

# Fabrication and oxidation resistance of mullite/yttrium silicate multilayer coatings on C/SiC composites

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Received: May 16, 2017; Revised: July 18, 2017; Accepted: September 12, 2017

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**Abstract:** A tri-layer coating of mullite/ $Y_2Si_2O_7$ /(70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$ ) was prepared on carbon fiber reinforced silicon carbide (C/SiC) composite substrate through dip-coating route for the sake of improving oxidation resistance of C/SiC composites. An  $Al_2O_3$ - $SiO_2$  sol with high solid content was selected as raw material for mullite, and a slurry of  $Y_2O_3$  powder filled silicone resin was used to synthesize yttrium silicate. The microstructure, phase composition, and oxidation resistance of the coating were investigated. The as-fabricated coating shows high density and favorable bonding to C/SiC substrate. After oxidation at 1400 and 1500 °C for 30 min under static air, the flexural strengths of coated C/SiC composite were both increased by ~30%. The desirable thermal stability and the further densification are responsible for excellent oxidation resistance. With the additional help of compatible thermal expansion coefficients among substrate and sub-layers in coating, the coated composite retained 111.2% of original flexural strength after 12 times of thermal shock in air from 1400 °C to room temperature. The carbothermal reaction at 1600 °C between free carbon in C/SiC substrate and rich  $SiO_2$  in mullite resulted in severe frothing and desquamation of coating and obvious degradation in oxidation resistance.

**Keywords:** oxidation resistance; thermal shock resistance; coatings; mullite; yttrium silicate; C/SiC composites

## 1 Introduction

Carbon fiber reinforced silicon carbide (C/SiC) composites have been well developed as a desirable candidate for thermo structural and tribological applications for about thirty years [1–3]. For applications in air, anti-oxidation should be paid much attention due to the inevitable formation of pores and microcracks. The pores and microcracks resulting from the intrinsic characteristics of fabrication technologies

and the mismatch of thermal expansion coefficient between fiber and matrix [4,5] make it easy for oxygen to diffuse into C/SiC composites and then oxidize carbon fibers and free carbon in matrix [6,7]. So far, several methods have been explored to protect C/SiC composites from oxidation in which external coating is demonstrated to be the most effective one [8,9].

Yttrium silicate ( $Y_2Si_2O_7$  and  $Y_2SiO_5$  phases), which possesses advantages such as high melt point, low oxygen permeability, favorable thermal and chemical stability, low modulus, and matchable thermal expansion coefficient with SiC, has been studied extensively as external anti-oxidation coating for C/C and C/SiC composites [10–13]. Some measures

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including plasma spray [10], electrophoretic deposition [11], sol–gel [12], slurry brushing, and sintering [13] have been employed to fabricate yttrium silicate coating. Recently, a new route to synthesize yttrium silicate from the pyrolysis of  $Y_2O_3$  powder filled silicone resin has been reported [14,15]. This route is a promising method because of good adhesive and covering ability of silicone resin, low process temperature, and relatively low cost [16,17]. Dense  $Y_2Si_2O_7$  coating was fabricated on SiC foam by pyrolysis in air of silicone resin containing  $Y_2O_3$  powders [14], but the oxidation protection for C/SiC composites was not investigated. The  $Y_2Si_2O_7$  coating in the study of Liu *et al.* [15] displayed good oxidation protection for C/SiC composites at 1400 °C. However, the residual free carbon and pores in coating due to the pyrolysis of silicone resin in inert atmosphere would be detrimental to oxidation resistance at higher temperature. In our previous work [18],  $Y_2Si_2O_7$  coating was fabricated directly on C/SiC composite substrate by dip-coating from  $Y_2O_3$  powder filled silicone resin and provided excellent oxidation resistance at 1400–1600 °C. At the same time, it was observed that C/SiC composites were damaged to a certain extent by the conversion of silicone resin to  $SiO_2$  at 800 °C in air.

