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Microstructure evolution and mechanical properties of Al_2O_{3sf}/AZ91D magnesium matrix composites fabricated by squeeze casting

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Abstract The squeeze casting process was used to fabricate Al_2O_{3sf}/AZ91D magnesium matrix composites before thixoforging. The microstructural evolution process in Al_2O_{3sf}/AZ91D was investigated during partial remelting. Tensile mechanical properties of thixoforged automotive component were determined and compared with those of squeeze casting formed composites. The results show that the microstructural evolution during partial remelting exhibited four stages: the formation of liquid, structural fragmentation, the spheroidization of solid particles, and final coarsening. As the holding time increases, the size of solid particles decreases initially and then increases. However, the size of solid particles decreases monotonously as the temperature increases. Increasing holding time or temperature promotes the degree of spheroidization. It is also shown that the cylindrical feedstock of the Al_2O_{3sf}/AZ91D composites can be thixoforged in one step into intricate shapes in the semi-solid state. The tensile tests indicate that the yield strength and ultimate tensile strength for Al_2O_{3sf}/AZ91D thixoforged from starting material fabricated by squeeze casting and partial remelting are better than those of Al_2O_{3sf}/AZ91D fabricated by squeeze casting. This research confirms that thixoforging is a practical method for the near net shape forming of magnesium matrix composites.

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Introduction

Magnesium alloys have received much attention as important materials in aerospace, automobile, and construction industries. However, their applications are often restricted because of their inherent deficiencies, such as poor high temperature strength, low stiffness, low creep resistance, and high thermal expansion coefficient [1]. In the last decades, magnesium matrix composites reinforced with discontinuous reinforcements have been developed to overcome these drawbacks of magnesium alloys, achieving low density composites with high specific strength, high specific stiffness, high wear resistance, high creep resistance, and low thermal expansion coefficient [2]. Although the use of discontinuous reinforcement leads to an improvement in the strength characteristics of magnesium alloys, the intrinsic limited ductility worsens due to the high brittleness exhibited by non-reacting ceramic-Mg formulations. Furthermore, because of their limited ductility, magnesium matrix composites have some difficulties in forming and machining.

One way to widen the application of high cost-effective magnesium matrix composites is to employ semi-solid processing. Semi-solid processing involves the rheo-route and the thixo-route [3–5]. Thixoforging, which refers to the thixo-route, is a relatively new route for the forming of metals and metal matrix composites (MMCs). In this process, metals or MMCs are shaped in the semi-solid state, exploiting the thixotropic behavior of feedstocks consisting of solid spheroids in a liquid matrix, i.e., when the material is sheared it flows but when it is allowed to stand it thickens again; the viscosity is time and shear rate dependent [6]. Thixoforging is basically a three step process, including the preparation of a feedstock material with thixotropic characteristics, rehearing the feedstock material

to semi-solid temperatures and forming the semi-solid feedstock to components.

Ward et al. [7] studied the semi-solid processing of novel MMCs based on hypereutectic aluminum–silicon alloys. They found that thixoformed Al–Si MMCs with high silicon contents processed all the properties for which the alloy system was renowned: high stiffness, wear resistance, fatigue life, and hot strength, combined with low density and thermal expansion. Zhang and Wang [8] reported that the thixoformed SiC_p/6066Al composites produced by spray forming had fine microstructure and high performance similar to the conventional forged ones. The thixoformability of spray deposits was excellent and the forming temperature was the most important factor in determining the formability. Chen et al. [9] determined the microstructural evolution of predeformed SiC_p/ZA27 composites during partial remelting. They found that high compression and remelting temperature were beneficial to achieve a desirable structure for semi-solid processing. Furthermore, the effect of the SiC_p content and size on the semi-solid structure was less than those of the amount of compression and melting temperature, but the large size and optimum content SiC_p were beneficial to getting an ideal structure needed by semi-solid processing. Qin et al. [10] determined the effect of holding temperature on the semi-solid microstructure of Mg_2Si/Al composites. They found at the low heat treatment temperature, the morphologies of primary Mg_2Si particles were identified as irregular shape and α-Al grains exhibited elliptic or rosette type. Furthermore, with the increasing of treatment temperature, the morphologies of primary Mg_2Si and α-Al grains changed to spherical type, and their sizes decreased and became relatively uniform. The results of literature search indicate that only limited work has been reported on the microstructure evolution of MMCs in the semi-solid state [7–10]. Furthermore, to the best of our knowledge, no attempt is made so far to research on the thixoforging of Al_2O_{3sf}/AZ91D fabricated by squeeze casting.

