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Effect of ultrasonic on morphology of primary Mg_2Si in in-situ Mg_2Si/Al composite

Jia-tao ZHANG, Yu-guang ZHAO, Xiao-feng XU, Xiao-bo LIU

Key Laboratory of Automobile Materials of Ministry of Education,
College of Materials Science and Engineering, Jilin University, Changchun 130025, China

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Abstract: Effects of ultrasonic on morphologies of primary Mg_2Si crystals in in-situ Mg_2Si/Al composite were investigated by metallographic microscopy and field emission scanning electron microscopy. The results show that the mean grain size of primary Mg_2Si crystals is refined from 150 to 20 μm by high intensity ultrasonic, and the morphologies of primary Mg_2Si crystals are changed as well. Optical microscopy reveals that primary Mg_2Si crystals without ultrasonic vibration exhibit coarse particles with cavities, in which eutectic structures grow. However, primary Mg_2Si crystals with ultrasonic vibration appear fine grains without any cavity. Three-dimensional morphologies of primary Mg_2Si without ultrasonic vibration display octahedron and tetrakaidecahedron with hopper-like hole in the crystals. After ultrasonic vibration, primary Mg_2Si particles become solid crystals with rounded corners and edges.

Key words: ultrasonic; Mg_2Si/Al composite; Mg_2Si; morphology; grain refinement

1 Introduction

Particulate reinforced aluminum metal matrix composites (MMCs) have significant implication potentiality in aircraft and automobile industries and attract worldwide increasing attention due to their improved properties, such as low density, excellent castability, good wear resistance and fine physical properties [1−3]. Magnesium silicide (Mg_2Si intermetallic compound), as a type of in-situ reinforcement phase, exhibits high melting temperature (1085 °C), low density (1.99×10^3 kg/m^3), high hardness (4.5×10^9 N/m^2), low thermal expansion coefficient (7.5×10^{-6} K^{-1}) and high elastic modulus (120 GPa) [1]. In-situ Mg_2Si particulates reinforced aluminum MMCs are excellent materials for producing auto pistons and brake discs due to their low density, high specific tensile strength and wear resistance [1,3,4]. However, Mg_2Si crystal often grows into coarse dendrite or grain during conventional solidification procedure [5], resulting in the decrease of mechanical properties of the composites.

For the purpose of refining primary Mg_2Si grains, many processing technologies have been developed, including modifier material addition, such as P [1], Sr [1], Y [5] and Sn [6], and rapid cooling casting [2]. In recent years, high-intensive ultrasonic shows large potential in refining solidification structure of light alloy. ZHANG et al [7] fabricated in situ (Mg_2Si+MgO)/Mg composite with assistance of the high-energy ultrasonic and found that the morphologies of in situ Mg_2Si particles were changed to smooth olive-shape or spherical shape. KO et al [8] used ultrasound to refine the primary Mg_2Si in Al−Mg−Si alloys and found that the morphology of primary Mg_2Si was also changed to polygonal from irregular shape. However, CHEN and LIN [9] revealed that high intensity ultrasonic vibration had no essentially influence on morphology of the primary Mg_2Si in Mg_2Si/Mg composite.

Despite extensive previous studies have been aiming at refinement of primary Mg_2Si, whether and how the morphologies of primary Mg_2Si would be changed by high energy ultrasonic is still wanting. This work attempts to reveal how high energy ultrasonic influences the morphologies of primary Mg_2Si by comparing the features of the section and the three-dimensional morphologies of primary Mg_2Si in Mg_2Si/Al composites with and without high energy ultrasonic vibration.

Corresponding author: Yu-guang ZHAO; Tel: +86-431-85094481; E-mail: zhaoyg@jlu.edu.cn
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2 Experimental

Commercial Al−20%Si (mass fraction) master alloy ingot and pure Mg ingot were used to prepare 20%Mg_2Si/Al composite. About 180 g of Al−Si alloy was melted in an alumina crucible in a 3 kW electric furnace. Then, about 19 g of pure Mg, wrapped by aluminum foil and preheated at 300 °C, was added into the melt protected by argon atmosphere. After holding at 780 °C for 10 min, the composite melt was poured into a steel crucible preheated at 600 °C and bolted with the horn. The schematic diagram of ultrasonic devices is shown in Fig. 1. The ultrasonic system with frequency of 20 kHz and whole power of 1 kW was started immediately to treat the melt until the solidification process was finished. Then the resulting composite with ultrasonic vibration was stripped away from the steel crucible. For comparison, the Mg_2Si/Al composite without ultrasonic vibration was fabricated.

