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# The Development of Spheroidal Grains and Thixoforming of AZ91D Magnesium Alloy Treated by Different Routes

**Gaozhan ZHAO [1] *, Yongjin LU [2, 3], Changjian SUN [1], Fuyou FAN [4], Chengzhou LONG [4]**

[1] *Southwest Technique and Engineering Institute, No.33 Yuzhou Road, Chongqing 400039, China*
[2] *China Ship Development and Design Center, No.268 Zhangzhidong Road, Wuhan 430064, China*
[3] *Key Laboratory for Advanced Materials Processing Technology, Ministry of Education, Tsinghua University, Haidian district, Beijing 100084, China*
[4] *Research Development Center of Changsha Electromechanical Products, Xingsha Road, Changsha 410100, China*

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A new process, squeeze casting-solid extrusion (SCSE), was introduced to prepare semi-solid billets for thixoforming together with partial remelting. The microstructure development and tensile properties of AZ91D alloy reheated from states of as-cast and SCSE-formed were studied. During partial remelting, SCSE-formed samples obtained finer and more uniform microstructure obviously compared with as-cast ones. As the holding time went on, both solid particles from the two states experienced continuous spheroidization. In the meantime, from SCSE-formed were always to coarsen, while from as-cast were refined initially and coarsened then. Microstructure development is determined by the combination effects of various factors, including distortion energy from SCSE deformation, grain coalescence, Ostwald ripening mechanism, etc. During thixoforming, components with good forming quality were prepared successfully. Excellent tensile properties were obtained for the thixoformed alloy prepared by SCSE deformation, mainly with the microstructure refinement and the decrease of defects related.

*Keywords*: AZ91D alloy, squeeze casting-solid extrusion, spheroidal grains, aggregation, tensile properties.

## 1. INTRODUCTION

As a new alloy processing method between forging and traditional diecasting, semi-solid forming has been considered to be a efficient and near-net forming process in the field of machine, mechanical, optical and electrical units manufacturing. Compared to traditional casting, semi-solid processing have several potential advantages, such as less segregation and lower forging force [1 – 3]. For this reason, the better tensile properties were expected.

To ensure the feasibility of thixoforming, as the reheated billet entering into the state of semi-solid, grains should be globular and homogeneous instead of usual dendritic [4]. In order to realize this, a number of methods have been studied, such as magneto hydrodynamic stirring, cooling slope, recrystallization and partial melting (RAP), strain-induced melt activation (SIMA) and near-liquidus casting [5 – 6]. During the RAP route, the target material is reheated into the semi-solid state from below the recrystallisation temperature originally [7].

The microstructural development and semi-solid processing of magnesium alloys have been widely studied. Lin et al. found that α-Mg in semi-solid AZ91D billet was obviously refined and mechanical properties were significantly improved after SIMA route. They concluded that main reasons were recrystallisation and refinement strengthening [8]. Yan et al. studied the development of semi-solid grains of AZ61 billet during reheating as the result of the SIMA route, and considered that coalescence mechanism results in coarsening initially [9]. Kleiner et al reported the semi-solid grains evolution of Mg-Al-Zn alloy during partial remelting after extrusion deformed. They suggested that the presence of small intragranular liquid droplets could be explained by the self-blocking remelting mechanism [10 – 11].

In this paper, a new process squeeze casting-solid extrusion (SCSE) was introduced to prepare semi-solid AZ91D billets. An aim has been focused on investigating the microstructure development of as-cast and SCSE-formed material during partial remelting. Moreover, the tensile properties of the material under the two states were also discussed.

## 2. EXPERIMENTAL PROCEDURE

In this experiment, the composition of the commercial AZ91D magnesium alloy used as the matrix alloy was Mg-9.09Al-0.86Zn-0.18Mn (wt.%). Firstly, based on the RAP route, SCSE is proposed for preparing semi-solid feedstock together with partial remelting. The preparation steps were summarized as follows: [10].

1) Preheat the mold cavity of the hydropress and pour proper molten alloy into it;

2) The mold cavity is closed by pressure and the liquid alloy is pressurized quickly for rendering solidification till the end;

3) When the temperature of the solidificated alloy decreases to that below the recrystallization temperature, solid extrusion is operated;

4) For partial remelting, divide the SCSE-formed billets into several slugs according to the shape of components. Heat slugs to the range of semi-solid temperature for a period of time, then fine and homogeneous grains can be obtained.

