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Effects of the single layer CVD SiC interphases on the mechanical properties of the SiCf/SiC composites fabricated by PIP process

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Abstract

Owing to the degradation of the mechanical properties of the SiC fiber reinforced SiC matrix (SiCf/SiC) composites with the pyrocarbon (PyC) and BN interphases under oxidation environment and neutron irradiation, single layer SiC interphases prepared by chemical vapor deposition (CVD) process were employed to substitute for them. Effects of the CVD SiC interphases on the mechanical properties and interfacial characteristics of the SiCf/SiC composites fabricated by precursor infiltration and pyrolysis (PIP) process were investigated. Compared with the as-received SiCf/SiC composites, the SiCf/SiC composites with the single layer CVD SiC interphases exhibit an obvious toughened fracture behavior, the flexural strength of which is about 4 times that of the as-received SiCf/SiC composites. From the microstructural analysis, it can be confirmed that the SiC interphases play a key part in protecting the fibers from damage during composite preparation and weakening interfacial bonding, which can provide high in situ fiber strength and appropriate interfacial bonding strength for the SiCf/SiC composites.

Keywords: C. Mechanical properties; Precursors; SiC interphases; SiCf/SiC composites

1. Introduction

SiCf/SiC composites generally exhibit excellent properties such as high strength at elevated temperature, oxidation resistance and microstructural stability under neutron irradiation [1,2]. Owing to these advantages, SiCf/SiC composites are known to be attractive materials for high-temperature structural applications, first wall and blanket components in fusion reactors [3–5]. For SiCf/SiC composites, the interphases between fibers and matrix are one of the key factors that determine the material mechanical properties, because the appropriate interphases can bring crack deflection, fiber pullout and interfacial debonding, which can provide excellent mechanical properties for SiCf/SiC composites [6,7]. Recently, it has been recognized that pyrocarbon (PyC) and BN with layered crystal structures are the most commonly used and effective interphase materials for improving the mechanical properties of the SiCf/SiC composites [8]. However, for fusion reactor components, PyC and BN interphases are not favorable because of their residual radioactivity after fusion-neutron irradiation. At the same time, it has been found that those two kinds of interphases will degrade under oxidation environment at elevated temperature [9,10].

SiC interphases prepared by CVD process, with high crystalline and near-stoichiometric characteristics [7], have excellent properties of oxidation resistance and stability during neutron irradiation. Many researchers have reported the applications of SiC interphases in the SiCf/SiC composites, but the SiC interphases always act as constituent layers in the $(X–Y)_n$ multilayer interphases [11,12], and the investigations on the effects of single layer SiC interphases on the mechanical properties of SiCf/SiC composites are rare.

The objective of this paper is to investigate the effects of single layer CVD SiC interphases on the mechanical properties and interfacial microstructure of SiCf/SiC composites fabricated by PIP process, and the reinforced mechanisms of the SiC interphases in the SiCf/SiC composites are also analyzed.

2. Experimental

The reinforcements used to prepare 2D-SiCf/SiC composites were plain-weave KD SiC fiber cloths, and the properties

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Table 1
Properties of KD SiC fibers.

Type	KD
Diameter (μm)	14–16
Number of filaments (fil/yarn)	∼1200
Tensile strength (MPa)	1300–1400
Density (g cm^{-3})	∼2.55
C/Si atom	1.23
Chemical compositions of the fiber surface	Si, C, O

of the KD SiC fibers (from National University of Defense Technology, China) are listed in Table 1.

Methyltrichlorosilane (MTS) was used for SiC fiber coating deposition. The deposition experiments were performed in a hot-wall vertical reactor. Hydrogen was used as carrier gas, which delivered the MTS source precursor through the bubbler to the reactor; argon was used as diluent and protective gas. The flow rate of hydrogen and argon were fixed at 200 and 75 sccm (cm^3/min), respectively. The deposition was performed at 1323 K under a total pressure of 5.0 kPa.

Polycarbosilane (PCS), the precursor of SiC matrix, with relative molecular mass ∼1300 and softening point ∼210 °C, was synthesized in our laboratory. The fabrication route of the 2D-SiCf/SiC composites is shown in Fig. 1.