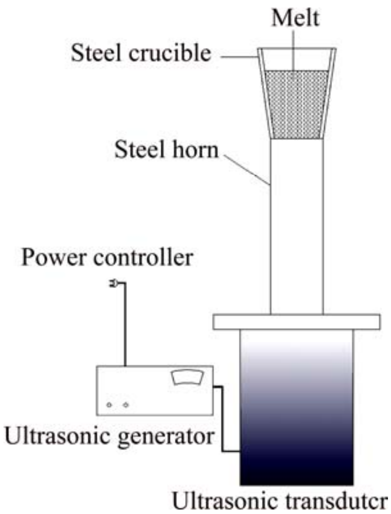

Fig. 1 Schematic diagram of ultrasonic devices

Specimens for optical microscopy analysis were cut from each resulting composite. The sections were polished through standard routines and etched by hydrofluoric acid (HF) water solution (0.5%) for 180 s. Characterization of the microstructure and the qualitative analysis were carried out on an OLYMPUSBX-60 metallographic microscopy with an image collection and analysis system. For observing the three-dimensional morphologies of primary Mg_2Si, one small piece of material was cut off from the center part of every resulting composite and cleaned with ultrasonic cleaner. NaOH water solution (25%) was used as etchant to extract primary Mg_2Si particulates. The extracted particulates were cleaned repeatedly and alternatively with an ultrasonic cleaner by water and alcohol to break up eutectic structures connected to primary Mg_2Si particles. The detailed 3D features of primary Mg_2Si were characterized by field emission scanning electron microscopy (FESEM, JSM-6700F).

3 Results and discussion

3.1 Grain refinement of primary Mg_2Si by ultrasonic

Figure 2 shows the optical microscopy microstructures of the composites and planar characters (polishing section of primary Mg_2Si grain) of primary Mg_2Si particles. Primary Mg_2Si crystals in the composite without ultrasonic vibration grow into coarse grains with the mean size of 150 μm, while primary Mg_2Si particles in composite with ultrasonic vibration grow into fine grains with the mean size of 20 μm.

For grain refinement during solidification procedure, increasing the solidification rate is an effective method. It is explicit that high intensive ultrasonic has great potential in grain refinement for primary phases precipitated during solidification procedure in light metals [10]. Ultrasonic cavitation can produce transient (in the order of nanoseconds) micro hot spots with temperatures of about 5000 °C, pressures above 101 MPa, and heating and cooling rates above 10^{10} K/s [11]. The velocity of acoustic streaming with a maximum speed of 1.37 m/s, produced by ultrasonic vibration [12], can break weak embryos into micro crystal fragments, which are turned into new nucleus for primary Mg_2Si grains. The nucleation rate can be increased by ultrasonic via this way.

It is well known that some impurities particulates in the melt can work as nucleus of heterogeneous nucleation during solidification. However, not all impurity particulates can act as the nucleus effectively. RAMIREZ et al [10] indicated that the high-intensive ultrasonic vibration could improve the nucleation potency of the nucleus. Therefore, those impurity particulates which have qualification as heterogeneous nucleation nucleus can work as nucleus of primary Mg_2Si effectively, and this results in the enhancement of nucleation rate. According to the Clapeyron equation, $\Delta T_m = T\Delta P\Delta V / \Delta H$, the elevation of pressure in the melt, induced by cavitation in a lot of small liquid volume, results in the increase of T_m, which is equivalent to increase of the undercooling and the nucleation rate.