Therefore, the primary aim of the present research was to fabricate Al_2O_{3sf}/AZ91D by squeeze casting, reheat Al_2O_{3sf}/AZ91D in the semi-solid state, and thixoform Al_2O_{3sf}/AZ91D. Furthermore, the aim is also to grain a better understanding of thixoforging for magnesium matrix composites and generate a database of mechanical properties.

Experimental procedures

The matrix alloy used was a commercial AZ91D magnesium alloy with a nominal composition of Mg–9Al–0.7Zn–0.15Mn (wt%) and the reinforcement was Al_2O_3 short fiber with a diameter of 5–8 μm and a length of 20–40 μm. The Al_2O_3 short fibers were fabricated into a preform by wet forming with a binder. The binder was composed of 5 g polyvinyl alcohol (PVA), 5 mL glycerine, 10 g $Al(H_2PO_4)_3$, and 200 mL H_2O. The preform with a diameter of 61 mm and a height of 80 mm was dried in the air at room temperature for 4 days. Then the preform was heated to 200 °C for 2 h to eliminate PVA. To improve the strength of the preform, the preform was calcined at 800 °C for 3 h in the air. Prior to infiltration, the preform was preheated in a furnace at 650 °C for 3 h. The preheating temperature of the die was about 450 °C. The AZ91D magnesium alloy was melted in an electric furnace with a graphite crucible under CO_2/SF_6 atmosphere. The casting temperature of the melt was selected as 760 °C. During infiltration, the pressure exerted by the punch on the molten metal was gradually increased to a pre-determined level and kept at this level for 2 min until the molten metal was completely solidified. To avoid the deformation of the preform, the applied pressures for infiltration were selected as 60 MPa. Colloidal graphite was used as a lubricant. The volume fraction of the short fibers in the composites was about 10%.

For investigation of microstructure evolution during partial remelting, the Al_2O_{3sf}/AZ91D magnesium matrix composites were machined into 8 mm diameter × 12 mm high cylindrical samples. The samples were heat-treated individually in the center of a furnace that had been carefully characterized for temperature distribution. A thermocouple, with an accuracy of 1 °C, placed in a hole of 2 mm diameter and 4 mm depth in the center of the sample ensured accurate temperature measurement and feedback control. Samples were heat-treated isothermally at various temperatures between the solidus and liquidus of the material for times in the range 8–40 min. On removal from the furnace, samples were immediately quenched in cold water.

For the thixoforging, the Al_2O_{3sf}/AZ91D magnesium matrix composites were machined into an ingot with a diameter of 60 mm and a height of 45 mm. Before thixoforging, the ingot was reheated in the center of a furnace that had been carefully characterized for temperature distribution. The furnace temperature was controlled by a K-type thermocouple embedded in the ingot. The ingot was then held at 550 ± 2 °C for 16 min to allow spheroidization of solid grains. The thixoforging operation was carried out with a laboratory hydraulic press. The die was preheated to 350 °C to avoid rapid cooling of the ingot during thixoforging. After the required holding time was reached, the thermocouple was removed. Then the reheated ingot was handled and put into the die. The pressure exerted by the punch on the ingot was rapidly increased to a pre-determined level of 350 MPa and kept at this level for 60 s.

Preparation of metallographic samples consisted of grinding with SiC paper of different fineness and subsequently polishing with 1 μm diamond paste. Scanning electron microscopy (SEM) was also used to examine the microstructure of Al_2O_{3sf}/AZ91D. The tensile tests were performed in an Instron 5569 machine with a crosshead speed 0.5 mm/min. The dog bone-shaped tensile ingots of the squeeze casting formed and thixoforged conditions had a gauge length of 15 mm and a thickness of 2 mm. At least three samples were tested in each condition. Mean grain size and shape factor of solid particles were calculated in each case by using the Eqs. 1 and 2,

$$d = \frac{\sum_{N=1}^{N} \sqrt{4A/\pi}}{N} \quad (1)$$

$$F = \frac{\sum_{N=1}^{N} \frac{4\pi A}{P^2}}{N} \quad (2)$$

where A and P are area and perimeter of grains, respectively, and N is the number of grains. For each sample, measurements were taken from the whole sectioned area with 200–250 grains per sample.