Three-point bending tests were carried out at ambient temperature. The sample geometry was about $60^l \times 4^w \times 3^t$ mm. mm. The support span was 50 mm, and the cross-head speed was 0.5 mm/min. The fracture surface of the composites after mechanical tests was observed by a scanning electron microscope (SEM, HITACHI FEG S4800). The surface of the pulled out fibers in the composites and KD SiC fibers was analyzed by EDS equipped with SEM. The phases of KD fibers before and after SiC coating deposition were characterized by X-ray diffraction (XRD) analysis using monochromatic Cu Kα radiation with a D8 ADVANCE diffractometer (Bruker, Germany).

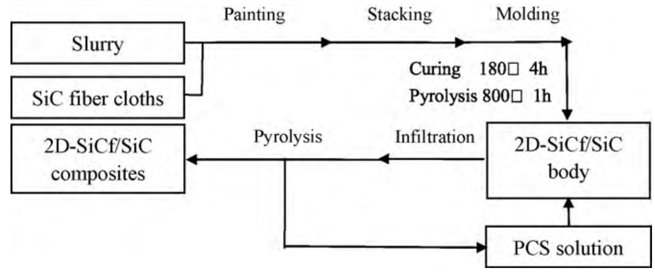


Fig. 1. Preparation route of 2D-SiCf/SiC composites.

3. Results

3.1. Characteristics of the CVD SiC fiber coating

SEM images of the SiC fiber with CVD SiC coating are shown in Fig. 2c–e. Those of uncoated KD SiC fibers are also shown in Fig. 2a and b for comparison. It can be found that

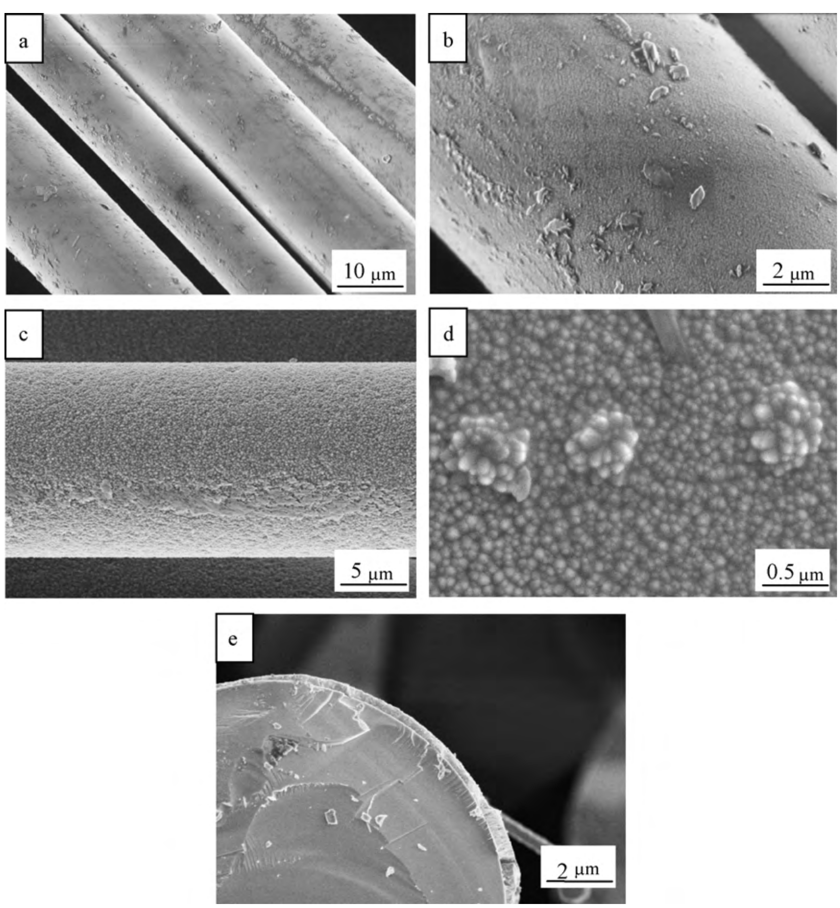


Fig. 2. SEM images of SiC fibers: (a and b) as-received and (c–e) with CVD SiC coating.

