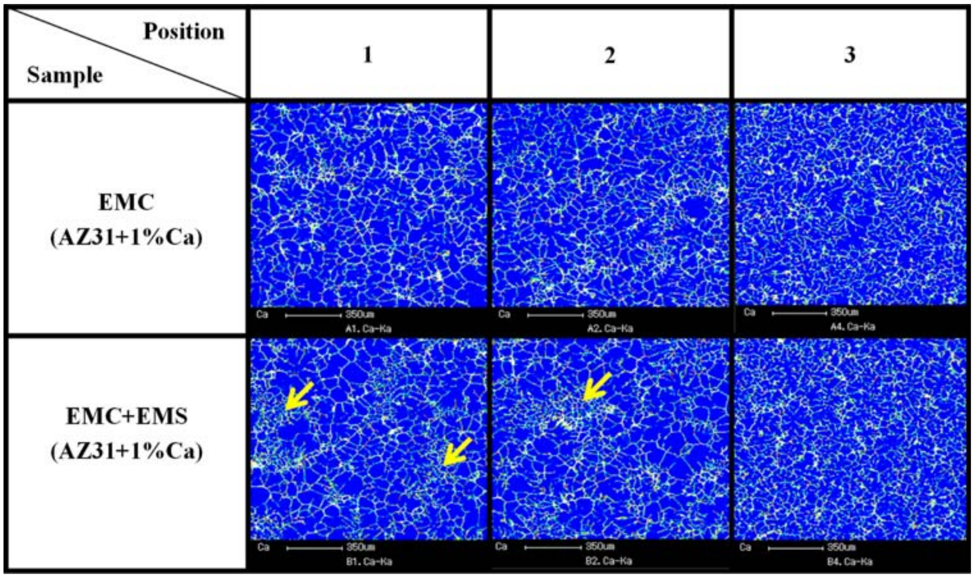

Fig. 7 The EPMA images showing that most areas of grain boundaries in the cross section of the billet processed by EMC and EMC + EMS are with high Ca content

The EPMA images in Fig. 7 show that grain boundaries in the cross section of the billet processed by EMC and EMC + EMS are with high Ca content. Presence of the nugget zones (indicated by arrows) is evident in the EPMA image of the EMC + EMS casting, indicating that most of grain boundaries in the cast are decorated with the Ca-containing compounds.

The eutectic compounds formed in Mg–Al–Ca alloys were reported to be Al_2Ca [9], Mg_2Ca [10], (Al, $Mg)_2Ca$ [11] or mixtures of these three phases [12] because of the similarity of their crystal structures. Recently, Han et al. [13] reported that three intermetallic phases exist in Mg–5%Al–Ca with Ca contents of 0.5 and 1.0 wt%; β-$Mg_{17}Al_{12}$, Al_8Mn_5, and (Al, $Mg)_2Ca$. In the present study, phase fractions (mol) of equilibrium phases in the AZ31 alloy without and with 1 wt%Ca were computed by a phase diagram calculation software (Pandat) as a function of temperature and the result is shown in Fig. 8. According to the analysis, there exist four major precipitation phases in the 1 wt%Ca-AZ31; (Al, $Mg)_2Ca$, β-$Mg_{17}Al_{12}$, Al_4Mn and Φ-Mg_{21}(Al, $Zn)_{17}$, while three phases exist in AZ31; β-$Mg_{17}Al_{12}$, Al_4Mn and Φ-Mg_{21}(Al, $Zn)_{17}$. With addition of Ca into AZ31, the phase fraction of β-$Mg_{17}Al_{12}$ markedly decreases, agreeing with the experimental observation by other investigator [11]. The factions of the other phases, on the other hand, remains almost unchanged. While the β-$Mg_{17}Al_{12}$ and Φ-Mg_{21}(Al, $Zn)_{17}$ phases completely dissolve at $\sim$450 K as in AZ31, the phase fraction of (Al, $Mg)_2Ca$ is almost invariant until it disappears at 823 K, indicating that (Al, $Mg)_2Ca$ is thermally very stable. Therefore, the (Al, $Mg)_2Ca$ phase is anticipated to play an important role in suppressing grain growth of α-Mg during solidification process, leading to formation of fine grains.