Results and discussion

Microstructure of squeeze casting formed magnesium matrix composites

Figure 1 shows the optical microstructure of squeeze casting formed Al_2O_{3sf}/AZ91D magnesium matrix composites fabricated under an applied pressure of 60 MPa. The short fiber Al_2O_3 reinforced phase was embedded in a magnesium matrix and there was no macroporosity in the ingot. Further SEM examination suggested that the as-cast

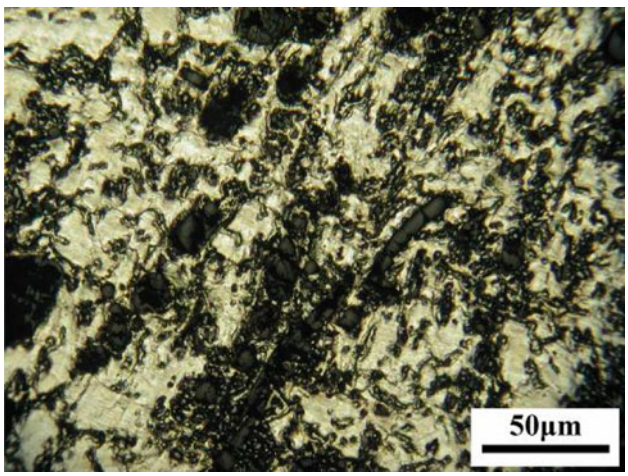

Fig. 1 Optical microstructure of squeeze casting formed Al_2O_{3sf}/AZ91D fabricated under an applied pressure of 60 MPa

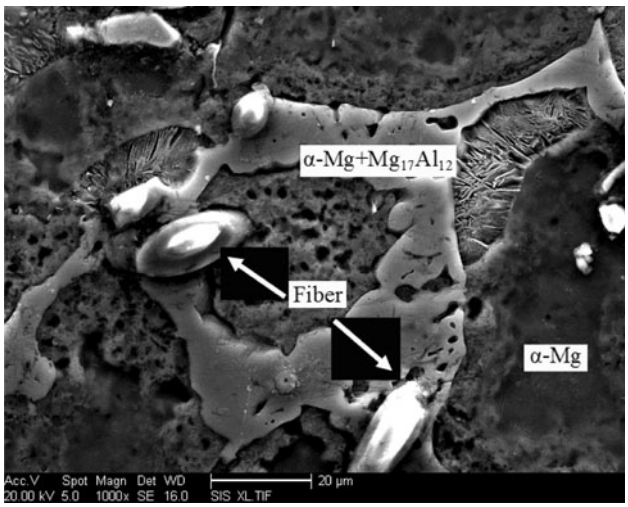

Fig. 2 Microstructure of squeeze casting formed Al_2O_{3sf}/AZ91D with 10% Al_2O_{3sf} with an applied pressure of 60 MPa

microstructure consisted of well-defined primary α phase (black), interdendritic $\alpha + \beta$ eutectics (gray), and homogeneous Al_2O_3 short fibers (white) (Fig. 2).

Structural evolution with increasing holding time during partial remelting

Figure 3 shows the microstructure evolution of the Al_2O_{3sf}/AZ91D during partial remelting at 550 °C for times in the range of 8–40 min. It can be seen from Fig. 3 that this process can be mainly divided into four stages: the formation of liquid, structural fragmentation, the spheroidization of solid particles, and final coarsening. During the initial stage, the dominant mechanism was the dissolution of the interdendritic eutectic phases with low melting temperatures, which formed as a result of the segregation during squeeze casting. With increasing holding time, the amount of liquid increased and the dendrite network inside each grain broke down. Under this condition, the dendrites were separated into individual solid particles. It appeared that this process did not obey the lowest energy principle, because the fragmentation of dendrites caused the increase of solid–liquid interfacial energy. However, with increasing holding time, the temperature of samples increases. Under this condition, the energy system in the sample could sustain the lowest level only when an amount of liquid phase was formed. Namely, the decrease of energy caused by the formation of liquid phase was larger than the increase of energy caused by the fragmentation of dendrites. Note that most of Al_2O_3 short fibers were present in the liquid phase (Fig. 3b). When the holding time was prolonged, these individual particles with polygonal shape were spheroidized into globular particles driven by the decrease of solid–liquid interface energy. The presence of

