Effect of Powder Composition on the Preparation of SiCfoam/Al Co-continuous Phase Composites by In-situ Reactive Pressureless Infiltration

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Abstract. The SiC foam/Al co-continuous phase composite was prepared by in-situ reaction pressureless infiltration (ISRPI). The influence of powder ratio on the impregnation effect and the mechanical properties of the material was studied. The relationship between the mechanical properties and the interfacial reaction was discussed. The results show that the addition of Mg powder of 8% mass fraction increases the impregnation effect, and the formed magnesium spinel strengthens the bonding interface, which makes the compacting of the composite improve. At the same time, adding 2% mass fraction of Si in the infiltrated powder can inhibit the production of brittle material Al4C3 and improve the mechanical properties of the composites. The compressive strength of SiC foam/Al co-continuous phase composites with good interfacial bonding reaches 832MPa.

Key words: SiC/Al, co-continuous composite, in-situ reactive pressureless infiltration.

1. Introduction
SiCp/Al composite has excellent mechanical properties, such as high specific strength, good wear resistance[1] and so on, and also has low expansion, high thermal conductivity and other thermal properties [2]. It has shown broad application prospects in aerospace [3], automobile industry [4] and other applications. Because of the discontinuous [5] of ceramic particles and the non-wetting of ceramic and metal, it is difficult to distribute the glumes evenly in the metal matrix, which affects the improvement of its properties. So Breslin[6] et al. Put forward a new composite material called Co-continuous ceramic composites (C4 materials). The prominent feature of this material is that the matrix and the reinforced phase are three dimensional continuous mesh in the whole material, and interspersed with each other. The comprehensive strengthening effect is usually better than that of the SiCp/Al composite, Therefore, it has received extensive attention from the [7].

The preparation of C4 materials is usually pressure infiltration method [8] and pressureless infiltration method[9]. Pressure infiltration method has become the main preparation method because of its good performance and high efficiency. However, because of its high requirements for ceramic performs and the expensive equipment, large-scale production is rather difficult. The pressureless infiltration method has the characteristics of simple equipment, convenient operation, low cost and easy industrialization. However, this method has the problems of long infiltration distance and low
efficiency. To this end, Jiang Guo-jian [10] proposes a new pressureless infiltration method, called In Situ Reactive Pressureless Infiltration (ISRPI). The method uses metal or alloy powder instead of metal alloy ingots to fill the pores of the three-dimensional mesh porous ceramics by the powder and make the metal contact with the ceramic fully. Compared with the traditional pressureless infiltration method, the in-situ reaction pressureless infiltration method has better infiltration efficiency and higher efficiency.

In this paper, the SiC\textsubscript{foam}/Al Co-continuous ceramic composites was prepared by in-situ reaction pressureless infiltration. The effect of infiltration powder composition on its infiltration and mechanical properties is mainly studied, and its infiltration mechanism is explored.

2. Experiment
The experimental materials for the preparation of SiC\textsubscript{foam}/Al co-continuous composites are aluminum powder (purity 99%, 200 mesh), magnesium powder (purity 99%, 200 mesh), silica powder (purity 99%, 200 mesh) and SiC foam ceramics (volume fraction 20%, average pore diameter about 1.8mm). First, the SiC foam ceramics were cut into 20mm\texttimes12mm. Then, the SiC foam ceramics were oxidized for 8h at 1000 °C. The metal powder is filled into the SiC foam ceramics and then sintered at 900 °C for 3h in Nitrogen atmosphere. The interface phase of the composite was characterized by XRD, and the reaction interface was characterized by metallographic microanalysis, and the relationship between the interface and the mechanical properties were analyzed.

3. Results and discussion
Fig. 1 is a photo of SiC\textsubscript{foam}/Al composite materials with different Mg powder content (2% mass Si powder) by ISRPI. It can be seen from the photo that the composite material is divided into two parts, the upper part is metal alloy and the lower part is SiC\textsubscript{foam}/Al composite. They are cylindrical, but their center do not coincide.

![Fig. 1 Composite materials with different Mg contents: 1%, 3%, 5%, 7%, 8%, 9%, 10% from left to right Mg content](image)

Compared with the photos of SiC\textsubscript{foam}/Al composites by ISRPI process with different Mg powder content. It was found that when the content of the Mg powder was less than 5%, the more metal remains in the upper part, the lower part of the ceramics is porous, which indicates that the amount of metal infiltrated in ceramics is less. With the increase of Mg content, the foam ceramics are surrounded by metal, which is formed by cooling molten metal. Thus, adding different content of Mg, the degree of infiltration of materials is quite different. When the content of Mg is less than 5%, most of the metal is above the ceramics.

The mechanism of impregnation has been thoroughly studied for traditional pressureless infiltration [11]. The process is generally as follows:

$$3\text{Mg} + \text{N}_2 = \text{Mg}_3\text{N}_2$$  \hspace{1cm} (1)
When the temperature reaches the melting temperature of Mg in the Al alloy ingot above the SiC ceramic skeleton, the Mg in the aluminum alloy ingot is melted and volatilized. In the process of volatilization, the Mg has a chemical reaction with nitrogen that is adsorbed on the surface of the SiC foam ceramic hole. A partial vacuum negative pressure is formed inside the foam ceramic, and a Mg₃N₂ coating is also formed on the mesh hole of the porous SiC ceramic skeleton. When Al solution contacts with Mg₃N₂, chemical reaction takes place to form AlN and Mg. Because the reaction is exothermic, it increases the temperature of the local Al liquid, and the wettability of Al solution on the surface of SiC skeleton ribs is promoted. Coupled with the local negative pressure formed by the chemical reaction in foam ceramic, the infiltration of the SiC foam ceramics is produced by the aluminum alloy solution under the condition of pressure difference and good wetting [12]. The ISRPI fills the metal powder to be infiltrated in the pore of SiC foam ceramic, although it is different from the traditional pressureless infiltration, but the mechanism of infiltration is based on the same mechanism of infiltration. The above mechanism is also applicable.

