Microstructure stability of as-extruded bimodal size SiCp/AZ91 composite

Jian-chao Li a, Kai-bo Nie a, Kun-kun Deng a,b,*, Shuan-jun Shang a, Shan-shan Zhou a, Fang-jun Xu a, Jian-feng Fan b

a College of Materials Science and Engineering, Taiyuan University of Technology, Taiyuan 030024, China
b Key Laboratory of Interface Science and Engineering in Advanced Materials, Ministry of Education, Taiyuan University of Technology, Taiyuan 030024, China

A R T I C L E   I N F O

Article history:
Received 20 April 2014
Received in revised form 29 July 2014
Accepted 31 July 2014
Available online 7 August 2014

Keywords:
Magnesium matrix composites
SiCp
Microstructure
Micro-hardness

A B S T R A C T

The microstructure stability and micro-hardness of as-extruded bimodal size (micron + submicron) SiCp/AZ91 composites were investigated. Results show that the significant grain refinement appears around micron SiCp at the temperature range of 643–693 K. Compared with monolithic AZ91 alloy and micron SiCp/AZ91 composite, the obvious submicron particle dense zones (SPDZs) are formed in the vicinity of micron SiCp in bimodal size SiCp/AZ91 composite during thermal stability test, which results in the small average grain size and improved microstructure stability. The micro-hardness of bimodal size SiCp/AZ91 composite is higher than that of monolithic AZ91 alloy and micron SiCp/AZ91 composite. The micro-hardness of bimodal size SiCp/AZ91 composite depends strongly on grain size and the lowest micro-hardness appears at 643 K.

1. Introduction

Magnesium alloys have received extensive attentions for their low density, high specific strength and stiffness in recent years [1–4]. But the low modulus, strength and abrasion resistance have limited their industrial applications. Previous studies have revealed that particle reinforced magnesium matrix composites (PMMCs) can overcome the demerits of the monolithic Mg alloys and expand their industrial application for their simple preparation process and low cost [5,6]. However, casting defects such as pore and particle agglomerates etc. that existed in the as-cast composite reduce their mechanical properties. Besides, the poor bonding strength between particles and Mg matrix will also reduce PMMCs’ mechanical properties [7,8]. Previous researches have revealed that the improvement of interface strength was an effective way to enhance mechanical properties [9,10]. The application of second process e.g. forging, rolling and extrusion, has a significant effect on refining grain size, improving particle distribution and enhancing bonding strength [11–16]. Deng et al. [17,18] found that the forging process has a significant effect on refining grain size, improving particle distribution and interfacial bonding between SiCp and Mg, which results in the enhancement of tensile properties. On Wang et al.’s investigation of SiCp/Mg–Zn–Ca composite [19], the interface between SiCp and the matrix was also modified by hot extrusion, which was thought to be one reason for the improved mechanical properties. Shang et al. [20] found that tensile properties of SiCp/Mg–Al–Zn composite can be affected by extrusion temperature through influencing grain size, texture, the amount and size of Mg17Al12 phase. Two-step deformation (forge + extrusion) has been applied by Wu et al. [16] on the 5 μm SiCp/AZ91 composite; the further improved tensile properties are obtained. Recently, Deng et al. [21] found that microstructure and mechanical properties of SiCp/AZ91 composites processed by two-step deformation can be affected significantly by particle size. In order to give full play to the merits of different particle sizes, the fine grained bimodal size (submicron–micron) SiCp/AZ91 composite has been obtained by author’s previous research [22]; the results showed that the mixture of a little amount fine particles and micron particles had significant influence on enhancing the tensile properties of Mg matrix.

However, less noted, but no less important, the microstructure stability which means the extent of grain growth during thermal treatment of fine grains will directly influence the application of magnesium matrix composite under high temperature conditions. Radi et al. [23] have reported that the introduction of 2 wt% Al2O3 nano-particles into the base AZ31 alloy could restrict grain boundary migration during the annealing treatment and increase the activation energy of grain growth, thus resulting in enhanced microstructure stability. Ferry et al. [24] have reported that the existence of ~5 nm diameter Al3Sc particles in fine-grained Al–Sc...
alloys could improve the microstructure stability significantly at temperatures up to 773 K. Compared with nanoparticles, micron particles have different influences on microstructure stability.