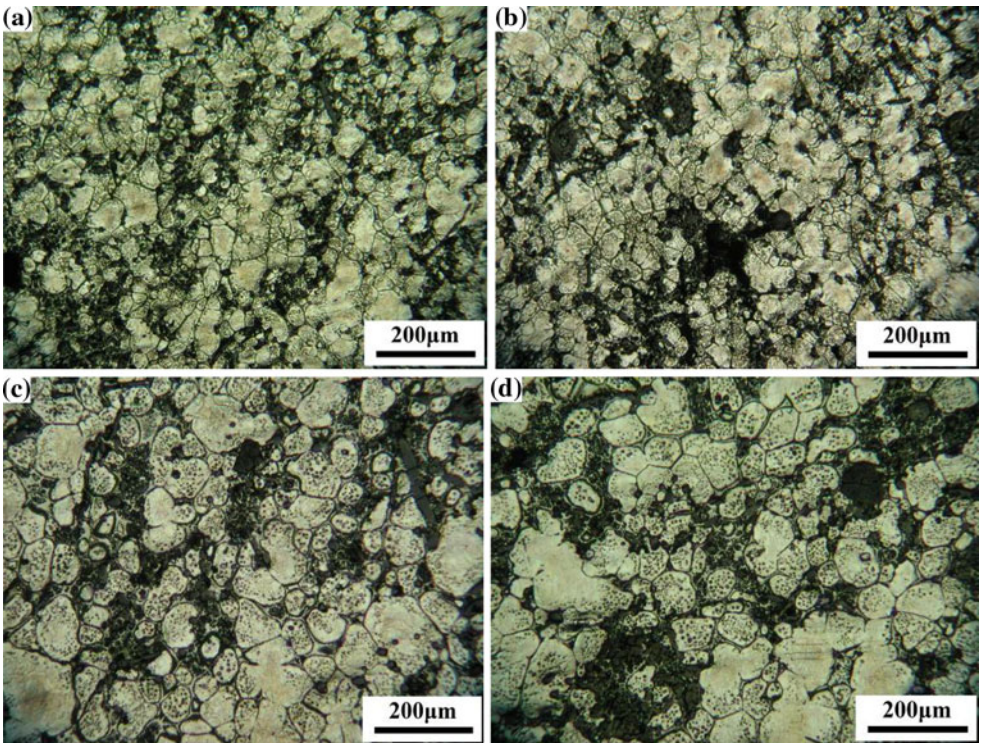

Fig. 3 Optical micrographs of squeeze casting formed Al_2O_{3sf}/AZ91D composites partially remelted at 550 °C for different holding times of **a** 8 min, **b** 16 min, **c** 32 min, and **d** 40 min

liquid phase enhanced spheroidization of the newly formed grains because the corners and edges of solid particles would melt. However, there were still some big and irregular grains in the sample after a long time isothermal holding. Such irregular grains could be present for a variety of reasons. First, the insoluble Al_2O_3 short fibers were present in liquid phase. These Al_2O_3 short fibers were of a size where they could potentially hinder Ostwald ripening of solid particles through inhibiting diffusion through the liquid from one boundary position to another. Tzimas and Zavaliangos [11] suggested that Ostwald ripening driven by the presence of variable curvature on the surface of each grain contributed to a large extent to grain spheroidization. The second, more likely explanation was that the degree of liquid penetration depended on the local balancing of surface energies, i.e., on the magnitude of the local grain boundary and solid–liquid surface energy, noted as γ_{gb} and γ_{sl}, respectively. A grain boundary was not penetrated by liquid when $\gamma_{gb} < 2\ \gamma_{sl}$. Comparison of Fig. 3a–d also shows that significant solid particle coarsening had occurred in the semi-solid state after 32 min. The holding time at the temperature should be short to avoid excessive solid particle coarsening, which in turn resulted in inferior mechanical properties in the thixoforged component.