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*Corresponding author. Tel.: +86-02368792232; fax.: +86-02368792100.
E-mail address: *chfzgz@126.com* (G. Zhao)

The matrix alloy was melted at 760 ºC under the protection of mixed gas of $SF_6$ and $CO_2$. The molten metal was cooled down to 720 ºC and kept for 10 min after being stirred for about 20 min at 760 ºC. The preheated temperature of the die was 350 ºC. Then poured the liquid alloy into the mold cavity of Ø 300 mm, and activated the punch to exert 300 MPa pressure on the alloy immediately and held for 600 s. The alloy cooled to 300 ºC after absolutely solidification, and extruded it to bar-shaped samples. During this operation, the extrusion rate and extrusion ratio were 3 mm/s and 6, respectively.

Investigation of microstructure developments during reheating and thixoextrusion of semi-solid billets was conducted both on as-cast and SCSE-formed AZ91D samples machined to the size of Ø 8 mm×12 mm. Samples were heated isothermally at 540 ºC (i. e. the semi-solid temperature) for different from 0 min to 30 min in the electric furnace under the argon protection environment, and quenched quickly into cold water.

As-cast and SCSE-formed slugs of Ø 120 mm×70 mm were both machined before thixoforming. These slugs were heated quickly with the aid of electromagnetic induction, and isothermally held for 20 min when the temperature reached 540 ºC, then started thixoforming into a die which had been preheated to 350 ºC. Thermocouples were embedded into slugs to ensure the accuracy of temperature measurement. During thixoforming, the working speed of the punch, the exerted pressure and the pressure holding time were 100 mm/s, 60 MPa and 60 s respectively.

After reheating, samples from SCSE and as-cast were processed into standard block and etched, and the microstructure was analyzed using various means such as optical microscope. Grain size and shape factor were tested and calculated using image analysis system. After components thixoforming, thin sheet-shaped samples were cut from them and used for tensile testing.

## 3. RESULTS AND DISCUSSION

### 3.1. As-cast and SCSE-formed microstructures

As-cast and SCSE-formed AZ91D microstructures are shown in Fig. 1. Fig. 1, a, shows the as-cast microstructure, which mainly included α-Mg matrix and β-$Mg_{17}Al_{12}$ intermediate phase. The intermediate phase with continuous or semi-continuous net-like structure was quite thick, most of which distributed nearby crystal boundaries. Due to applied pressure for solidification of liquid alloy , the microstructure is obviously refined (Fig. 1, b). According to the Clausius-Clapeyron equation [12]:

$$\frac{dP}{dT} = \frac{T_m \Delta V}{H_f} \tag{1}$$

the application of external pressures ($P$) can result in increase of the melting point of the alloy, where $T_m$ is the equilibrium solidification temperature, $\Delta V$ is the volume difference during solidification, and $H_f$ is the latent heat of fusion. During solidification, $\Delta V$ and $H_f$ are both negative because of the shrinkage of alloy and heat release, respectively. Therefore, $dT/dP$ is normally positive, that is, an increase in applied pressure can lead to a rise in solidification temperatures. The higher freezing point, the greater cooling rate in the initially superheated alloy, and finer microstructure can be obtained finally.

Further more, obvious directional band grains appeared along the deformation direction after SCSE-formed (Fig. 1, c). The crystal boundaries became pretty muddied and the intermediate phase was refined into small and diffuse particles. In addition, the RAP route can be classified to be suitable for the SCSE-formed microstructure in Fig. 1, c, attributed to the typical unrecrystallized grains.

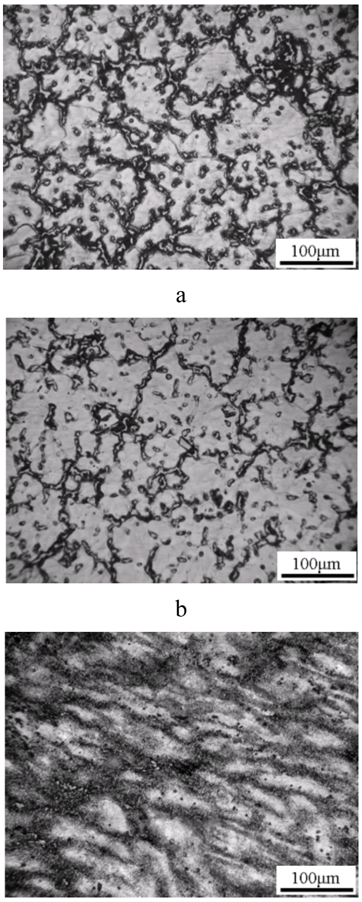

**Fig. 1.** Microstructures of as-cast and SCSE-formed AZ91D alloy: a – as-cast; b – squeeze casting formed; c – squeeze casting-solid extrusion formed

