The influencing mechanism of modification layer on the performance of SiC$_{3D}$/Al multi-function gradient composite

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Abstract. SiC skeleton surface was oxidized in this paper. Vacuum-pressure infiltration method is used to prepare SiC$_{3D}$/Al composite. The effects of the thickness of the interface modification layer were investigated. The results showed that the thickness of SiO$_2$ layer increases with the prolonged time of the skeleton oxidation. The brittle phase basically disappeared at the interface of the composite with 9 hours pre-oxidized, which lead to the high interface bonding strength. As a result, fracture morphology of the oxidized composite is mainly composed with plastic toughening of pure aluminum. Therefore, the static compressive strength of the composite raises up to 1165.2Mpa.

Keywords: SiC$_{3D}$/Al composite; thickness of the interface modification layer; fracture mechanism;

1. Introduction

Due to the unique network interpenetrating structure of the SiC$_{3D}$/Al composite, with high specific strength of the SiC as well as plastic deformable ability of aluminum, the SiC$_{3D}$/Al composite is widely used in many fields, such as electronics, mechanics, aviation and etc$^{[1]}$.

The best way to produce completely infiltrated SiC$_{3D}$/Al composite is vacuum pressure infiltration, which fulfilled with most of standards to create shape-perfect, structure compact composite$^{[2]}$. Therefore, this method was used to prepare the composite in this paper.

During the special artistry in producing SiC$_{3D}$/Al with vacuum pressure infiltration, the SiC structure and Al matrix experienced a long directly contacting period under high temperature and high pressure. This lead to the formation of fragile phase along interfaces$^{[3-5]}$. Therefore, the interface cannot efficiently conduct and distribute stress with applied load on the composite. Consequently, the composite property will be discounted. The interface bonding is a key factor to elevate performance of this composite material. One of the solutions is to reconstruct the outer shell of the SiC structure by oxidization, creating a compact oxygen layer of SiO$_2$, to detach the SiC from Al during infiltration$^{[5,6]}$. However, if the thickness of SiO$_2$ layer is insufficient to block the direct contact of Al from SiC, Al$_4$C$_3$ will formed. So, the thickness of the SiO$_2$ layer is very important to the vacuum pressure infiltration. In this paper, a series of SiC$_{3D}$/Al composites with different SiC pre-oxidation processes were investigated and the performance-microstructure relationship was discussed.

2. The experiment procedure and method

2.1. Experimental materials

The three-dimensional connected porous SiC with the density of 2.5g/cm$^3$, relative porosity of 21.88% was used as the ceramic phase of the experimental composite, while industrial pure aluminum was used as the metal phase during the preparation.
The ZYQ250/400-2.1000 model vacuum pressure infiltration furnace was used to produce SiC$_3$D/Al composite material at temperature between 750 ~ 800 °C, and infiltration pressure of 5-6.5 Mpa. The infiltration sketch was shown in Fig.2.

![Image 1](https://example.com/image1.png)  ![Image 2](https://example.com/image2.png)

**Figure 1** Skeleton microstructure.  **Figure 2** Infiltration sketch.

### 2.2. SiC skeleton surface oxidation

To estimate the effects of the thickness of SiO$_2$ layer on the properties of the composites, this experiment was carried out under 1200°C in an oxygen sufficient environment where the SiC surface oxidation would take place. The oxidation time under 1200°C were 3h, 6h, 9h respectively.

### 2.3. Composite performance and organization structure characterization.

The microstructural morphology of the polished composites was observed using ice emission scanning electron microscope (S4800). The Rigaku X-ray diffraction analyzer was used to analyze the phase composition of the reaction product (Cu target, K, Ni filter, scanning range 15 ~ 90; tube voltage 40 kv, tube current 50 mA; Slit size DS = 1, RD = 0.15, SS = 1, scanning speed 5 / min).

The compression properties of the composite were determined by WDW - E100D tester, with the strain rate controlled in the range of $10^{-4}$ s$^{-1}$ ~ $10^{-3}$ s$^{-1}$.

The composite was etched for 30s using solution of HNO$_3$(33 vol.%) [5] with corrosion voltage of 20V, corrosion current of 1.5A, so as to observe the surface morphology and phase composition.

### 3. Results and analysis

#### 3.1. Microstructure morphologies and mechanical properties

Figure 3 shows the microstructure of composites with skeletons treated by different pre-oxidation procedures. The samples shown in figure 3a, 3b, 3c, 3d were pre-oxidized for 0 hr, 3 hrs, 6 hrs and 9hrs, respectively. It is evidently in figure 3 that the integrities of all the samples were very good. The density of these composite are above 99%.

In order to study the influence of pre-oxidation, the static compressive strength of composites was tested in this paper. The results are shown in Table 1.
The compressive strengths of the composite with oxidized skeleton were significantly higher than those with skeleton unprocessed. Besides, the compressive strength improved with the increasing oxidation time and reached its highest level when the oxidation time prolonged to 9 hours.