Figure 4 shows the variations of the solid particle size and shape factor after being isothermally held at 550 °C for

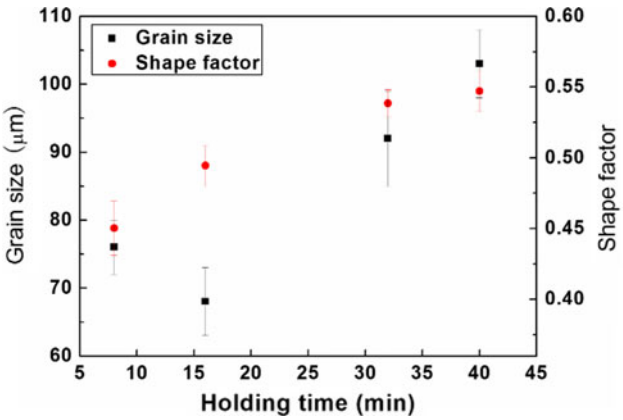

Fig. 4 Grain size and shape factor of the Al_2O_{3sf}/AZ91D composites plotted as a function of holding time

times in the range of 8–40 min. As shown in Fig. 4, the shape factor monotonously increased with prolonged holding time, while the solid particle size slightly decreased during the period from 8 to 16 min and then obviously increased. The decrease of solid particle size in the early stage of isothermal holding attributed to the fragmentation of dendrites. Solid particle coarsened due to Ostwald ripening and calescence. Increasing holding time was beneficial for the formation of liquid, which promoted the degree of spheroidization.

Structural evolution with increasing holding temperature during partial remelting

Figure 5 shows the microstructure evolution of the composites after partial remelting with different predetermined temperatures of 540, 550, 560, and 570 °C for 10 min, respectively. It can be seen from Fig. 5 that the solid particle size gradually decreased and the shape changed from polygonal to globular with increasing temperature. At the same time, the amount of liquid phase increased with increasing temperature. When the holding temperature was relatively low, the amount of liquid was limited. Solid particle coalescence and Ostwald ripening were thought to operate simultaneously and independently as soon as liquid was formed [11]. Because the holding temperature was relatively lower, diffusion was slower and the process of liquid penetration toward dendrites was slower. Coalescence was a dominant mechanism of solid particle coarsening. Furthermore, because the thickness of liquid film was relatively low, polygonal solid particles tended to merge into newly formed solid particles. With increasing temperature, Ostwald ripening was more active and the spheroidization of solid particles promoted. Furthermore, with increasing temperature, the edges of solid particles would be melted. At the same time, the distance between solid particles increased due to the increase of liquid phase and thus the probability of the merged decreased. Compare Fig. 5a with Fig. 5d, increasing temperature promoted the speed of microstructure, including the formation of liquid, fragmentation, and the degree of spheroidization. Therefore, the holding temperature should be as high as possible to shorten holding time. However, if the holding temperature was too high, it was difficult for holding semi-solid feedstocks.

Figure 6 shows the variations of the solid particle size and shape factor after being isothermally held for 10 min at different temperatures. As shown in Fig. 6, as the holding

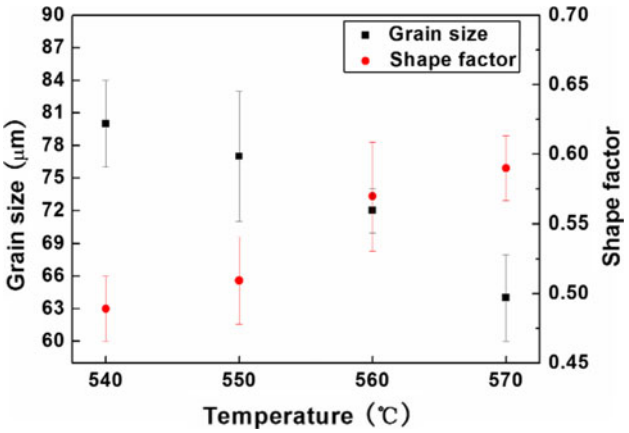

Fig. 6 Grain size and shape factor of the Al_2O_{3sf}/AZ91D composites plotted as a function of holding temperature