### 3.2. Microstructures development during remelting treated by different routes

Figs. 2 and 3 show the microstructure development of as-cast and SCSE-formed AZ91D alloy isothermal held at 540 ºC for different time (0, 10, 20 and 30 min). Meanwhile, the change curves of both mean grain size and shape factor after isothermally holding are showed in Fig. 4.

Fig. 2, a, shows that the microstructure became coarse and nonuniform obviously, when heated up to 540 ºC from as-cast. The net-like $Mg_{17}Al_{12}$ decreased significantly and a small amount of liquid formed in local areas after the temperature reached 540 ºC. Prolonging isothermal holding time, finer solid particles began to spring up and the net-like crystal boundary areas was taken over by liquid film completely (Fig. 2, b – d), due to thickening of liquid films. Also it was shown that solid particles had been spheroidized greatly over time, but the tendency of grains evolution were refined at the beginning and then coarsened.

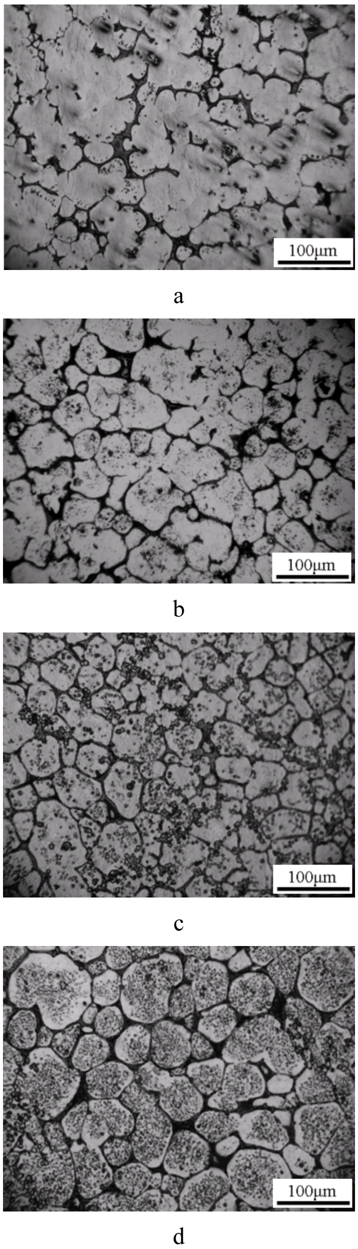

**Fig. 2.** Microstructures of as-cast AZ91D alloy isothermal held at 540 ºC for different times: a – 0 min; b – 10 min; c – 20 min; d – 30 min

As it is shown in Fig. 3, a, partial remelting occurred in the vast bulk of areas of microstructures from SCSE-formed state without isothermal holding at 540 ºC, even with existence of some incipient spheroidal grains and pools of liquid. Then the liquid film becomes thicker little by little in the original crystal boundary areas. With the continued isothermal holding, partial remelted areas spreaded to the whole microstructure. Fig. 3, c, shows the microstructure after 20 min heating, it can be observed that the relatively fine and uniform spheroidal grains had been obtained, but continued to be remelted, the grains of samples coarsened seriously further. In the mean time, both the mean grain size and degree of spheroidization increased continuously during reheating. Moreover, further examination of the microstructures revealed that the spheroidization rate slowed down after isothermal held for 20 min. In general, through comparing Fig. 2 to Fig. 3 and analyzing Fig. 4, samples remelted from SCSE-formed obtained finer and more uniform microstructure obviously than from as-cast in almost every stage of holding time.