In fact, inner SiC coating was usually prepared on C/C or C/SiC composite substrate before yttrium silicate coating [10] in order to enhance bonding strength and prevent carbon diffusion. Nevertheless, oxygen would diffuse through cracks and pores in yttrium silicate coating and oxidize SiC coating during long-time oxidation, resulting in desquamation or split of external coating [19]. To avoid the above problem, inner SiC coating was replaced by mullite coating derived from  $Al_2O_3$ – $SiO_2$  sol [20], and mullite/ $Y_2Si_2O_7$  double-layer coating with desirable anti-oxidation performance and thermal shock resistance has been obtained [21]. However, some pores and microcracks were still formed due to the intrinsic characteristics of sol–gel and slurry method. Although a top layer of borosilicate glass could seal these pores and microcracks and improve oxidation resistance [22], its low melt point and high reactivity in water-vapor are disadvantageous to the thermal stability of coating.

It has been found that the yttrium silicate coating with gradient composition showed higher density and better oxidation resistance than single-layer yttrium silicate coating [10]. The layer of 70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$  not only had a thermal expansion coefficient close to those of C/SiC and  $Y_2Si_2O_7$  but also

showed desirable oxidation resistance [15,19]. Once the content of  $Y_2SiO_5$  was more than 30wt%, the thermal mismatch between  $Y_2Si_2O_7$  and 70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$  would be enlarged, very likely leading to delamination and desquamation. So, in order to improve oxidation resistance of mullite/ $Y_2Si_2O_7$  double-layer coating, a top layer of 70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$  was introduced in this paper. The fabrication and oxidation resistance of mullite/ $Y_2Si_2O_7$ /(70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$ ) tri-layer coating were investigated.

## 2 Experimental

### 2.1 Raw materials

The substrate was C/SiC composites with an apparent density of 1.92 g/cm<sup>3</sup> and an open porosity of 9.8%. The C/SiC substrates with a size of 70 mm × 5 mm × 4 mm were fabricated by polycarbosilane infiltration and pyrolysis in our own lab. The  $Al_2O_3$ – $SiO_2$  diphasic sol with a solid content of 22% and an  $Al_2O_3$ / $SiO_2$  mass ratio of 1:2 was selected as raw material for mullite coating. Commercially available flake silicone resin with a trademark of DC249 (Dow Corning Co. Ltd., USA) and  $Y_2O_3$  powders (Sinopharm Chemical Reagent Co. Ltd., Shanghai, China) with a diameter of 2–3 μm and a purity of 99.99% were used to synthesize yttrium silicate. According to the chemical compositions of  $Y_2Si_2O_7$  and 70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$  and the ceramic yield (~63%) of silicone resin in air, the mass ratios of  $Y_2O_3$  to silicone resin were set to 54.2:45.8 and 58.8:41.2, respectively. Silicone resin was dissolved in anhydrous ethanol, to which  $Y_2O_3$  powders were added. After stirred for 4 h, the slurries for yttrium silicate coatings were obtained.

### 2.2 Fabrication of coatings

C/SiC composite substrate was dipped in  $Al_2O_3$ – $SiO_2$  sol for 5 min, then pulled out with a constant speed and dried at 100 °C for 1 h. After the dipping–drying cycle was repeated 5 times, C/SiC composite substrate was heated at 1400 °C for 1 h under flowing high purity argon. The mullite coating was obtained by repeating 4 times of the process of dipping–drying (5 times) and heat treatment.

The preparation of  $Y_2Si_2O_7$  coating included three steps, namely, dip-coating, pre-pyrolysis, and heat treatment. In the first step, mullite-coated C/SiC

composite substrate was dipped in slurry for 5 min, then drawn out with a constant speed and dried at 80 °C for 1 h. After repeating 4 times of dip-coating process, C/SiC composite substrate was heated at 250 °C for 2 h to cure silicone resin, followed by pre-pyrolysis at 800 °C for 1 h in air to convert silicone resin into SiO<sub>2</sub>. The process consisting of dip-coating (4 times) and pre-pyrolysis was repeated for 3 times. Finally, the mullite/Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> coated C/SiC composites were obtained after heat treatment at 1400 °C for 1 h under flowing high purity argon. The fabrication of 70wt%Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>+30wt%Y<sub>2</sub>SiO<sub>5</sub> coating was almost the same as Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> coating, except the slurry.