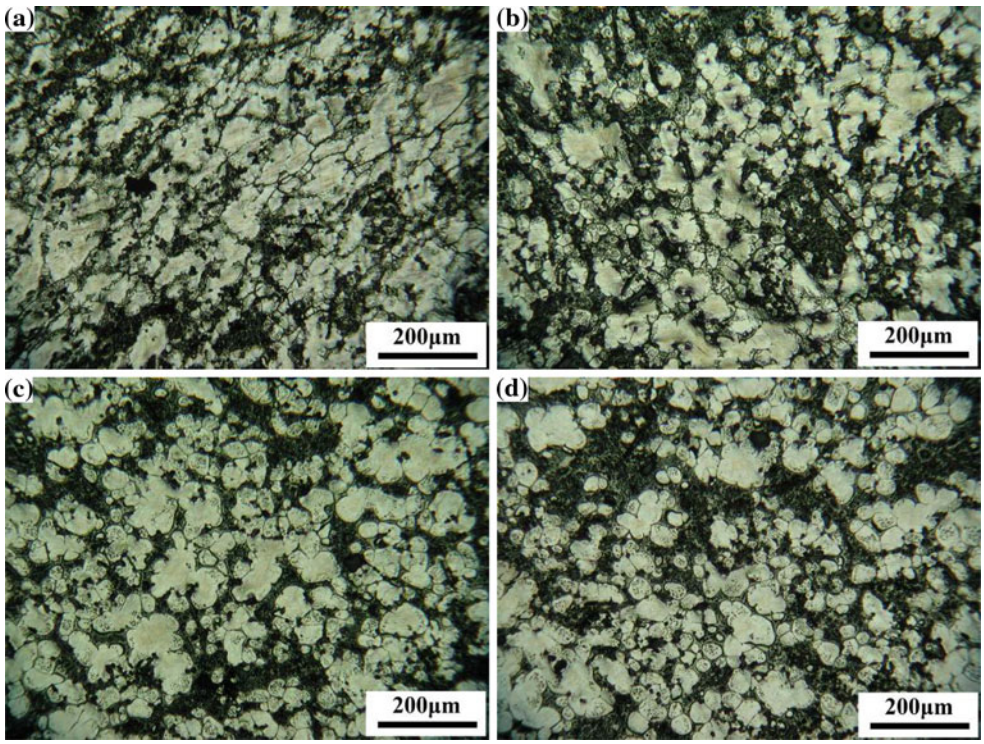

Fig. 5 Semi-solid microstructure of Al_2O_{3sf}/AZ91D composites with different partial remelting temperatures of **a** 540 °C, **b** 550 °C, **c** 560 °C, and **d** 570 °C for 10 min isothermal holding

temperature increased, the solid particle size decreased continuously, while the shape factor increased monotonously. The former was attributed to fragmentation and the latter was attributed to the dissolution-reprecipitation mechanism driven by the presence of variable curvature on the surface of each solid particle.

Component production trials and mechanical properties of thixoforged component

Component production trials have been conducted to confirm the formability of Al_2O_{3sf}/AZ91D composites in the semi-solid state. Figure 7 shows photography of thixoforged automotive component. The result indicated that the thixoforged component had a very good surface finish and close-to-zero porosity. The production of the thin wall was shown to be good, and no problem was experienced in filling the thin-walled sections. Figure 8 shows mechanical properties obtained on the squeeze casting formed and the thixoforged conditions. In each condition at least three samples were tested in each condition and there is no considerable scatter in the results. As shown in Fig. 8, yield strength (YS) and ultimate tensile strength (UTS) in the thixoforged condition were found to be higher than those in the squeeze casting formed condition. For example, the average YS and UTS of squeeze casting formed sample increased from 206 and 256 MPa to 216 and 273 MPa in the thixoforged condition, respectively. Although the elongation to facture for thixoforged sample was a little worse than that of squeeze casting formed sample, the mechanical properties of thixoforged sample were still encouraging. Thixoforging was the shape of metal or MMCs components in the semi-solid state [12]. The processing temperature was the solidus and liquidus of metal or matrix phase and globular solid particles were distributed in the liquid matrix