A SEM micrograph and the elemental maps for the EMC + EMS casting (Fig. 9) show that Al and Ca are preferentially located at the grain boundaries to form Al–Ca compounds. The Al–Ca compounds at the grain boundaries in the EMC + EMS billet are thinner than those in the EMC billet, especially in the nugget zones, due to increased grain boundary area by forming smaller grains. Zn element also preferentially exists at the grain boundaries. The Al–Mn compounds, either Al_4Mn or Al_8Mn_5

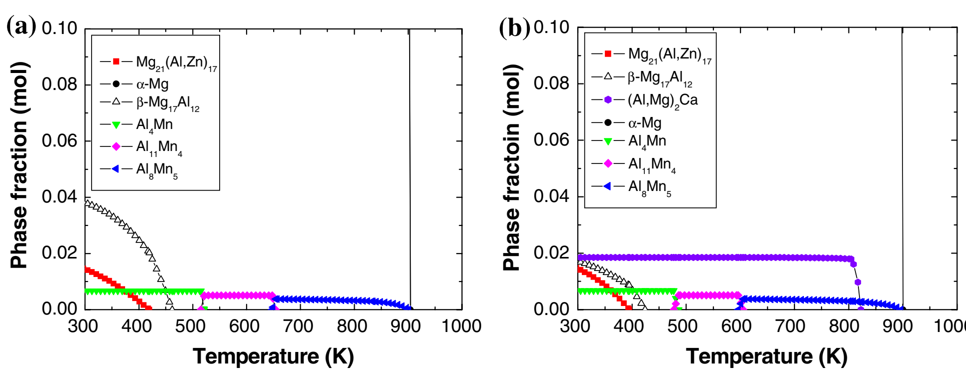

Fig. 8 Phase fractions of the equilibrium phases in the AZ31 alloy **a** without and **b** with 1 wt%Ca computed by Pandat software as a function of temperature