Robson et al. [25] have reported that the addition of micron particles (>1 μm) could stimulate DRX during deformation. While during the subsequent annealing, fine static recrystallized

Fig. 1. (a) SEM micrographs of as-extruded bimodal size SiCp/AZ91 composite before thermal stability test and (b) the EDS results of point “A” marked in (a).

Fig. 2. OM micrographs of as-extruded bimodal size SiCp/AZ91 composite before (a) and after thermal stability test (30 min) at (b) 543 K, (c) 593 K, (d) 643 K, (e) 693 K and (f) 743 K.
grains forms in the particle-stimulated nucleation (PSN) zones around micron particles. However, no research on the microstructure stability of fine grained SiCp/AZ91 composites at high temperature conditions has been reported in the open literature. Therefore, the aim of the present work is to investigate the microstructure stability of as-extruded bimodal size SiCp/AZ91 composites. In order to characterize the mechanical properties of the composites after thermal treatment, the micro-hardness test is conducted.

2. Materials and methods

2.1. Materials preparation

AZ91 magnesium alloy was selected as the matrix alloy. Two sizes of SiC particles with the average particle diameter of ~10 μm and ~0.2 μm were used as reinforcement. The fine-grained bimodal size (0.2 μm 1 vol% + 10 μm 9 vol%) SiCp/AZ91 composites were fabricated by the combination of stir casting technology and the following two-step hot deformation. The details of fabrication process are described in Refs. [22,26].

2.2. Thermal stability tests

The as-extruded bimodal size SiCp/AZ91 composites bars were sliced into 5 mm-thick samples to conduct the thermal stability tests. The samples were held at the temperatures of 543 K, 593 K, 643 K, 693 K and 743 K. For each temperature, the holding time was chosen as 30 min, 4 h, and 8 h. For the purpose of comparison, the fine-grained AZ91 alloy and 10 μm 10 vol% SiCp/AZ91 composite were prepared under the same conditions and then held at 693 K for 4 h.

2.3. Microstructure characterization

The microstructure characterization was carried out by a 4XC optical microscope (OM) and a MIRA 3XMU scanning electron microscope (SEM) equipped with energy spectrum (EDS). The annealed samples were ground, polished and etched in acetic picral (5 ml acetic acid + 6 g picric acid + 10 ml H2O + 100 ml ethanol (95%)) to investigate the morphological characteristics of grains. The average grain size was measured by Image Pro plus software. To make the measured grain size more representative, at least four micrographs for each sample were used.

2.4. Micro-hardness

The Vickers micro-hardness (Hv0.2) of the composites was measured by an EM-1500TK digital Vickers hardness tester with a load of 200 g for 15 s. The specimens were polished before the test. At least six points for each sample were tested to obtain the average micro-hardness.
3. Results and discussions

3.1. Microstructures

SEM micrographs of as-extruded bimodal size SiCp/AZ91 composites before thermal stability test are demonstrated in Fig. 1. As shown in Fig. 1(a), the grain size of composite matrix is fine. And the micron SiC particles are mainly distributed around grain boundaries while the submicron SiC particles are distributed both around grain boundaries and within grains. In addition, the precipitated secondary phase is found mainly at grain boundaries, as shown in Fig. 1(a). The EDS result of point “A” in Fig. 1(a) demonstrates that the secondary phase is $\beta$-Mg$_{17}$Al$_{12}$, as illustrated in Fig. 1(b). Feng et al. [27] have reported that Mg$_{17}$Al$_{12}$ can be precipitated from AZ91 alloys at ~523 K, which is lower than the present extrusion temperature (643 K). Thus, the Mg$_{17}$Al$_{12}$ phase might be precipitated at grain boundaries during the cooling process after extrusion.