3.2 Effect of ultrasonic on morphology of primary Mg_2Si

As shown in Fig. 2(a), primary Mg_2Si grains, in composite without ultrasonic vibration, have incomplete polygonal planar character with long straight edges and sharp corners. Inside the primary Mg_2Si crystal, cavities are formed during solidification procedure, as shown by the marked sign *A* in Fig. 2(a). It can be seen clearly that

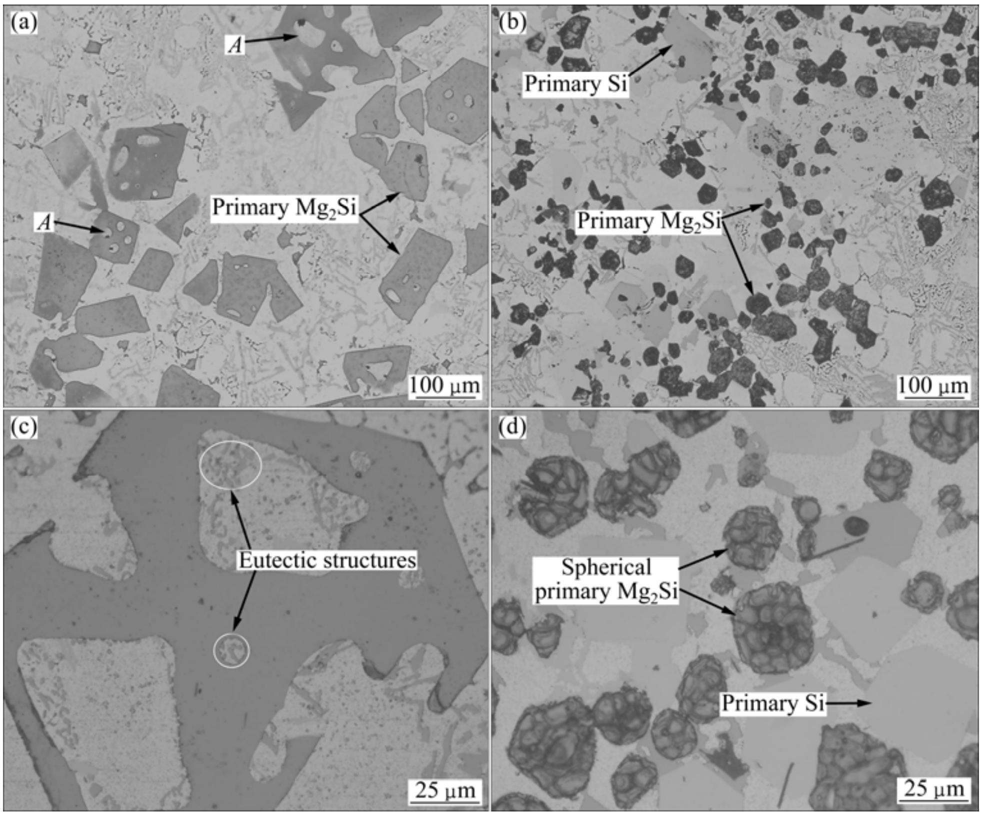

Fig. 2 Morphologies of primary Mg_2Si in Mg_2Si/Al composites without (a) and with (b) ultrasonic vibration, high resolution planar characters of primary Mg_2Si without (c) and with (d) ultrasonic vibration

eutectic structures grow in those cavities, as shown in Fig. 2(c). However, primary Mg_2Si crystals with ultrasonic vibration take shape appearing as solid grains with round corners. Even some of them become spherical particles, as shown in Fig. 2(d). It is interesting that the section of primary Mg_2Si without ultrasonic vibration is clean. However, the sections of primary Mg_2Si with ultrasonic vibration have pattern-like character, revealing that these primary crystals may have different sub-structures.