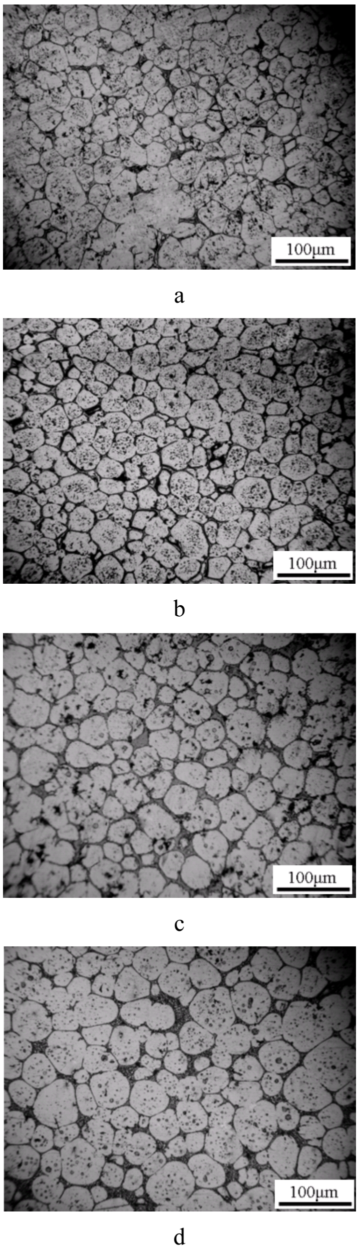

**Fig. 3.** Microstructures of SCSE-formed AZ91D alloy isothermal held at 540 ºC for different times: a – 0 min; b – 10 min; c – 20 min ; d – 30 min

In addition, Fig. 5 shows the grain growth plot for as-cast and SCSE-formed AZ91D alloys during partial remelting at 540 ºC. The relationship between size of the growing solid grain and isothermal holding time can be described according to the classic LSW theory:

$$d^3 - d_0^3 = K\,t\,, \tag{2}$$

where $d_0$ is the mean grain size initially, $d$ is the size at holding time $t$, and $K$ is a constant of coarsening rate [11]. From the present data, the coarsening rate $K$ of as-cast is found to be 293 $\mu m^3/s$, which is slightly higher than that obtained from SCSE-formed (i. e., 272 $\mu m^3/s$).

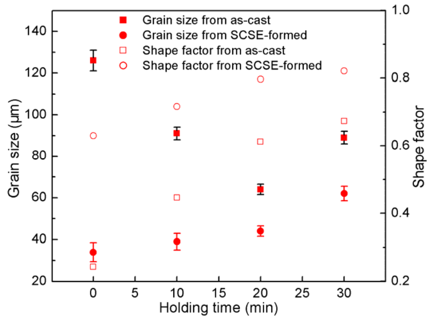

**Fig. 4.** Mean grain size and shape factor of as-cast and SCSE-formed AZ91D alloy after isothermal holding at 540 °C for different times

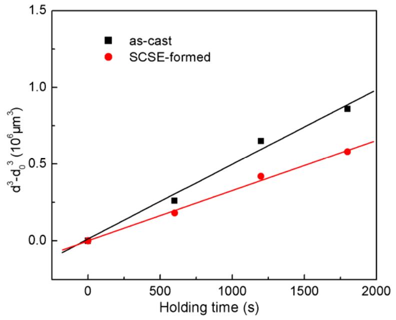

**Fig. 5.** Grain growth plot for as-cast and SCSE-formed AZ91D alloys during partially remelting at 540 °C

### 3.3. Mechanisms of microstructure development during remelting

During isothermal holding, the SCSE deformation can lead to finer and more homogeneous spheroidal grains than as-cast. Furthermore, it also causes excessive growth of grains. The reason can be analyzed as follows. As SCSE deformation went on, more and more distortion energy was stored in the alloy in the form of vacancies, dislocation multiplication and lattice distortion. When the alloy was heated below solidus temperature, recovery and recrystallisation would be activated by drawing the energy above. As a result, nucleation and growth of grains were accelerated. The more distortion energy was stored, the finer grains would be obtained after recrystallization. During SCSE deformation, distortion energy was stored due to solidification under applied pressure and extrusion with a relatively high extrusion ratio. By contrast, distortion energy of as-cast was virtually nil. However, the increment of nucleation rate is always slower compared with growth rate, and two reasons are proposed. Firstly, with the recrystallisation going on, the distortion energy stored from SCSE deformation gradually decreases. With the consumption of limited distortion energy, recrystallisation can not be maintained [13]. Secondly, aggregation among recrystallized grains will start first from grain boundaries where energy is relatively lower [14]. By this law, thicker and more irregular solid particles are formed, which can be easily identified from Figs. 2 and 3. Therefore, once the distortion energy for drawing is run out, determinant of grains evolution will transform into aggregation.