Fig. 2 illustrates the OM micrographs of as-extruded bimodal size SiCp/AZ91 composites before and after thermal stability test under different temperatures. The holding time of the thermal stability test is

![Fig. 4. The statistical grain size distributions of as-extruded bimodal size SiCp/AZ91 composite before (a) and after thermal stability test (30 min) at (b) 543 K, (c) 593 K, (d) 643 K, (e) 693 K and (f) 743 K.](image-url)
30 min for the composites. Fig. 3 shows the magnified SEM micrographs of fine-grained bimodal size SiCp/AZ91 composites after thermal stability test under the same condition with Fig. 2(d) and (f). Fig. 4 shows the distribution of grain size in the as-extruded bimodal size SiCp/AZ91 composite before and after thermal stability test for 30 min. It can be found that the grains grow up after thermal stability test. At 543–593 K, the average grain size increases with increasing temperature, as shown in Figs. 2 and 4. In the initial microstructure, the grain size is uniform and the peak of grain size distribution mainly concentrates at 0.6–1.8 μm, as shown in Fig. 4(a). This peak is transferred to 1.6–3.0 μm and 1.6–3.8 μm as the temperature increases to 543 K and 593 K, respectively, as shown in Fig. 4(b) and (c). However, as the temperature increases to 643 K, the grain size distribution becomes non-uniform. Fine static recrystallized grains appears around micron SiC particles while those away from SiCp grow up, as depicted in Figs. 2(d) and 3(a) and (b). And inconspicuous bimodal grain size distribution, with two peaks at 0.8–1.8 μm and 2.0–3.0 μm, appears in Fig. 4(d). As the temperature increases to 693 K, the volume fraction of fine grains around micron SiCp particles increases, while the grains in the region away from micron particles grow up, as shown in Fig. 2(e). The peak also transfers from 0.8–1.8 μm (643 K) to 0.2–1.6 μm (693 K) and the amount of fine grains is also increased, which results in the small average grain size (~1.86 μm), as shown in Fig. 4(e). However, the grain size distribution of composite matrix changes a little as the temperature increases to 743 K by comparing Fig. 2(f) with (e). It can be illustrated that the amount of fine grains decreases while the amount of coarse grains increases as the temperature increases to 743 K.

By the observation and analysis of Figs. 1–4, it can be concluded that grain growth plays a main role when the composites are held at 543 and 593 K. As the temperature increases to 643 K, static recrystallization occurs in the zone around micron particles, which results in the bimodal distribution of grain size. The bimodal size distribution of grain size has also been reported on nano-Al2O3p reinforced AZ31 magnesium alloys [23] and nano or ultrafine-grained magnesium [28,29]. Habibnejad-Korayem et al. [30] have reported that the difference between the coefficient of thermal expansion (CTE) values of nano-particles and composite matrix generates geometrically necessary dislocations and thermally induced residual stresses. For the present bimodal size SiCp/AZ91 composites, the stored energy in the zones around micron particles can be produced during the cooling process. When the composites are held at higher temperatures than 643 K, the stored energy will be released, which is propitious to promote the nucleation of static

Fig. 5. OM micrographs of as-extruded (a) and (b) AZ91 alloy, (c) and (d) 10 μm 10 vol% composite before and after thermal stability test: (a) and (c) the initial microstructure and (b) and (d) after the thermal stability test at 693 K for 4 h; (e) bimodal size SiCp/AZ91 composite after the thermal stability test at 693 K for 4 h; (f) the variation of average grain size of above three materials before and after the thermal stability test.
recrystallization. However, the grain growth is hindered by micron particles. So the above two reasons lead to the fine grain size around micron particles. The extent of static recrystallization is enhanced and the nucleation rate increases as the temperature increases from 643 K to 693 K, which results in the small average grain size. As the temperature continuously increased from 693 K to 743 K, the static recrystallization nucleation and grain growth occur at the same time. However, the grain growth rate is slightly higher than the nucleation rate, so the average grain size increases slightly at 743 K.