### 2.3 Characterization of coatings

Static oxidation was performed in a muffle which was preset at 1400, 1500, and 1600 °C. The as-received coated C/SiC composites were placed on Al<sub>2</sub>O<sub>3</sub> bracket and oxidized for 30 min. Thermal shock in air from 1400 °C to room temperature was carried out 12 times. The muffle was heated to 1400 °C in advance. The as-received coated C/SiC composites were put into muffle and soaked at 1400 °C for 10 min, followed by rapid removal to ambient atmosphere. After the temperature lowered to room temperature, the C/SiC composites were put into muffle again.

The weight loss and the flexural strength retention after oxidation and thermal shock were recorded to characterize the oxidation and thermal shock resistance of tri-layer coating. The flexural strength was measured by a three-point bending method with a cross-head speed of 0.5 mm/min and a span/height ratio of 15. Scanning electron microscope (SEM), energy dispersive spectroscopy (EDS) (Quanta-200 EDAX), and X-ray diffraction (XRD) were employed to observe the changes of microstructure, element, and phase of coating after oxidation and thermal shock, respectively. XRD measurements were carried out on a diffractometer (D/max-2400, Rigaku) with Cu K $\alpha$  radiation. Data were digitally recorded during a continuous scan in the range of angle ( $2\theta$ ) from 10° to 80° with a scanning rate of 4 (°)/min.

## 3 Results and discussion

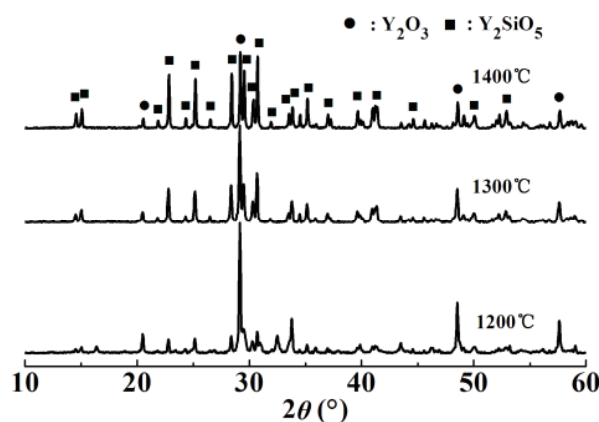
### 3.1 Fabrication and microstructure of tri-layer coating

The synthesis of mullite [20] and Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> [18] has been

studied in our previous studies. For the synthesis of Y<sub>2</sub>SiO<sub>5</sub> phase, the relative mass ratio of Y<sub>2</sub>O<sub>3</sub>/silicone resin in slurry was 70.3/29.7. The slurry was dried, cured, and pre-pyrolyzed in air, followed by heating in high purity Ar at 1200–1400 °C. In the same way, there are only Y<sub>2</sub>O<sub>3</sub> diffraction peaks after pre-pyrolysis at 800 °C in air, as shown in Ref. [18]. Figure 1 shows XRD patterns of the production after heat treatment.

The emergence of obvious diffraction peaks of Y<sub>2</sub>SiO<sub>5</sub> phase is observed at 1200 °C, indicating that the reaction between Y<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> takes place at this temperature. The intensity of Y<sub>2</sub>SiO<sub>5</sub> diffraction peaks is gradually enhanced by increasing temperature, accompanied by the gradual decline of Y<sub>2</sub>O<sub>3</sub> peaks. At 1400 °C, most of peaks can be assigned to Y<sub>2</sub>SiO<sub>5</sub> phase, implying that the reaction between Y<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub> is basically finished. The completion of Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> synthesis at 1400 °C has also been confirmed in previous work [18,21]. Moreover, the formation of Y<sub>2</sub>SiO<sub>5</sub> phase was demonstrated to be prior to Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> phase [14]. However, diffraction peaks of Y<sub>2</sub>O<sub>3</sub> can be found in the spectrum of 1400 °C. This may be explained by two reasons. One is a slight deviation of Y<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> mass ratio from the stoichiometric composition of Y<sub>2</sub>SiO<sub>5</sub> due to the fluctuation of ceramic yield of silicone resin in air. The other is that mixture uniformity is not good enough.