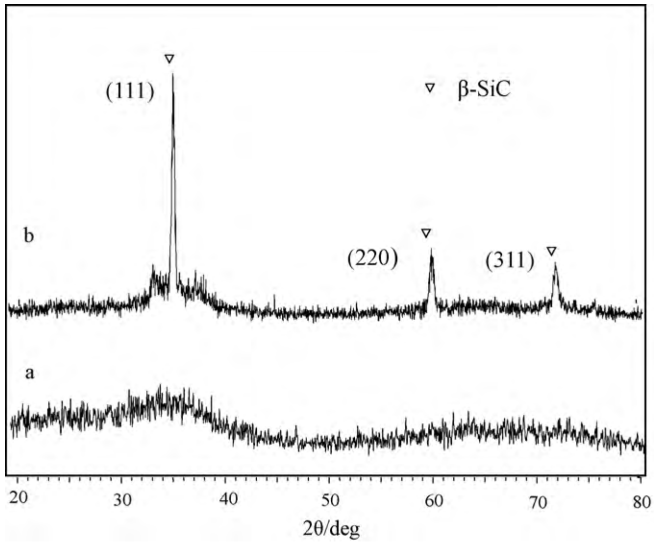


Fig. 3. XRD patterns of SiC fibers: (a) as-received and (b) with CVD SiC coating.

some strumae structure defects distribute on the uncoated SiC fibers. After deposited by SiC coating, the surface of the SiC fibers is smooth, and the defects are made up (Fig. 2c). In the higher magnification SEM image of the SiC fiber coating (Fig. 2d), some traces of strumae are observed on the fiber surface. From the SEM cross-sectional image of the SiC fiber (Fig. 2e), the thickness of the SiC fiber coating is about 0.4 μm.

XRD pattern of SiC fibers with CVD SiC coating is represented in Fig. 3b. For comparison, that of the as-received KD SiC fibers is also shown in Fig. 3a. No obvious XRD peaks are observed in Fig. 3a, which show that the KD SiC fibers are amorphous structures. The very broad band around a 2θ value of 35.7° in KD SiC fibers is attributed to β-SiC. In Fig. 3b, three obvious XRD peaks at about the 2θ values of 35.7°, 60.4° and 71.8° ($d = 0.251$, 0.154 and 0.131 nm) are detected, which correspond to the (1 1 1), (2 2 0) and (3 1 1) reflections of β-SiC. Therefore, it can be confirmed that the CVD SiC coating is β-SiC phase.

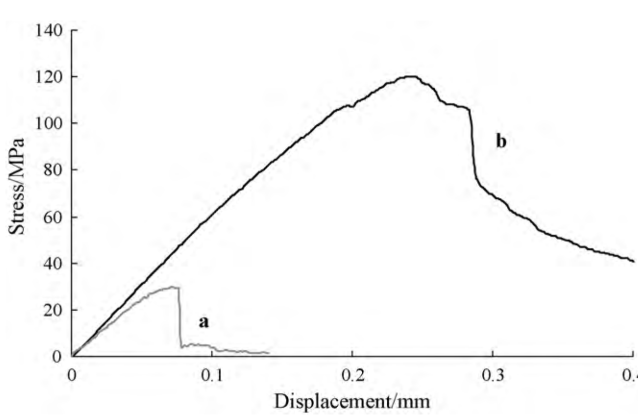


Fig. 4. Stress/displacement curves of SiCf/SiC composites: (a) as-received and (b) with CVD SiC interphases.

3.2. Mechanical properties of the SiCf/SiC composites

The stress/displacement curves of the SiCf/SiC composites without and with CVD SiC interphases are shown in Fig. 4. As shown in Fig. 4a, the SiCf/SiC composites without SiC interphases (as-received SiCf/SiC composites) exhibit an obvious brittle fracture behavior. In contrast, the SiCf/SiC composites with CVD SiC interphases (SiC–SiCf/SiC composites) show a standard toughened fracture behavior (Fig. 4b), the flexural strength of which is about 4 times that of the as-received SiCf/SiC composites. When the load reaches maximum, it will not decrease sharply, but drop off gradually.

4. Discussion

4.1. Microstructural analysis of the SiCf/SiC composites

From the stress/displacement curves of the two kinds of SiCf/SiC composites, it can be concluded that the CVD SiC interphases play a key part in determining the mechanical properties and failure behaviors of the SiCf/SiC composites. In order to investigate the effects of the CVD SiC interphases on the mechanical properties of the composites deep, the microstructure of the SiCf/SiC composites was analyzed by SEM.