3.2. Microstructure of Mg matrix affected by bimodal size SiCp

To investigate the influence of bimodal size SiCp on the microstructure of composite matrix during thermal stability test, the monolithic AZ91 alloy and micron SiCp/AZ91 composite were prepared under the same conditions and then held at 693 K for 4 h as a comparison. OM micrographs of above three kinds of material before and after thermal stability test at 693 K for 4 h are demonstrated in Fig. 5. Before thermal stability test, the optical microstructures of monolithic AZ91 alloy and micron SiCp/AZ91 composite are shown in

![Fig. 6. SEM micrographs of bimodal size SiCp/AZ91 composites after thermal stability test at 693 K for (a) and (b) 30 min, (c) and (d) 4 h and (e) and (f) 8 h.](image-url)
Fig. 5(a) and (c), respectively. After the tests, the equiaxed and coarse 
grains are observed in the AZ91 alloy, as shown in Fig. 5(b). The 
grains in the composites is significantly refined as compared with 
AZ91 alloy, as given in Fig. 5(d) and (e). However, the grain size 
distribution is different in micron and bimodal size SiCp/AZ91 
composite. The grains are much more uniform in micron SiCp/AZ91 
composite while large amounts of fine grains appear around micron 
SiC particles in bimodal size SiCp/AZ91 composite, as shown in Fig. 5 
(d) and (e). The average grain size of the three kinds material before 
and after the thermal stability test is illustrated in Fig. 5(f). After being 
treated at 693 K for 4 h, the grains of AZ91 alloy are about 
8.3 times larger than that of the original one. While, the increased 
grain size of micron and bimodal size SiCp/AZ91 composites are 
~284% and ~135%, respectively. The above phenomenon means 
that the addition of micron SiCp has an obvious inhibiting effect on 
grain growth. While, the microstructure stability of micron SiCp 
and AZ91 composites can be further improved by the addition of 
submicron SiC particles.

To reveal the influence of bimodal size SiCp on the composite 
matrix during thermal test, the SEM micrographs of bimodal size 
SiCp/AZ91 composites at 693 K for 30 min, 4 and 8 h are given in 
Fig. 6. By comparing Fig. 6(a) with Fig. 1(a), it can be found that the 
precipitated Mg17Al12 phase disappears and grain growth occurs in 
the zone away from micron particles at 30 min. The temperature 
(693 K) for the thermal test is higher than the solution tempera-
ture of Al in Mg (688 K [27]), so the Mg17Al12 phase can be 
dissolved into Mg. Besides, the submicron SiCp mainly distributes 
at grain boundaries in static recrystallization zones and the 
regions away from micron SiCp, as shown in Fig. 6(a) and (b). 
With the increase of holding time, the amount of submicron SiC 
particles at grain boundaries and in the region around micron SiC 
increases, as shown in Fig. 6(c)–(f). Robson et al. [31] and Cao et al. 
[32] have reported that the existence of secondary phase particles 
have a hinder effect on grain boundary migration. Compared with 
the 10 μm SiCp, the pinning effect of submicron SiCp on grain 
boundary is weaker. So the submicron SiCp moves together with 
grain boundary which leads to the increase of submicron SiCp at 
grain boundary. Usually, the grain boundary migration is stopped 
once they meet with micron SiC particles. Thus, the submicron 
particle dense zones (SPDZs) can be formed in the regions around 
micron particles, as shown in Fig. 6(e) and (f). In these regions, the 
grain boundaries are pinned by large amount of submicron 
particles, thus it is hard for them to move during the subsequent 
thermal treatment, which results in the fine grain size around 
micron SiC particles. On the contrary, the grains grow up easily in 
the region away from micron particles owing to the less amount of 
micron SiC particles and SPDZs.

