



A novel method of coating ex-situ SiC particles with in-situ SiC interlayer in Al-Si-C alloy



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ABSTRACT

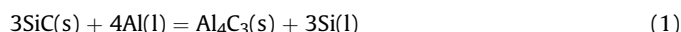
For SiC_p/Al composites, the stability of the interface between SiC particles (SiC_p) and matrix is crucial. A novel method, in-situ coating of ex-situ SiC_p, is developed in hypereutectic Al-Si alloys within this research. The microstructure, growth mechanism and strengthening behavior of the ex-situ SiC_p with in-situ SiC coating were studied. Microstructure observation shows that due to the formation of in-situ coating, the interfacial bonding of ex-situ SiC_p and matrix is improved. Further research reveals the epitaxial growth character of the in-situ SiC coating which has the same structure with ex-situ SiC_p. The ex-situ SiC_p with in-situ SiC coating is easily introduced into complex Al-Si alloys and the tensile strength of the alloy at both room temperature and 350 °C is improved remarkably.

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1. Introduction

Metal matrix composites (MMCs) are considered more suitable for aerospace, automotive and other structural applications than its monolithic alloy, owing to their enhanced mechanical, thermal and frictional properties [1,2]. SiC_p reinforced Al matrix composites are among the most promising MMCs, and have been widely used in electronic packaging and aerospace areas [3–6]. Because of the load transfer from Al matrix to SiC_p through the interface, the incorporation of SiC_p in Al matrix can significantly increase strength, lower the coefficient of thermal expansion, and improve wear resistance [7,8]. Therefore, in order to obtain the best combination of these properties, optimum interfacial bonding between SiC_p and Al matrix is required.

The interfacial bonding strength between SiC_p and Al depends on two factors. On one hand, interfacial reaction between SiC_p and Al, as shown in equation (1), is unavoidable in Al melt. The reaction product, Al₄C₃, is generally considered a brittle phase and is detrimental to the interfacial bonding between SiC_p and Al [9–11]. Hence, this reaction should be limited. On the other hand, the wettability between SiC_p and Al is poor and should be improved [12,13].



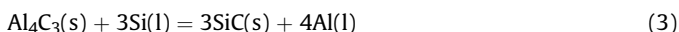
Traditionally, SiC_p (α -SiC with hexagonal structure in most cases) is externally added, and pre-treatment of the particles beforehand is necessary to balance the above two factors. Adding of alloying elements, such as magnesium and titanium, can improve the wettability between SiC_p and Al to some extent [14,15]. However, the most common pre-treatment method is to introduce an interlayer between SiC_p and Al matrix, so as to avoid the contact and reaction between them, and prevent the formation of brittle Al₄C₃. One effective interlayer is SiO₂, which can be generated by exposing SiC_p in oxidative atmosphere, like air, at high temperature or via sol-gel method [16–18]. Metal coating such as nickel or copper is also common, and it can be formed by electroless deposition [19–21]. Introduction of these interlayers can effectively enhance the interface bonding between SiC_p and Al matrix, but this method is costly and impurity elements may be added into composites.

In Al-Si system, the reaction in equation (1) can be depressed or reversed, owing to the existence of silicon. In order to avoid the formation of Al₄C₃, the least Si content corresponding to 1000 °C (the extreme temperature in melt process) is 12.8%, close to eutectic Si content in Al-Si alloy [22]. Based on this, in-situ synthesis of β -SiC_p (face-centered cubic structure, F-43 m) has been reported in Al-Si-C alloy by our group [23–25]. In this system, β -

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SiC_p is synthesized via gradual reaction by equations (2) and (3), and Al₄C₃ is the intermediate phase. Al element, less than 5 at%, is usually doped in in-situ β-SiC_p. The interface of in-situ β-SiC_p and Al matrix bonds well in the alloy.



Inspired by the above results, in-situ SiC coating on the surface of ex-situ SiC_p (hexagonal structure, *P63mc*) has been developed in this study. Core-shell structured SiC_p with curved surface were obtained via in-situ coating in Al-Si-C alloy, in which the Si content was high enough to depress the formation of Al₄C₃. Strong interfacial bonding between SiC_p and the matrix is obtained. The morphology and the growth mechanism of the in-situ coating were studied. The mechanical properties and the failure mechanisms of the ex-situ SiC_p with in-situ SiC coating (in-situ coating SiC_p) reinforced Al-Si matrix composites were investigated.

2. Experiment

Commercial green SiC_p powder (98.5 wt% purity), Al (99.7 wt% purity), Si (99.9 wt% purity, 75 μm, Sinopharm Chemical Reagent, China), and graphite (99.2 wt% purity, 1.3 μm, Huatai Graphite, China) were used in this research. The morphologies of SiC_p are shown in Fig. 1. Sharp corners and steps can be observed at the surface, as marked in Fig. 1b. SiC_p were cleaned by 5 vol% hydrofluoric acid to remove the impurities and increase the activity of their surface beforehand. The composition of the green samples was Al-20Si-3C-10SiC_p (wt%): 20% Si can guarantee that graphite is totally reacted and no Al₄C₃ is formed, while 3% C (graphite) and 10% SiC can guarantee that liquid Al-Si alloy can be hold by solid SiC at high temperature, and that the reacted samples can be introduced into other systems easily. SiC and graphite were mixed in blender mixer for 2 h. After that, they were mixed with Al and Si for another 2 h. The sufficient mixed blends were pressed into cylindrical mould and pressed at 250 MPa in cold isostatic press, with the size of φ60 mm × 50 mm. After that, the compacted samples were heated in vacuum resistance furnace, with vacuum degree higher than 10⁻³ Pa, at 750 °C for 0.5 h or 2 h.