When the content of Mg is less, the local vacuum pressure is smaller because of the chemical reaction between Mg powder and N₂, and the wettability of Al and SiC is not good, which causes the Al liquid to flow from the SiC foam ceramics. With the increase of Mg content, the reaction between Mg and N₂ was continuous, the surface of SiC foam ceramic skeleton gradually forms a Mg₃N₂ coating which promotes wetting. At the same time, Mg can react with SiO₂ on the surface of SiC ceramics and Al₂O₃ in molten Al. The equation[13] is as follows:

\[
3\text{Mg} + 4\text{Al}_2\text{O}_3 = 3\text{MgAl}_2\text{O}_4 + 2\text{Al} 
\]

\[
\text{Mg} + \text{SiO}_2 + \text{Al} = \text{MgAl}_2\text{O}_4 + 2\text{Si} 
\]

The volatilization of Mg can destroy the oxide film on the surface of molten aluminum at the infiltration temperature, which makes the wettability of metal melt and SiC ceramic increase. On the other hand, the Mg steam pressure of the molten state is high, especially when the temperature rises, the vapor pressure increases rapidly. The spinel MgAl₂O₄, produced by the reaction, reduces the surface tension of the metal, making the molten metal spread smoothly to the ceramic surface.

Fig.2 is a XRD porous SiC ceramics before and after infiltration. The phase composition of the porous SiC ceramics before and after infiltration is compared, it can be seen that after infiltrated, MgAl₂O₄ is found, which proves the reaction of the above interface.

Fig.3 shows the relation between the volume density of the composite and the content of Mg. It can be seen from the figure that the density of the composite increases with the increase of the amount of Mg added. When its addition reaches 8%, the density is 2.784 g/cm³. Further increasing Mg content, the density of the composites decreased, which may be related to the high content of residual Mg.
It is known from the experimental results that it is not a simple physical process to reduce the surface tension of the molten Al in order to improve the molten Al wettability on the surface of SiC ceramic, but because of its chemical reaction with the SiO₂ on the SiC skeleton, which changes the combination of the metal and the ceramics.

Fig. 2 Relationship between bulk density and Mg amount of composite

Fig. 3 Relationship between bulk density and Mg amount of composite

Fig. 4 is the compressive strength of composite materials with different Mg powder. It shows that with the increase of Mg content, the bonding strength of molten metal and ceramics increases gradually. When the content of Mg is higher than 8% mass fraction, the compressive strength of the composite decreases with the increase of Mg content.

In the process of preparing SiC foam/Al composite by, when the temperature is higher than 933K, the bonding interface between ceramic and metal will generate Al₄C₃[14]. Al₄C₃ is a brittle material that reacts with water. Therefore, the formation of Al₄C₃ is an adverse reaction which seriously affects the properties of composite materials.

$$4\text{Al} + 3\text{SiC} = \text{Al}_4\text{C}_3 + 3\text{Si}$$ (5)
When the degree of reaction increases, the strength of SiC ceramic skeleton will be greatly reduced, and it is easy to deliquesce and extremely sensitive to corrosion environment [15]. The reaction (5) is controlled by the dissolution and diffusion mechanism. The process [16] is as follows:

SiC dissolves, C and Si melt into the Al solution; C is saturated, SiC continues to dissolve, and the interface generates Al₄C₃, and grows in Al liquid in the form of thin rod or lump.

\[
\ln A_{Al_{4}C_{3}} + 3 \ln A_{Si} = -\Delta G^{\theta} / RT
\]

It can be seen from the equation that the activity of temperature T and Si is the main factor affecting the formation of Al₄C₃. From the XRD of the composite material after infiltration of Fig.3, there is no fragile substance Al₄C₃. Therefore, the incorporation of Si into the infiltration powder can effectively inhibit the interfacial reaction.

At the same time, the mechanical properties of the materials were characterized, and the amount of Si powder was discussed. Fig.5 shows the compressive strength of composites with different amounts of Si powder. As the content of Si increases, the compressive strength of the composite increases gradually. When the content of Si exceeds 2%, the compressive strength of the composite tends to be slow.
Fig. 5 Compressive strength of the composites with different Si amount

Fig. 6 is a metallographic photo of SiC\textsubscript{foam}/Al co-continuous composite. The white part of the composite is aluminum and the black part is SiC. It can be seen from the diagram that the interface between metal and ceramic is compact, and dense SiC\textsubscript{foam}/Al co-continuous composite are prepared.

Fig. 6 Metallurgical structure of SiC\textsubscript{foam}/Al co-continuous composite

4. Conclusion
In this paper, the effect of powder ratio on the infiltration and mechanical properties of SiC\textsubscript{foam}/Al co-continuous composite is studied by the new technology of ISRPI. The main results are as follows:

(1) In the process of ISRPI, the wettability of metal and SiC ceramics can be improved by adding Mg in the metal powder, and Si powder is added to control the interfacial reaction between the ceramic and the metal.

(2) The incorporation amount of Mg affects infiltration effect. When the content is less than 5%, the compressive strength of the composites is low, and the addition of 8% is the most suitable. The compressive strength of the composites reaches 832MPa.

(3) The addition of 2% Si powder in the metal powder can effectively inhibit the production of Al\textsubscript{4}C\textsubscript{3} and improves the mechanical strength of SiC\textsubscript{foam}/Al co-continuous composite.

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