The fracture surface morphology of the SiCf/SiC composites with and without SiC interphases is shown in Fig. 5. For the as-received SiCf/SiC composites, the fracture surface is very even, and nearly no pulled out fibers can be found (Fig. 5a). As shown in Fig. 5b, a strong interfacial bonding occurs at the interfaces, and no interfacial debonding behaviors can be observed. Contrarily, concerning the SiC–SiCf/SiC composites, the fracture surface shows an evident fiber pullout (Fig. 5c), and the maximum length of the pullout fibers can exceed 20 μm. The surface of the pulled out fibers is relatively smooth. The crack extending paths are shown in Fig. 5d. When cracks extend into the SiC interphases, owing to their non-layered crystal structures, the cracks cannot deflect, but traverse the SiC interphases. While the cracks propagate to the SiC fibers, due to the relatively weak bonding between the SiC interphases and SiC fibers resulted from the CVD process, they will deflect at the interfaces between the SiC fibers and SiC interphases, which can lead to interfacial debonding (as shown in Fig. 5d) and fiber pullout in the composites.

In order to confirm the interfacial debonding mechanisms in the SiC–SiCf/SiC composites further, the EDS analysis of the fiber surface was done. As shown in Fig. 6, the difference of the chemical compositions between the surface of pulled out fibers in the SiC–SiCf/SiC composites and that of the as-received KD SiC fibers is not evident, which indicates that the interfacial debonding does not occur within the SiC interphases, but at the interfaces between the SiC interphases and SiC fibers.

4.2. Effects of the CVD SiC interphases

In general, the mechanical properties of the continuous fiber reinforced ceramics matrix composites are determined by the in situ fiber strength and interfacial characteristics. In our previous

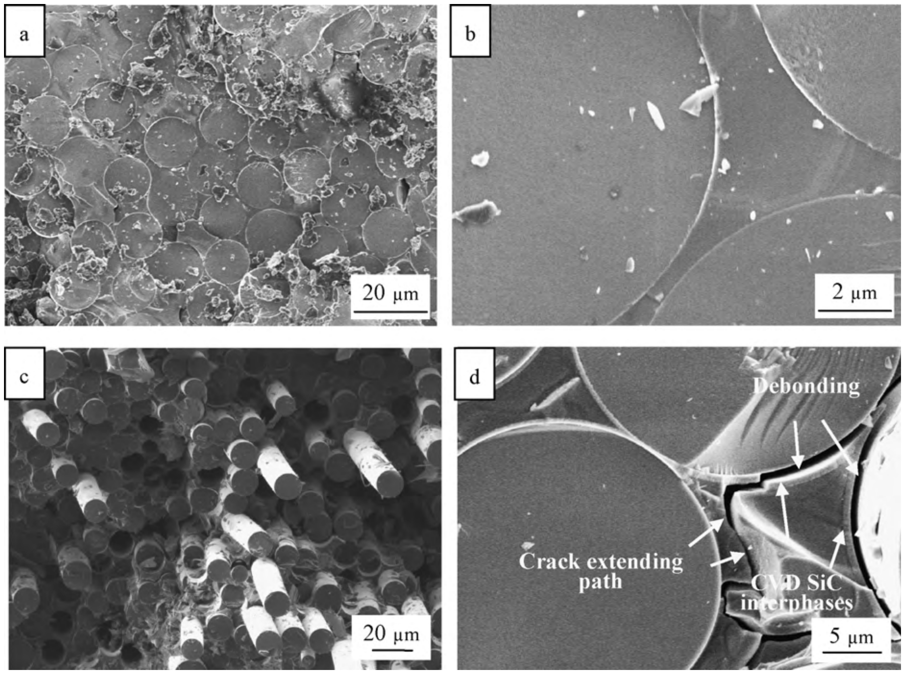
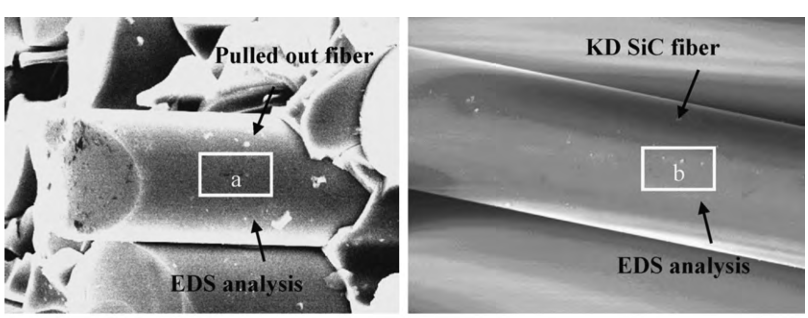