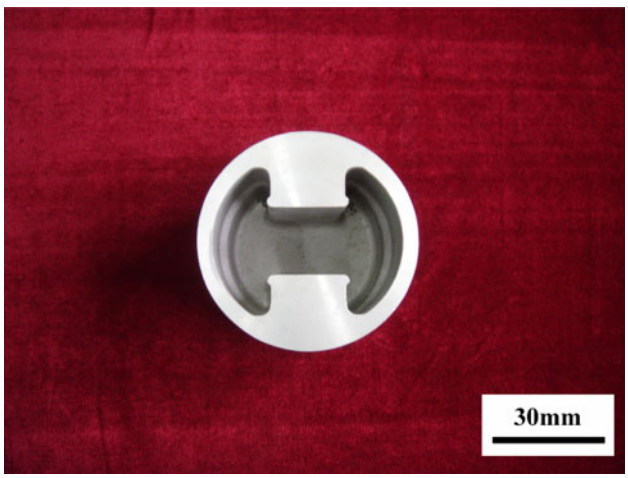

Fig. 7 Automotive piston component made from Al_2O_{3sf}/AZ91D and thixoforged under an applied pressure of 350 MPa

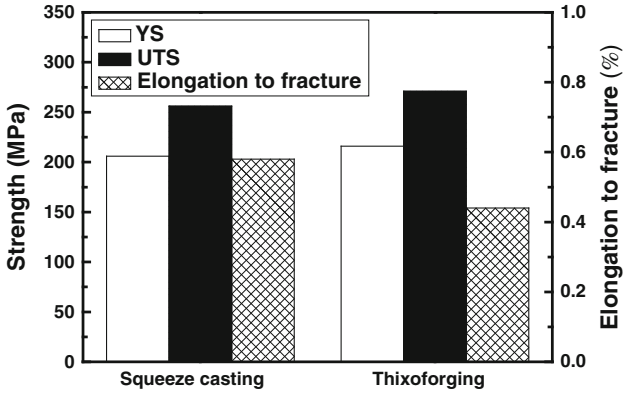

Fig. 8 Tensile mechanical properties of squeeze casting formed Al_2O_{3sf}/AZ91D and thixoforged automotive piston component with thin wall at room temperature (Each tensile value was the average of at least three measurements)

homogeneously in the thixotropic feedstock before thixoforging. Partial remelting and isothermal holding of the dendritic structures produced by squeeze casting would produce globular structures with adequate rheological properties for thixoforging. A key question was why the mechanical properties (YS and UTS) of thixoforged sample increased, compared with those of squeeze casting formed sample. This issue was related to the change of microstructure during partial remelting and applied pressure in thixoforging. For a squeeze casting formed sample, its microstructure referred to as-cast microstructure in essential. After partial remelting, due to fragmentation and liquid penetration, original dendritic structures were changed into fine semi-solid structures, in which relatively globular primary phase particles were surrounded by liquid phase and Al_2O_3 short fibers mainly distributed in liquid phase. It was reasonable to expect fairly higher YS and UTS, as shown in Fig. 8. On the other hand, second solidification during thixoforging was a process of nucleation and growth. This process was influenced by the rate at which heat was transferred which in turn influenced the structure and mechanical properties of the thixoforged component. During thixoforging, due to the relatively high applied pressure of 350 MPa, not only porosity existing in squeeze casting formed sample was decreased, but also the rate of cooling was increased. Under this condition, good mechanical properties of thixoforged component were obtained.

Conclusions

The microstructure evolution of squeeze casting formed Al_2O_{3sf}/AZ91D composites experienced four stages during partial remelting: the formation of liquid, structural fragmentation, the spheroidization of solid particles, and final coarsening. The shape factor monotonously increased with

prolonging holding time, while the solid particle size slightly decreased during period from 8 to 16 min and then obviously increased. As holding temperature increased, the solid particle size decreased continuously, while the shape factor increased monotonously.

Thixoforging for Al_2O_{3sf}/AZ91D composites resulted in successful filling of the die. The yield strength and ultimate tensile strength of the thixoforged part were satisfactory and exceed the squeeze casting quality for Al_2O_{3sf}/AZ91D.

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