Figure 2 shows the XRD spectrum of 70wt%Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>+30wt%Y<sub>2</sub>SiO<sub>5</sub> sub-layer in as-received coating. It is evident that Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> and Y<sub>2</sub>SiO<sub>5</sub> phases are well developed after heat treatment at 1400 °C. At the same time, it is noted that the diffraction peaks of Y<sub>2</sub>SiO<sub>5</sub> phase are in the majority, which is not corresponding to the molar fraction (34.1%) of Y<sub>2</sub>SiO<sub>5</sub> phase. On one hand, prior formation of Y<sub>2</sub>SiO<sub>5</sub> has been affirmed [14],



**Fig. 1** XRD patterns of Y<sub>2</sub>O<sub>3</sub> powder filled silicone resin after heat treatment at various temperatures.

and higher temperature (>1400 °C) is necessary for the conversion from  $Y_2SiO_5$  to  $Y_2Si_2O_7$  because  $SiO_2$  in 70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$  is not enough as compared with that in  $Y_2Si_2O_7$ . On the other hand, the positions of  $Y_2Si_2O_7$  and  $Y_2SiO_5$  peaks are very close to each other, even the same [15]. Accordingly, the  $Y_2Si_2O_7$  peaks are likely to be enshrouded by those of  $Y_2SiO_5$  phase.

SEM appearance and EDS of as-received tri-layer coating are displayed in Fig. 3. It is clear from Fig. 3(a) that there are several microcracks besides some pores in the top layer of 70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$ . The microcracks were not obvious in mullite/ $Y_2Si_2O_7$  coating [21]. Thus, the microcracks result from the thermal expansion mismatch between  $Y_2Si_2O_7$  and 70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$  since the thermal expansion coefficient of  $Y_2SiO_5$  is much higher than that of  $Y_2Si_2O_7$  [15,23,24]. Moreover, Fig. 2 indicates that the content of  $Y_2SiO_5$  in top layer is more than 30wt%. As shown in Fig. 3(b), tri-layer coating shows tight bonding to C/SiC substrate and indistinct interfaces among sub-layers. The thickness of tri-layer coating is estimated to be 200–250 μm from cross-section. The tight bonding can be ascribed to the viscous flow effect of rich  $SiO_2$  in mullite [20] and the thermal expansion matching between substrate and mullite. The incompletely dense mullite coating [20],  $Y_2Si_2O_7$  coating [18,21], and the viscous flow effect of amorphous  $SiO_2$  derived from silicone resin are responsible for the indistinct interfaces. In addition, the oxidation damage to C/SiC substrate from pyrolysis of silicone resin in air [18] is not observed in this study because of the protection of mullite coating, which has been confirmed in previous work [21]. Theoretical element compositions of the top layer of 70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$  are 63.32 at% O, 16.63

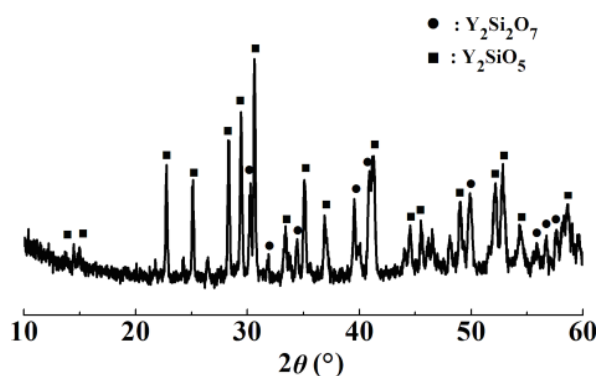


Fig. 2 XRD pattern of 70wt% $Y_2Si_2O_7$ +30wt% $Y_2SiO_5$  sub-layer in as-received coating.