Theoretically, primary Mg_2Si crystal tends to form faceted octahedron with minimized total surface free energy in conventional solidification conditions [13]. Primary Mg_2Si without any refining treatment, such as refining elements addition and mechanical stirring, often exhibits typical hopper-like crystal shape or enormous dendrite. If primary Mg_2Si is in 15%Mg_2Si/Al composite or modified by P and Sr, it can form perfect octahedron, tetrakaidecahedron and cube respectively [3,13,14]. Figure 3 provides detailed information about the differences between three-dimensional morphologies of primary Mg_2Si with and without ultrasonic vibration. From Fig. 3(a), it can be seen that {111} planes of primary Mg_2Si without ultrasonic vibration are incomplete, on which holes are formed. The edges of them are sharp. This type of morphology is typical hopper crystal. Fig. 3(b) shows a tetrakaidecahedron crystal shape of primary Mg_2Si without ultrasonic, attached with another primary Mg_2Si particle. However, it is an incomplete tetrakaidecahedron, of which there is a hole on the (001) plane, as shown by the marked sign *K* in Fig. 3(b).

Three-dimensional morphologies of primary Mg_2Si with ultrasonic vibration are shown in Figs. 3(c) and (d). It can be seen that the crystals are complete particles without any hole on the planes of the crystal. The edges and corners of these particles are not as sharp as those without ultrasonic vibration, as shown by the marked sign *B* in Fig. 3(c). The shape of primary Mg_2Si is still faceted octahedron, which is determined by the crystal structure. However, {111} planes of primary Mg_2Si with ultrasonic vibration are neither as smooth as perfect octahedron nor as incomplete as those particles without ultrasonic vibration. There are some depressions but not holes on {111} planes, as shown by the marked sign *D* in Figs. 3(c) and (d).

The structure type of Mg_2Si crystal belongs to FCC, whose advantaged growth directions are in $\langle 100 \rangle$ directions via faceted manner, which leads to the transformation of {100} faces into corners [1]. If the high growth rates of $\langle 100 \rangle$ directions are limited, the octahedron will change into other morphologies [13].

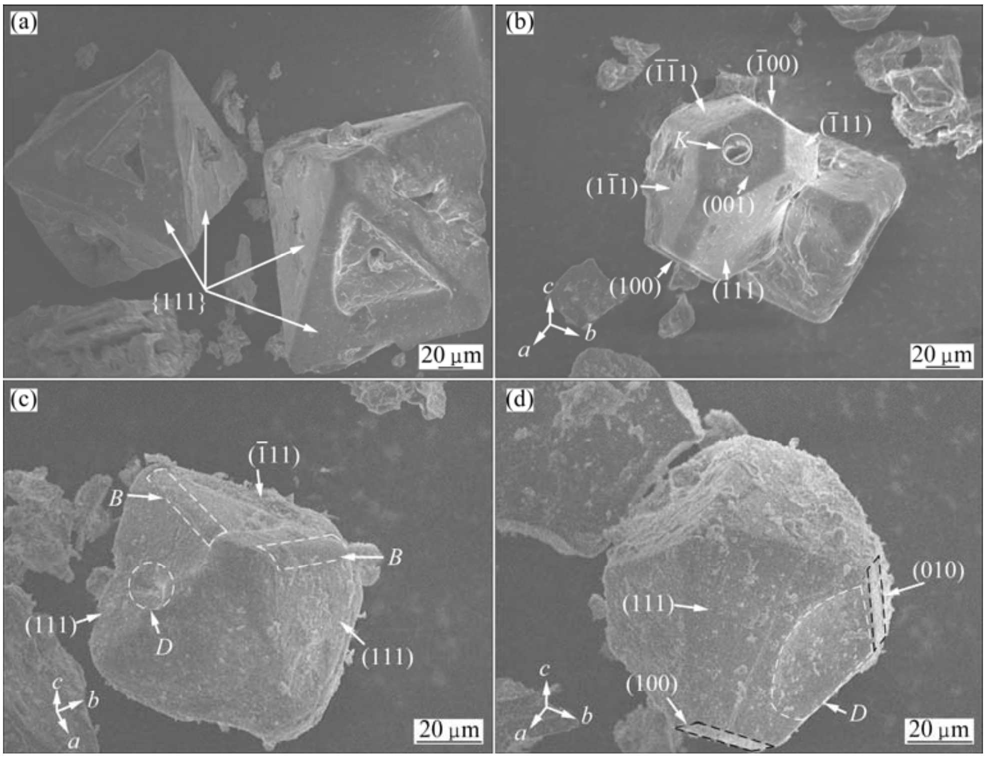

Fig. 3 Three-dimensional morphologies of primary Mg_2Si: (a) Hopper; (b) Incomplete tetrakaidecahedron; (c) Octahedron; (d) Truncated octahedron

