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Fabrication of advanced SiC fiber/F-CVI SiC matrix composites with SiC/C multi-layer interphase

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Abstract

SiC/SiC composite with SiC/C multi-layer interphase coated on advanced SiC fibers was fabricated by the forced thermal-gradient chemical vapor infiltration (F-CVI) process. SEM and TEM observations verified that SiC/C multi-layer interphase was formed on SiC fibers. Both flexural and tensile strengths of SiC/SiC composite with SiC/C multi-layer interphase were approximately 10% higher than composites fabricated with single carbon interphase. The SEM observation of fracture surface for the composite with SiC/C multi-layer interphase revealed cylindrical steps formed around the fiber. Apparently several crack deflections occurred within SiC/C multi-layer interphase. Moreover, the SiC/C multi-layer applied in this study operated efficiently to improve the mechanical properties.

1. Introduction

The continuous silicon carbide fiber reinforced silicon carbide (SiC/SiC) composites are attractive candidate materials for fusion reactors because of their excellent mechanical properties at high temperature and low induced radioactivity after neutron irradiation [1–3]. Recently, highly crystalline near-stoichiometric SiC fibers have been produced including Tyranno SA [4]. They have superior mechanical properties under irradiation and oxidation compared with their SiC-based predecessors. Various processes have been employed for advanced fiber composite fabrication including chemical vapor infiltration (CVI), polymer impregnation and pyrolysis, reaction bonding and nano-infiltration and transient eutectic-phase process [5–8]. Among the various fabrication processes, CVI is attractive process for the fabrication of SiC/SiC composites because it yields highly crystalline, near-stoichiometoric SiC fiber damage during processing is minimized. The forced flow thermal-gradient CVI (F-CVI) process is a further improvement since the low rates of producing and high porosity, which are the peculiar problem of CVI, are overcome [9]. Previous work by authors has reported on F-CVI process optimization of SiC/SiC composite with carbon (C) interphase including the effect of interphase thickness on tensile strength [10,11].

It was reported that SiC/C multi-layer interphase composite with conventional fibers achieved superior mechanical properties compared to single C interphase [12]. However, the effect of SiC/C multi-layer with the advanced SiC fiber on the composite tensile strengths has not been reported. In this study, new concept (SiC/C)×6 multi-layer interphase was applied with advanced SiC fibers for further improvement in mechanical properties. The first SiC layer is designed to strengthen the bond between fiber and interphase. The second to fourth SiC layers are designed to provide multi-stage pull-out of fibers. The fifth and sixth SiC layers are designed to provide multi-stage pull-out of fiber bundles.

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The intermediate C layers are used only to separate each SiC layers. It was also reported that thin C layer reduces the sensitivity of oxidation [13] and irradiation [14]. A thin C layer (~50 nm) was therefore chosen in this study. The characterization of SiC/C multi-layer and the effect of SiC/C multi-layer on the mechanical properties were investigated in this study.

2. Experimental procedure

Tyranno SA fiber SiC fabrics (Ube Industries, Ube, Japan) were used as reinforcement for the SiC/SiC composite in this study. The precursor for C deposition was propylene (C_3H_6). Technical grade methyltrichlorosilane (MTS, CH_3SiCl_3, Gelest Inc., Tullytown, PA, USA) was also used for SiC infiltration. SiC fabric layers with fabric layer orientation of (0°/90°) were restrained in a graphite fixture. The SiC and C interphase coating on the fiber were sequentially deposited at 1100 °C and at 5 Pa. The C deposition condition of flow rate is 50 cm^3/min C_3H_6 and 1000 cm^3/min Ar. The SiC deposition condition of flow rate is 0.15 g/min MTS carried by 250 cm^3/min of H_2. Table 1 shows the condition parameters of SiC/C multi-layer interphase deposition. After the interphase layer was deposited, the preform was infiltrated at 1200 ° C under atmospheric pressure with a MTS flow rate of 0.3 g/min carried by 750 cm^3/min of H_2. A composite with single C layer as an interphase was also fabricated.

Mechanical properties of the composite were evaluated by 3-point bending and tensile testing. Three-point bending was carried out at ambient temperature with cross-head speed of 0.1 mm/min and a support span length of 20 mm. The test specimen was $2\times5\times25$ mm^3. Tensile testing was carried out at ambient temperature with cross-head speed of 0.5 mm/min. Further details of tensile test and specimen is described elsewhere [10,11]. The each number of specimens measured by bending and tensile testing is three.

Microstructure was observed by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). Composite fracture surface was evaluated by SEM. Electron probe microanalysis (EPMA) was carried out to evaluate the chemical composition of the fracture surface within the SiC/C multi-layer.

3. Results and discussion

The porosity of SiC/SiC composite fabricated in this study was approximately 20% with fiber volume fraction of approximately 39%.

A typical cross-sectional SEM microphotograph of SiC/SiC composite is shown in Fig. 1. The average thickness of each layer in SiC/C multi-layer interphase is given in Table 1. Although the deposition time of each C layer was same, the thickness of outer C layer was thicker than that of inner C layer. The reason is that the deposition area decreased at the outer C layer compared to inner C layer since the space among vicinal fibers was filled with the inner SiC and C layers.