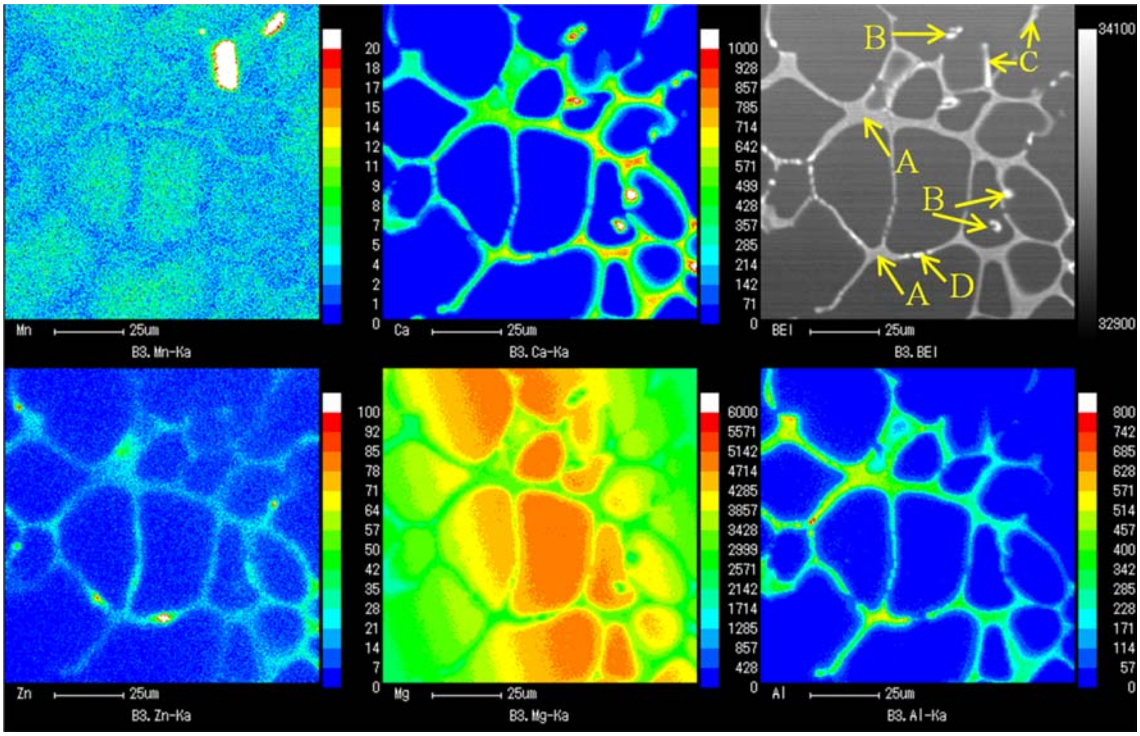

Fig. 9 The EPMA images of the EMC + EMS billet showing that high Ca, Al and Zn contents are with the grain boundaries: A[(Al, Mg)$_2$Ca], B[Mg$_2$Ca], C[Al$_4$Mn or Al$_8$Mn$_5$], D[Φ-Mg$_{21}$(Al, Zn)$_{17}$]

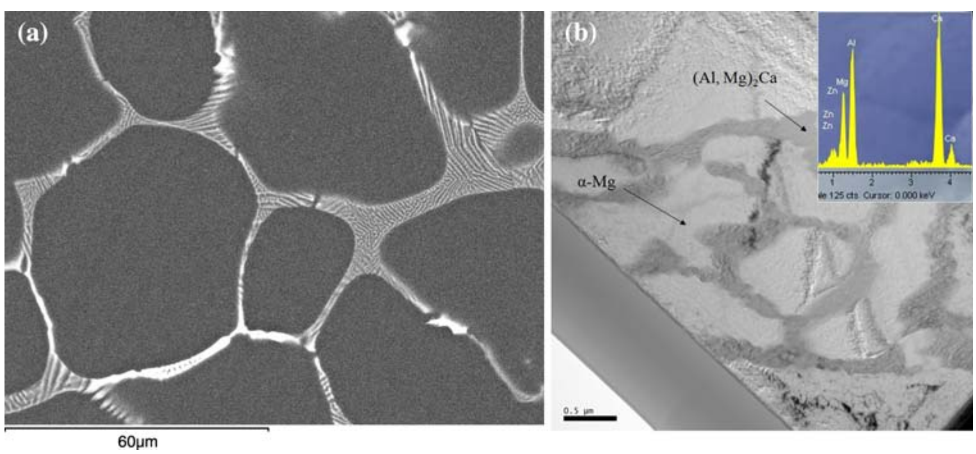

Fig. 10 **a** SEM and **b** TEM micrographs of the phases on grain boundaries in the microstructure of the billet produced by EMC + EMS

phase, exist in the form of particles. Mg–Ca compounds, most probably Mg$_2$Ca phase, in the form of particles are also found. It is hard to distinguish the β-Mg$_{17}$Al$_{12}$ phase from the EMPA image since it exists at grain boundaries in mixture with (Al, Mg)$_2$Ca phase. Figure 10 shows the SEM and TEM micrographs of the phases at grain boundaries in the microstructure of the billet produced by EMC + EMS, which show lamellae structures that are arranged in alternating layers of the Al–Ca phase and α-Mg phase. Qualitative analysis of the Al–Ca phases by energy dispersive X-ray analysis confirmed that the phase is (Al, Mg)$_2$Ca phase. As Zn is always detected in the phase at grain boundaries, however, its coexistence with Φ-Mg$_{21}$(Al, Zn)$_{17}$ phase is possible. The EDS results also showed that there were very small amounts of Al and Ca in the grain interior regions.

The mechanical properties of the AZ31 billet processed by EMC are compared with the same composition billet casted by the conventional casting in Fig. 11. The yield strength and ductility of the EMC AZ31 billet are higher by

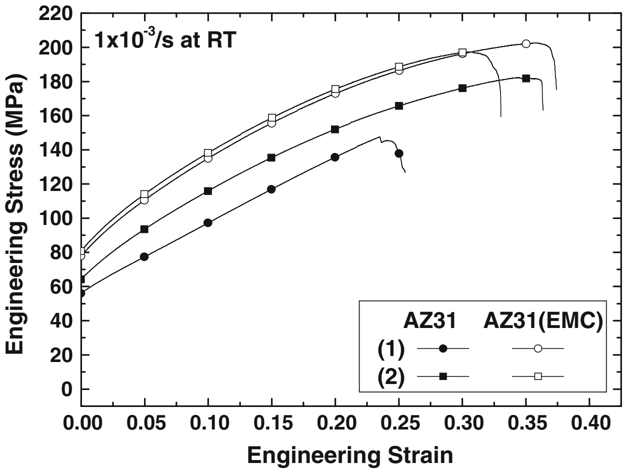

Fig. 11 Engineering stress–engineering strain curves of the AZ31 billet casted by EMC compared with the same composition billet casted by the conventional ingot casting

about 33 and 12%, respectively. Furthermore, the strength and ductility at the center and periphery regions in the EMC billet are similar, agreeing with the observation that microstructure is homogeneous over the entire cross section of the EMC AZ31 billet (Fig. 5).