at% Si, and 20.05 at% Y, and the EDS result of selected point A in Fig. 3(a) is shown in Fig. 3(c). In consideration of the semi-quantitative characteristic of EDS analysis and the incompletely uniform distribution of  $Y_2O_3$  powders in silicone resin, the two results are well coincident.

### 3. 2 Oxidation resistance of tri-layer coating

After oxidation at 1400, 1500, and 1600 °C for 30 min, the weight change and flexural strength retention of coated C/SiC composites are showed in Table 1. It is noted that the coated composites exhibit tiny weight gain after oxidation at 1400 and 1500 °C. Theoretically speaking, the weight of tri-layer coating should not be increased during oxidation. It is very likely that the tiny weight gain is due to the slight cohesion of  $Al_2O_3$  bracket to coating. In the work of Huang *et al.* [25], it

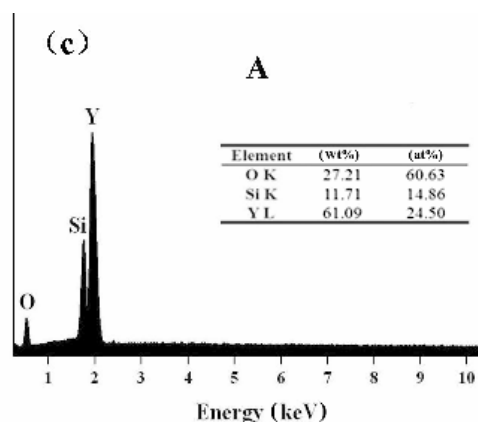
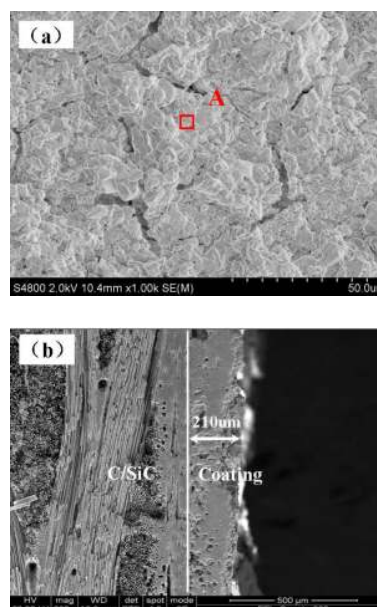
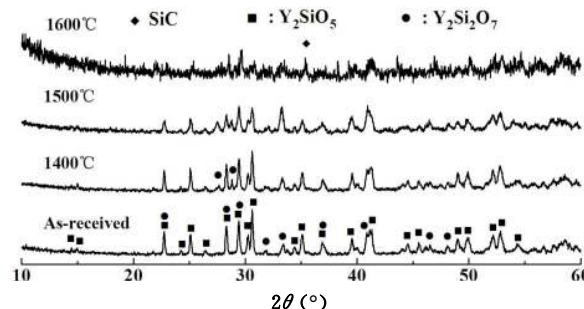


Fig. 3 SEM photos of (a) surface and (b) cross-section, and (c) EDS result of as-received coating.

was confirmed that the reaction between yttrium silicate and  $\text{Al}_2\text{O}_3$  bracket took place at  $1500\text{ }^\circ\text{C}$ . Our previous study [18] has validated this phenomenon. As for the weight loss of 18.66% at  $1600\text{ }^\circ\text{C}$ , it is not the fact and will be explained later. The flexural strength retention suggests that the tri-layer coating provides desirable oxidation protection for C/SiC composites.