To check the improvement of interfacial bonding between SiC and Al matrix treated by the novel method mentioned above, the Al-20Si-3C-10SiC_p alloy was introduced into an Al-12Si-4Cu-2Ni-1Mg (Al-Si-Cu) alloy at 800 °C in resistance furnace. Al, Si, Cu and Ni were melt at first, and Al-20Si-3C-10SiC_p alloy was added

secondly. The melt was then stirred for 3 min at 900 °C and after that, Mg was added. After refined by high purity argon, 0.5 wt% Al-3P was added to refine Si phase. Finally, the melt was cast into an iron mould at 800 °C. The mechanical properties of the cast samples at room temperature and 350 °C were detected and the strengthening behavior was studied.

Metallographic specimens were cut from the center of the samples, and prepared in standard routines. SiC_p were extracted from the samples by 2 vol% hydrochloric acid. The metallographic specimens and the extracted SiC_p were analyzed with X-ray diffraction (XRD, Rigaku D/max-rB, Cu Kα radiation) and field emission scanning electron microscopy (FESEM, Hitachi SU8010) equipped with an energy dispersive X-ray spectroscopy (EDS) detector. Transmission electron microscopy (TEM) samples were prepared by Precision Ion Polishing System (PIPS, Gatan Model 691) before being observed with TEM and high-resolution TEM (HRTEM, JEM-2100, Japan).

Tensile tests were conducted with 2 mm/min constant cross-head speed with extensometer. The samples tested at room temperature and 350 °C were prepared according to ASTM E8/E8M-15 and ASTM E21-2009 with 25 mm and 50 mm gauge length respectively. Surface and the cross section of the tensile fracture were observed with FESEM.

3. Results and discussion

3.1. Microstructure of SiC_p/Al-Si composites and ex-situ SiC_p

The microstructure of the designed composites is shown in Fig. 2. Besides α-Al and Si, two kinds of particles with different sizes can be observed, and both of them distribute uniformly in the matrix (Fig. 2a&b). XRD patterns of the composites (Fig. 3a) detect the appearance of α-SiC with hexagonal structure and β-SiC with fcc structure in the composites. The coarse particles shown in Fig. 2d are the ex-situ green α-SiC particles. The fine particles with sub-micron size and irregular spherical shape are in-situ synthesized. EDS analysis (Fig. 2c) show that the in-situ particles in Fig. 2b are mainly composed of C and Si with atomic ratio of nearly 1:1, together with nearly 5 at% of Al element. As mentioned in the background [23–25], and combined with the XRD patterns, the in-situ particles are β-SiC.

A noticeable structure, as shown in Fig. 2e, is the interlayer between ex-situ addition SiC_p and matrix. Due to the existence of the interlayer, two kinds of interfaces, i.e. the interface between ex-situ SiC and interlayer and the interface between interlayer and Al matrix, are formed.

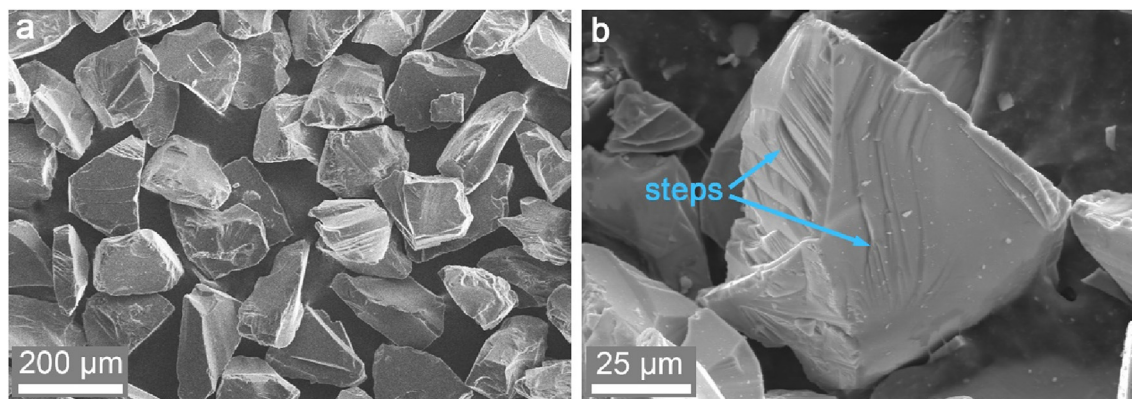


Fig. 1. (a) Shape and (b) surface morphology of raw green SiC_p. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

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