Fig. 5. Fracture surface morphology of SiCf/SiC composites: (a and b) as-received and (c and d) with CVD SiC interphases.

work [13], the microstructures and characteristics of the interphases in the as-received SiCf/SiC composites have been investigated detailedly. It has been concluded that the elemental diffusions and chemical reactions can occur at the interfaces, which is owing to the existence of silicon-based oxide interphases introduced by the surface layers of the as-received KD SiC fibers. At the same time, as shown in Fig. 2a and b, the rough surface characteristics of as-received KD SiC fibers would lead to strong physical interfacial bonding, and the defects on the SiC fibers may result in a larger stress concentration, and the largest stress introduced by the volume shrinkage of SiC matrix during precursor pyrolysis may cut off the SiC fibers (see Fig. 7), which will bring a tremendous physical damage to SiC fibers. The stronger interfacial bonding and serious fiber damage lead to an execrable interfacial state and low in situ fiber strength, respectively, which can make the fiber reinforced mechanisms be out of service, and a brittle fracture behavior occurs in the as-received SiCf/SiC composites.

In contrast to the in situ fiber strength and interfacial characteristics of the as-received SiCf/SiC composites, those of the SiC–SiCf/SiC composites all can be improved by introducing the CVD SiC interphases. As is well known that CVD SiC with high-crystalline and near-stoichiometric characteristics is an inert material, it can play a diffusion barrier part at the interfaces, which can prevent chemical reactions and elemental diffusions from occurring at the interfaces during composite preparation. Therefore, the CVD SiC interphases can weaken the chemical interfacial bonding and decrease chemical damage to the fibers. At the same time, from Fig. 2c, it can be seen that the defects on the KD SiC fibers can be made up by the SiC coating, and the smooth surface characteristics of SiC fibers would reduce the physical interfacial bonding and stress concentration. So, the appropriate interfacial bonding strength and relatively high in situ fiber strength make the SiC–SiCf/SiC composites exhibit an evident toughened fracture behavior.



a			b		
C (at. %)	Si (at. %)	O (at. %)	C (at. %)	Si (at. %)	O (at. %)
34.47	34.66	30.87	31.19	36.42	32.39

Fig. 6. EDS analysis of the SiC fiber surface: (a) pulled out fibers in the SiC–SiCf/SiC composites and (b) KD SiC fibers.

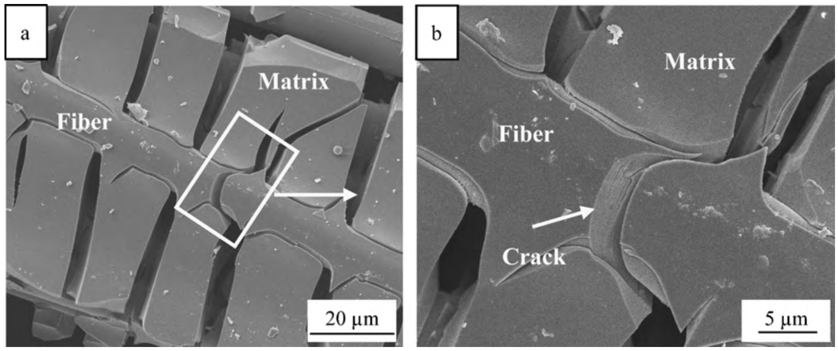


Fig. 7. Physical damage to SiC fibers in the as-received SiCf/SiC composites.

5. Conclusions

Effects of the single layer CVD SiC interphases on the mechanical properties and interfacial microstructure of the SiCf/SiC composites were investigated, and the following conclusions have been reached:

(1) Uniform β-SiC fiber coating about 0.4 μm thick is deposited on the SiC fibers by CVD process. The defects on the as-received KD SiC fibers can be made up by the SiC coating, and the surface of the SiC fibers with CVD SiC coating is smooth.
(2) Compared with the as-received SiCf/SiC composites, the SiCf/SiC composites with CVD SiC interphases exhibit an obvious toughened fracture behavior, the flexural strength of which is about 4 times that of the as-received SiCf/SiC composites.
(3) The CVD SiC interphases can provide protection for SiC fibers and appropriate interfacial bonding for SiCf/SiC composites. The relatively high in situ fiber strength and low interfacial bonding strength can lead to interfacial debonding and fiber pullout in the composites, which are advantageous of the improvement of the mechanical properties of the SiCf/SiC composites.

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