Figure 4 presents load-displacement curves of coated C/SiC composites before and after oxidation. As shown, all the coated composites illustrate non-catastrophic fracture behavior with a maximum displacement of  $\sim 2.0\text{ mm}$  at invalidation point. The fracture surfaces of coated composites, which are similar to those of mullite/ $\text{Y}_2\text{Si}_2\text{O}_7$  coating [21] and not presented in this paper, show extensive fiber pull-out and long pull-out length before and after oxidation. In combination with the data in Table 1, it is indicated that tri-layer coating is a desirable candidate of anti-oxidation for C/SiC composites.

To elucidate the anti-oxidation mechanism of tri-layer coating, XRD patterns and surface morphology after oxidation were recorded and presented in Figs. 5 and 6, respectively. In Fig. 5, the spectrum at  $1400\text{ }^\circ\text{C}$  is almost the same as that of as-received coating except the two faint peaks at  $\sim 27.5^\circ$  and  $\sim 28.8^\circ$ . The two faint peaks are assigned to  $\text{Y}_2\text{Si}_2\text{O}_7$  phase. After oxidation at



**Fig. 5** XRD patterns of tri-layer coating before and after oxidation.

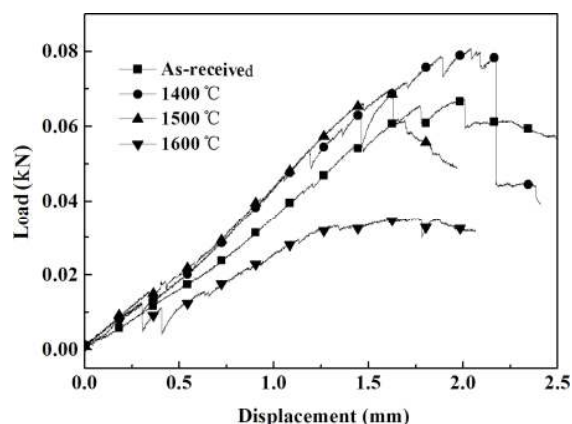
$1500\text{ }^\circ\text{C}$ , the peaks at  $\sim 27.5^\circ$  and  $\sim 33.3^\circ$ , which belong to  $\text{Y}_2\text{Si}_2\text{O}_7$  phase, are obviously strengthened. At the same time, the peaks at  $\sim 25^\circ$ ,  $\sim 31^\circ$ , and  $\sim 35^\circ$ , which are assigned to  $\text{Y}_2\text{SiO}_5$  phase, are weakened. As mentioned above,  $\text{Y}_2\text{SiO}_5$  phase is predominant in the top layer of as-received coating (Fig. 2) although the nominal composition is 70wt% $\text{Y}_2\text{Si}_2\text{O}_7$ +30wt% $\text{Y}_2\text{SiO}_5$ . This implies that the synthesis of 70wt% $\text{Y}_2\text{Si}_2\text{O}_7$  was not completed at  $1400\text{ }^\circ\text{C}$  for 1 h. XRD results indicate that the transformation from  $\text{Y}_2\text{SiO}_5$  to  $\text{Y}_2\text{Si}_2\text{O}_7$  can be promoted by prolonging heat treatment time or increasing heat treatment temperature. So, the fabrication of coating should be performed at  $1400\text{ }^\circ\text{C}$  for a long time or at higher temperature if the complete synthesis of  $\text{Y}_2\text{Si}_2\text{O}_7$  phase is expected.