Addition of Ca into AZ31 improved the yield strength of the EMC billet from 80 to 100 MPa but largely reduced the tensile elongation from 35 to 11% (Fig. 12a). The mechanical properties were homogeneous. The improvement of strength should be primarily attributed to the reduction in grain size by Ca addition. Presence of (Al, Mg)$_2$Ca phase at the grain boundaries may also contribute to the strengthening by blocking movement of dislocations. The ductility reduction by the Ca addition, on the other hand, is linked with the presence of (Al, Mg)$_2$Ca phase at the grain boundaries of α-Mg in network form, which easily debond from the soft magnesium matrix during deformation [14]. Application of EMS improved the strength of the 1%Ca-AZ31 billet further (Fig. 12b), especially at the periphery region (114 MPa) where the measured grain size is smallest. Improvement of tensile ductility is also seen, which can be related to formation of finer matrix grains and thinner (Al, Mg)$_2$Ca phase that tends to be more easily breakable to finer particles during plastic deformation.

Conclusions

The effects of EMC and EMS on the microstructure and mechanical properties of the AZ31 and 1 wt%Ca-AZ31 billets were examined. The obtained results illustrate a great potential that high-quality magnesium billets can be fabricated using electromagnetic continuous casting method.

1. Electromagnetic field produced the high-quality surface in the AZ31 and 1 wt%Ca-AZ31 billets. Oscillation marks were totally suppressed and homogeneity of the initial solidified shell was greatly improved. Furthermore, the casting speed could be increased up to 0.4 m/min when the billet with a diameter of 150 mm was used.
2. Application of EMC to the AZ31 resulted in a high degree of micrstructural homogeneity in the billet. The yield strength and elongation of the EMC AZ31 billet were increased by about 33 and 12%, respectively, compared with the AZ31 billet produced without EMC.
3. The Ca addition (1.0 wt%) to AZ31 with EMC application resulted in significant grain refinement. The average grain size of the fine globular grains was 56 μm. Strength improvement was obtained by grain refinement but presence of coarse (Al, Mg)$_2$Ca phase at grain boundaries reduced tensile ductility.
4. Application of EMC + EMS to the 1 wt%Ca-AZ31 refined the microstructure to the average grain size of 45 μm. Attainment of the fine-grained microstructure was attributed to reduction of temperature gradient and fragmentation of dendrite structure by enhanced fluid motion.

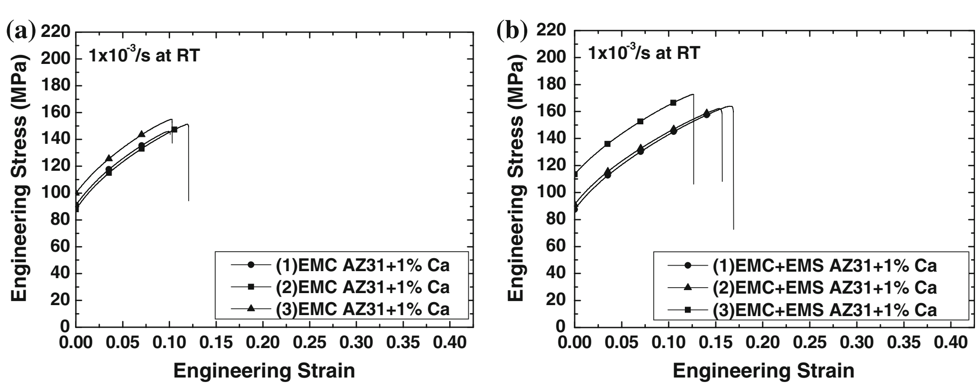

Fig. 12 Engineering stress–engineering strain curves of the **a** EMC and **b** EMC + EMS 1 wt%Ca-AZ31 alloys

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