Many investigations have been done for the SiO$_2$ layer on the skeleton surface, in which the existence of SiO$_2$ layer is considered to suppress the formation of Al$_3$C$_3$ and increase the wettability between SiC skeleton and Al matrix$^{[7]}$. But the formation of Al$_3$C$_3$ could not be completely prevented if the thickness of SiO$_2$ layer is insufficient. Therefore, the SiO$_2$ layer thickness plays an important role in influencing of the interface reaction during the infiltration process.

3.2 The role of the oxide layer thickness

The thickness of SiO$_2$ layer increases with the prolonged time of the skeleton oxidation. As analyzed before, the thickness of the layer is a sensitive factor that influences the property of the composite. Hence, the thickness of the layer should be quantitatively analyzed first. In this paper the experience formula$^{[8]}$ is used to calculate the thickness of the SiO$_2$ layer.

$$h = \frac{100 \cdot \Delta W}{S \cdot (M_{SiO_2} - M_{SiC}) \cdot \rho_{SiC}}$$

where h is the thickness of the oxidized layer, S represents the specific surface of SiC skeleton (cm$^2$/g), $\rho_{SiC}$ represents the SiC density (3.2g/cm$^3$); $\Delta W$ the weight gain (%), being $M_{SiC}$ and $M_{SiO_2}$ the molecular masses of SiC and SiO$_2$, respectively.

Table 1 The compressive strength of composites with un-oxidized and oxidized skeleton.

| Sample No. | un-oxidized | 3hrs oxidation | 6hrs oxidation | 9hrs oxidation |
|------------|-------------|----------------|----------------|---------------|
| compressive strength | 300.3 | 414.0 | 1096.1 | 1074.0 |
| average | 365.7 | 737.6 | 401.6 | 1119.8 |
|         | 300.5 | 593.1 | 433.2 | 1301.9 |
|         | 322.5 | 505.8 | 643.6 | 1165.2 |

Figure 3 Microstructure of composites.
Table 2 The thickness of SiO$_2$ layer

| Sample No. | 3hrs oxidation | 6hrs oxidation | 9hrs oxidation |
|------------|----------------|----------------|----------------|
| thickness/nm | 121.1          | 179.6          | 223.4          |

![Image](https://example.com/table2.png)

Figure 4 Micro morphology of SiC/Al composite with oxidized skeleton.

Figure 4 shows the microstructural morphology of the skeletons surfaces which were obtained from the electrolytic solution etched composites sample. Al phase was removed in etching process. The hexagonal crystals attached on the surface of skeleton in Figure 4.(a) is analyzed by EDS, the result is shown in figure 5.

![Image](https://example.com/eds.png)

Figure 5 The EDS analysis of SiC skeleton.

It can be concluded from the EDS results that the hexagonal substance in figure 4a is Al$_4$C$_3$. The XRD results for all the skeleton surface are shown in figure 6, which further demonstrated the existence of Al$_4$C$_3$. Thus, the interface reaction product is Al$_4$C$_3$. It can be concluded from the intensities of the peaks that amounts of Al$_4$C$_3$ in the skeletons decreased with the pre-oxidation time increasing, which is also accordance with the morphologies shown in figure 4. As shown in figure 4a, numerous brittle hexagonal crystals generated on the interface of the composite with 3 hours of pre-oxidized skeleton. There was a significant reduction of brittle crystals in the composite with 6 hours pre-oxidized skeleton, which is exhibited in Figure 4b. Al$_4$C$_3$ crystal was barely generated in the composite with 9 hours pre-oxidized skeletons.
The existence of hexagonally stacked Al₄C₃ would cause the irregular arrangement of the atoms at the interface and also induced the stress concentration in the deformation area, which finally lead to the growth of the cracks and influenced the mechanical property of the composite. Based on the results above, it can be concluded that the existence of SiO₂ layer could suppress the formation of Al₄C₃ and elevate the mechanical property of composite.

3.3. Fracture morphology of the composite

Figure 7 shows the fracture morphology of composites. The relatively dark and regular phase is SiC, while the light and rugged phase is Al or interface reaction product. The figure shows that the
primarily fracture mode of the composite without oxidation treatment is interfacial brittle cracking. The brittle phase stacking at the interface leads to the interfacial premature cracks, so the plastic deformation capacity of Al phase cannot be well performed. Based on the analysis above, the mechanical properties of the composites was degraded. From the labeled zone in figure 7(b), (c), (d), the interface of the composite bonded closely and the fractured mechanism of the composites pre-oxidized was Al phase ductile tearing. The content of the brittle phase decreased due to the increasing of oxidation time. After 9 hours of pre-oxidation, the brittle phase disappeared essentially, which was in accordance with the analysis results mentioned before. There was obviously dimple in the Al phase surface. Pure Al played an important role in improving the mechanical properties of composite.

4. Conclusions

The thickness of SiO$_2$ layer increased with the prolonged time of the skeleton oxidation. The brittle phase basically disappeared at the interface of the composite with 9 hours pre-oxidized skeleton, which lead to the high interface bonding strength and plastic toughening of pure aluminum, as a result, the static compressive strength of the composites raised up to 1165.2Mpa.

The fracture morphology of the unprocessed composite is mainly constituted with brittle cracking in the interface, while the fracture morphology of the oxidized composite is mainly composed with plastic toughening of pure aluminium. Hence, the mechanical property of composite improves.

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