3.3. Micro-hardness

The variation of bimodal size SiCp/AZ91 composite's micro-hardness 
with temperature (at 30 min) is plotted in Fig. 7(a). It is obvious that the 
micro-hardness decreases as the temperature increases from 543 K to 
643 K. Then the micro-hardness increases slightly as the temperature 
increases from 643 K to 743 K. Fig. 7(b) shows the micro-hardness and 
the footprint of AZ91 alloy, micron and bimodal size SiCp/AZ91 
composite before and after thermal stability test (at 693 K for 4 h). 
Even though the micro-hardness of the three kinds materials decreases 
after thermal stability test, the micro-hardness of SiCp reinforced 
magnesium matrix composites is higher than that of AZ91 alloy. After 
thermal stability test, the micro-hardness of AZ91 alloy is 68.3 Hv0.2, 
while that of the micron and bimodal size SiCp/AZ91 composite is 
100.3 Hv0.2 and 124.2 Hv0.2, respectively. It can be seen that the largest 
micro-hardness is obtained in bimodal size SiCp/AZ91 composite. 
Besides, the decrease of micro-hardness in AZ91 alloys is about 22.4% 
after thermal stability test. However, the decrease of micro-hardness is 
17.7% and 10.3% for micron and bimodal size SiCp/AZ91 composite, 
respectively. It means the existence of bimodal size SiC particles is 
propitious to the improvement of microstructure thermal stability in 
the Mg matrix at elevated temperatures.

The Hall–Petch (H–P) relationship has been frequently used to 
investigate the contribution of grain boundary strengthening as 
well as other strengthening mechanisms to strength and hardness. 
The general form of the H–P relationship for hardness is given by

$$H_V = H_0 + KD^{-1/2}$$  \((1)\)

where $H_V$ is the measured micro-hardness, $H_0$ is the lattice fraction 
hardness which is a material constant and represents the overall 
resistance of the crystal lattice to dislocation movement, $D$ is the 
average grain size, and $K$ is a material constant which reflects the 
hardening contribution of grain boundaries. Eq. (1) demonstrates 
that the change of micro-hardness depends on grain size. The 
smaller the grain size, the larger the micro-hardness. As discussed 
in Section 3.1, the average grain size of the bimodal size SiCp/AZ91 
composite increases as the temperature increases from 543 K to 
643 K, which leads to the decrease of micro-hardness. However, 
the average grain size decreases as the temperature is higher than 
643 K owing to the occurrence of static recrystallization, which 
leads to the increase of micro-hardness.
After the thermal stability test at 693 K for 4 h, the average grain size of SiCp reinforced magnesium matrix composites is much finer than monolithic AZ91 alloy, as mentioned in Section 3.2. According to the H–P relationship, the significant refinement of grains of SiCp/AZ91 composite is one reason contributing to the high micro-hardness. Besides, the mismatch of CTE between SiC particles and AZ91 matrix leads to geometrically necessary dislocation generation in the vicinity of particle during cooling process after thermal stability test [30]. The increased dislocation density may also lead to the increase of micro-harness of SiCp/AZ91 composite.

4. Conclusions

The microstructure stability of as-extruded bimodal size SiCp/AZ91 composites was investigated and the following conclusions can be obtained:

(1) The grains grow up with the increase of time and temperature during thermal stability test. However, the significant grain refinement appears at the temperature range of 643–693 K due to the occurrence of static recrystallization around micron particles.

(2) The SPDZs are formed around micron particles during the treatment, which exhibit high stability at elevated temperature. Compared with monolithic micron SiCp/AZ91 composite, the existence of bimodal size SiCp is propitious to the stability of microstructure at elevated temperatures.

(3) Micro-hardness of the composites do not vary linearly with the increase of temperature and the lowest micro-hardness value appears at 643 K. The micro-hardness of bimodal size SiCp reinforced magnesium matrix composite is higher than that of monolithic AZ91 alloy and micron SiCp/AZ91 composite.

Acknowledgments

This work was supported by the “National Natural Science Foundation of China” (Grant nos. 51201112 and 51174143), the “Specialized Research Fund for the Doctoral Program of Higher Education” (Grant no. 20121402120004), and the “Natural Science Foundation of Shanxi” (Grant no. 2013021013-3).

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