As shown in Fig. 6(a), the microcracks and pores in Fig. 3(a) disappear and a dense surface is observed after oxidation at  $1400\text{ }^\circ\text{C}$  for 30 min. At  $1500\text{ }^\circ\text{C}$ , further densification of coating surface is discovered. Owing to the transformation from  $\text{Y}_2\text{SiO}_5$  to  $\text{Y}_2\text{Si}_2\text{O}_7$  during oxidation, volume expansion takes place since the density of  $\text{Y}_2\text{SiO}_5$  ( $4.53\text{ g/cm}^3$ ) is higher than that of  $\text{Y}_2\text{Si}_2\text{O}_7$  ( $4.15\text{ g/cm}^3$ ) [14], and the thermal expansion mismatch between the top layer and the interlayer of  $\text{Y}_2\text{Si}_2\text{O}_7$  is reduced because of the lower thermal expansion coefficient of  $\text{Y}_2\text{Si}_2\text{O}_7$  as compared with  $\text{Y}_2\text{SiO}_5$  [15,23,24]. As a result, the microcracks and pores are healed. In addition, solid-state sintering of yttrium silicate is also responsible for the further densification of coating [18]. Profiting from the densification of coating, the diffusion of oxygen into composites is effectively impeded, and the resistance to crack creation and propagation is enhanced. Therefore, the flexural strengths of coated C/SiC composites are increased by  $\sim 30\%$  after oxidation at 1400 and  $1500\text{ }^\circ\text{C}$ .

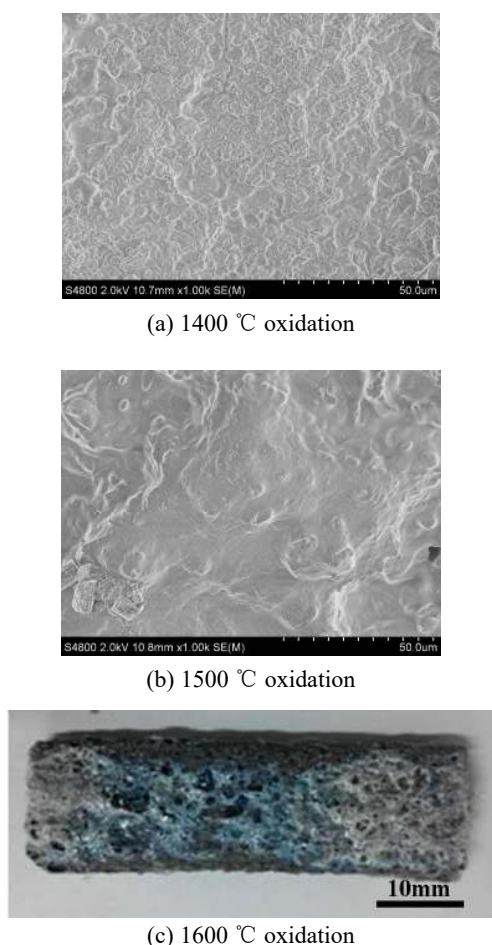
By comparing Fig. 2(b) in Ref. [21] with Fig. 3(b) in this paper, it is found that the pores in the cross section

**Table 1** Weight change and flexural strength of coated C/SiC composites after oxidation

Oxidation temperature	Room temperature	1400 °C	1500 °C	1600 °C
Weight change	—	0.58%	0.93%	-18.66%
Flexural strength (MPa)	163.4	208.8	212.6	138.7
Flexural strength retention	—	127.8%	130.1%	84.9%



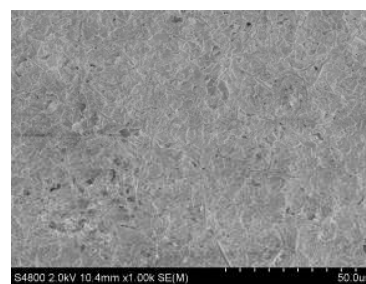
**Fig. 4** Load-displacement curves of coated C/SiC composites before and after oxidation.