The formation process of coalescence from two adjacent spheroidal grains to a irregular one can be divided into two cases: sizes of the two have significant difference or little to no. In the former case, after the small merged into the large, energy of solid-liquid interface decrease slowly during the isothermal holding, and the newly formed grain will be spheroidized gradually. But in the latter case, the relatively low energy is not enough to affect the irregular shape dramatically.

Besides, as is shown in Fig. 4, the shape factor of AZ91D alloy were improved gradually both from as-cast and SCSE-formed during isothermal holding at 540 °C. During partial remelting, the recrystallisation occurred firstly.With the rapid growth of new grains, deformed regions are replaced by strain-free ones, so the energy stored within grains decreases dramatically until close to zero. Meanwhile, as the holding time goes on, liquid film starts to surround the recrystallized grains little by little and thicken continuously, which separate the adjacent grains and make them hard to gather again. Consequently the grains are spheroidized gradually.

Solidifying thermodynamics provides the relationship between the interfacial energy and the decrease in equilibrium melting temperature $\Delta T_r$ as follows:

$$\Delta T_r = -\frac{2T_m V_s \sigma}{\Delta H_m} K \,, \tag{3}$$

where $\Delta H_m = H_s - H_l$ is the difference between enthalpy of the liquid and solid phase, $T_m$ is the transformation temperature in equilibrium, $V_s$ is the volume of solid phase, $\sigma$ is the interfacial tension and $K$ is the mean curvature of the solid surface [15]. Therefore, as the billet temperature reaches the semi-solid range, compared with the bulge parts of recrystallised grain the concave are melt more easily to make the grain become rounded gradually. Also, Ostwald ripening mechanism indicates that the minor solid particles and closed angles of the large have lower melting points and melt more easily, because of the greater radiuses of curvature [16 – 17]. In addition, with the interfacial energy reducing, grain coarsens gradually and the liquid film becomes thinner [18]. When the film is too thin to separate the two adjacent grains which are comparable in size, a larger size, irregular-shaped new grain will be formed as shown in Figs. 2 and 3.

### 3.4 Thixoforming and its effect on tensile properties of components

Fig. 6 presents AZ91D magnesium alloy slugs cut from SCSE-formed billets and successfully thixoformed barrel-shaped components using this kind of slugs. The starting alloy was treated by the RAP route (SCSE deformation plus partial remelting). As is shown in Fig. 6, the surface forming quality and thin-wall filling effect are both good.

Excellent tensile properties were obtained for the thixoformed alloy that was prepared by SCSE, with a yield strength of 207 MPa, a ultimate tensile strength of 284 MPa and an elongation to fracture of 8.3 %. Compared with as-cast formed, i. e. 171 MPa, 238 MPa and 6.2 %, all the three were improved significantly, which increased by about 21.1 %, 19.3 %, and 33.9 % respectively.

SCSE deformation and partial remelting can bring finer grains to the thixoformed components, and that is what the yield strength of the alloy depends on, such that it increases with the decreasing of the grain size. But the ultimate tensile strength and the elongation are mainly correlated with the amount of defects in the alloy, e. g. pin holes and shrinkage porosity [19]. When billets are predeformed by SCSE, defects will be reduced dramatically, which leads to higher strength and elongation compared to as-cast state.

a b

**Fig. 6.** AZ91D slugs cut from SCSE-formed billets and successfully thixoformed components using this kind of slugs: a – AZ91D slugs; b – thixoformed components

## 4. CONCLUSIONS

Squeeze casting-solid extrusion was introduced to prepare semi-solid billets of AZ91D magnesium alloy for thixoforming together with partial remelting. This new process can break the net-like β-$Mg_{17}Al_{12}$ of as-cast AZ91D microstructure into small and diffuse particles and provide sufficient energy for recrystallization.

Samples remelted from SCSE-formed state obtained finer and more uniform microstructure obviously than from as-cast. As the holding time went on, both the solid particles from the two states experienced continuous spheroidization, and in the meantime from SCSE-formed were always to coarsen, while from as-cast were refined initially and coarsened then. Microstructure development during remelting is determined by the combination effects of various factors, including distortion energy from SCSE deformation, grain coalescence, Ostwald ripening mechanism, etc.

During thixoforming, components with good forming quality were prepared successfully. Excellent tensile properties were obtained for the thixoformed alloy prepared by SCSE deformation, mainly with the microstructure refinement and the decrease of defects related.

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