Those factors, influencing the growth rates along ⟨100⟩ directions, include the density of Mg and Si atoms in solidification front, modifier atoms and impurities atoms (Fe, Mn, Zn and Ga). During the growing procedure along ⟨100⟩ directions, Al and impurities atoms are expelled from the growing primary Mg_2Si, accumulated at centers of {111} planes [13]. The forming of hopper-like Mg_2Si crystal is due to the difficulty of diffusion of aluminum atoms in the centers of {111} surfaces, so that the aluminum-rich blanket over these facets grow thicker with time increasing [1]. Therefore, the growth of {111} faces is restricted, which leads to hollows formation on {111} facets.

When the ultrasonic vibration is introduced into the melt, the effects of cavitation will become the main factors controlling the distribution and diffusion of alloy atoms instead of thermal convection. The reason is that the velocity of acoustic streaming with a maximum speed of 1.37 m/s, produced by ultrasonic vibration, is about $10-10^3$ times of that of the melt thermal convection, based on theoretical calculations [12]. Because of the strong influence of ultrasonic, primary Mg_2Si crystal becomes solid particle. The growth rates along ⟨111⟩ directions can be increased by ultrasonic via the way that the acoustic streaming mixes Al and impurities atoms accumulated at the centers of {111} planes with the melt far from the vicinity of the growing primary Mg_2Si particle, resulting in the suppression of growing of the aluminum-rich blanket over {111} facets. So no hole can be observed on {111} facets of primary Mg_2Si with ultrasonic vibration.

When high energy ultrasonic is introduced into liquid medium, acoustic cavitation involves the formation, growth, pulsating, and collapsing of tiny bubbles under cyclic high-intensity ultrasonic waves. By the end of one cavitation cycle, the tiny bubbles implosively collapse, producing transient cavitation [15−17]. Cavitation has great influence on the growth of primary Mg_2Si. Edges of primary Mg_2Si particles are the most vulnerable position in the ultrasonic field. Those edges are scoured intensively by cavitation. So they become roundness during the growth procedure, as shown by the marked sign B in Fig. 3(c). This is in keeping with the round section characters of primary Mg_2Si in Fig. 2(d). It is worth mentioning that some depressions can be observed on {111} facets, as shown by the marked sign D in Figs. 3(c) and (d), which may be resulted from cavitation.

4 Conclusions

1) The mean grain size of the primary Mg_2Si particles was refined from 150 μm to 20 μm by high intensity ultrasonic.

2) The morphologies of primary Mg_2Si crystals are changed by ultrasonic vibration. The planar character of

the primary Mg_2Si with ultrasonic vibration is complete round section without holes. The three-dimensional morphology of primary Mg_2Si crystal is changed into solid particle with roundness edges and depressions on {111} facets by ultrasonic vibration.

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超声波对原位 Mg_2Si/Al 复合材料中初生 Mg_2Si 形态的影响

张家陶，赵宇光，徐晓峰，刘晓波

吉林大学 材料科学与工程学院，汽车材料教育部重点实验室，长春 130025

摘　要：采用光学显微镜和场发射扫描电镜，研究超声波对原位 Mg_2Si/Al 复合材料中初生 Mg_2Si 形态的影响。研究结果表明：超声波处理使初生 Mg_2Si 的晶粒尺寸从 150 μm 降低到 20 μm，初生 Mg_2Si 形态发生改变。在二维形貌中，未实施超声波振动处理的初生 Mg_2Si 晶粒生长为含有空腔的粗大颗粒，共晶组织生长于其中，相应的三维形态为含有漏斗状空腔的八面体和十四面体。超声波处理后的初生 Mg_2Si 晶粒变成细小、实心三维形态的颗粒，颗粒棱角已发生钝化效应。

关键词：超声波；Mg_2Si/Al 复合材料；Mg_2Si；形态；晶粒细化

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