**Fig. 6** SEM photos of tri-layer coating after oxidation.

of double-layer coating were not observed in tri-layer coating. This suggests that the tri-layer coating has higher density and validates the results of Huang *et al.* [10]. On one hand, the pores in interlayer could be filled by the slurry during the dip-coating of top layer. On the other hand, the top layer exerted compressive stress on the interlayer due to the higher thermal expansion coefficient of top layer. The compressive stress was helpful to the densification of interlayer. As mentioned above, the cracks in the top layer resulting from tensile stress would be healed during high temperature oxidation. By comparing Fig. 6(b) and Fig. 6(c) in Ref. [21] with Fig. 6(a) and Fig. 6(b) in this paper, it is obvious that the tri-layer coating exhibits more compacted surface after oxidation at 1400 and 1500 °C. As a result, the flexural strength retention ratios of the composites with tri-layer coating (127.8% at 1400 °C and 130.1% at 1500 °C) are much higher than those of the composites with double-layer coating (91.9% at 1400 °C and 102.4% at 1500 °C).

However, severe frothing and desquamation of



**Fig. 7** SEM photo of tri-layer coating after thermal shock.

coating is observed after oxidation at 1600 °C for 30 min, as shown in Fig. 6(c). This phenomenon can be attributed to the carbothermal reaction between rich SiO<sub>2</sub> in mullite sub-layer and free carbon in composite substrate [20]. Accordingly, the disappearance of some diffraction peaks of yttrium silicate and the emergence of SiC diffraction peak are observed in XRD pattern (Fig. 5). Porous C/SiC composite substrate is inevitably oxidized without the protection of tri-layer coating. Coating desquamation and subsequent oxidation of substrate result in the obvious weight loss of 18.66% and the remarkable degradation in flexural strength.

### 3.3 Thermal shock resistance of tri-layer coating

After 12 times of thermal shock from 1400 °C to room temperature in air, the coated C/SiC composites show a tiny weight gain of 0.36% and a flexural strength retention ratio of 111.2%. The load-displacement curve of coated C/SiC composite after thermal shock, which is very similar to that of 1400 °C in Fig. 4, illustrates typical non-catastrophic fracture behavior. Figure 7 shows SEM photo of coating surface after thermal shock. The XRD pattern of coating after thermal shock is not presented here since it is very similar to that of 1400 °C in Fig. 5.

During thermal shock test, the coated C/SiC composites endured not only sudden change of temperature but also oxidation at 1400 °C for 120 min. As mentioned above, the oxidation can promote the transformation of Y<sub>2</sub>SiO<sub>5</sub> to Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, leading to volume expansion and reduction in thermal expansion coefficient. So, the large-size cracks in as-received coating (Fig. 3(a)) are eliminated after thermal shock. With the increase of Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> phase content, the difference in thermal expansion coefficient between the top layer and the interlayer of Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> decreases. Thanks to the matched thermal expansion coefficients among substrate, mullite, and yttrium silicate, no macroscopic cracks and coating desquamation are

found after thermal shock. However, some micropores and microcracks are still observed in Fig. 7. As compared with Fig. 6(a), the densification is inferior. It is deemed that the micropores and microcracks are created by the local thermal stress derived from the repeating thermal shock and the incompletely uniform distribution of coating composition. Thus, the flexural strength retention ratio is lower than those after static oxidation at 1400 and 1500 °C.

#### 4 Conclusions

A tri-layer coating consisting of SiO<sub>2</sub>-rich mullite, Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, and 70wt%Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>+30wt%Y<sub>2</sub>SiO<sub>5</sub> has been fabricated on C/SiC composite substrate through dip-coating route, and provides excellent oxidation protection for C/SiC composite at 1400 and 1500 °C due to desirable thermal stability and further densification during high temperature oxidation. The carbothermal reaction between free carbon in substrate and rich SiO<sub>2</sub> in mullite at 1600 °C leads to severe frothing and desquamation of coating, degrading oxidation resistance.

After 12 times of thermal shock from 1400 °C to room temperature in air, the flexural strength of coated composites is increased by 11.2% due to the preferable physical and chemical compatibility among C/SiC substrate, mullite, and yttrium silicate as well as the further densification of coating.

#### Acknowledgements

The authors are grateful to the financial supports from the Aid Program for Science and Technology Innovative Research Team in Higher Educational Institutions of Hunan Province, and the Science Innovation Foundation of Shanghai Academy of Spaceflight Technology (No. SAST2015043).

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