Fig. 2 shows the cross-sectional TEM micrographs of SiC/SiC composite. The SiC/C multi-layer consisted of six (SiC/C) layers. Electron diffraction patterns for the corresponding SiC layers and SiC fiber are also shown in Fig. 2. The electron diffraction pattern of second SiC layer was similar to that of SiC fiber with fine grain size (~40 nm) [4]. The TEM observation reveals that the inner SiC layers (first to fourth SiC layers) consisted of finer SiC grains compared to the outer SiC layers (fifth and sixth SiC layers) and SiC matrix. The grain sizes of fifth and sixth SiC layers were almost the same as that of SiC matrix. A very thin C layer formed between SiC fiber and first SiC layer. The SiC fibers were coated by poly vinyl alcohol (PVA) as sizing material [4]. A very thin C layer formed by carbonizing the PVA since the fibers were heated at 1100 °C prior to the initial SiC interphase deposition.

Table 1
Condition parameters of SiC/C multi-layer interphase deposition

	Material	Designed thickness (nm)	Measured thickness (nm)	Deposition time (min)
(SiC/C)×6	SiC	50	56	4
	C	50	37	30
	SiC	200	119	15
	C	50	35	30
	SiC	200	132	15
	C	50	40	30
	SiC	200	153	15
	C	50	41	30
	SiC	500	432	38
	C	50	62	30
	SiC	500	572	38
	C	50	83	30

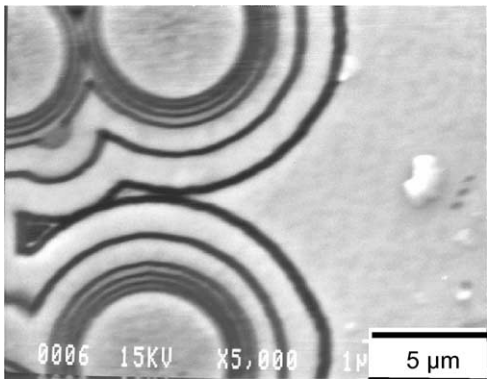

Fig. 1. Typical cross-sectional SEM microphotograph of SiC/SiC composite with SiC/C multi-layer interphase.

The tested composites had different porosities and fiber volume fractions. Araki et al. [15] reported that the flexural strength increased with decreased porosity of composite. Since the load is mainly maintained by unfractured fibers and friction between fractured fiber and interphase above the proportional limit stress, the flexural and tensile strengths depend on the porosity and fiber volume fraction of composite. The flexural and tensile strengths were, therefore, normalized by the following equation:

$$\text{Normalized strength} = \frac{\text{original strength}}{1-\text{porosity}} \frac{V_f}{\overline{V}_f},$$

where V_f and $\overline{V}_f$ are fiber volume fraction and the average fiber volume fraction of the composites.

The normalized flexural and tensile strengths of the SiC/SiC composite with SiC/C multi-layer and the composite with single C interphase are shown in Fig. 3. The normalized flexural and tensile strengths of the SiC/SiC composite with SiC/C multi-layer were approximately 10% higher than those of the composite with single C interphase.

The SEM micrographs of fracture surfaces of SiC/SiC composite with SiC/C multi-layer after flexural and tensile tests are shown in Fig. 4. The cylindrical steps around the fiber were observed after flexural and tensile tests and several crack deflections occurred within SiC/C multi-layer interphase. Furthermore, pull-out of fiber bundles occurred. These results indicate that the second to fourth SiC layers and fifth to sixth SiC layers operated efficiently to improve the fracture behavior. Since both multiple pull-out of fibers and fiber bundles occurred, higher fracture energy was absorbed compared to single C layer. The grain size of inner SiC layer was

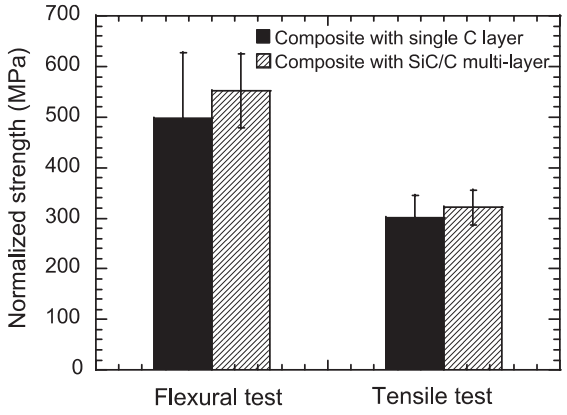

Fig. 3. Normalized flexural and tensile strengths of the SiC/SiC composite with SiC/C multi-layer and the composite with single C interphase.

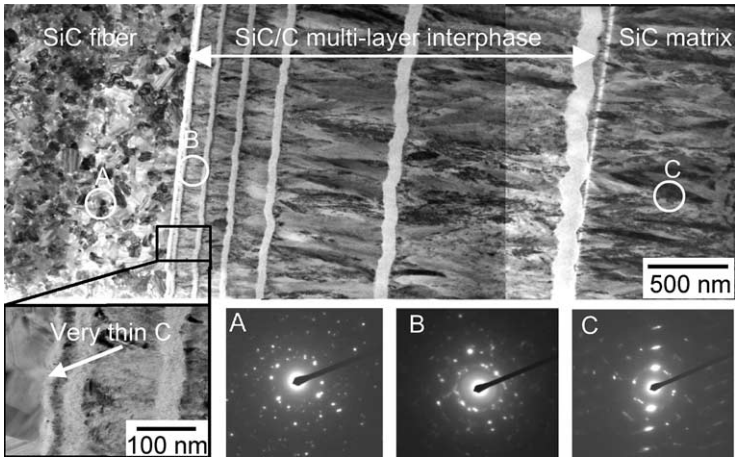

Fig. 2. Typical cross-sectional TEM microphotographs of SiC/SiC composite with SiC/C multi-layer interphase and the electron diffraction patterns for (A) SiC fiber, (B) second SiC layer and (C) SiC matrix.

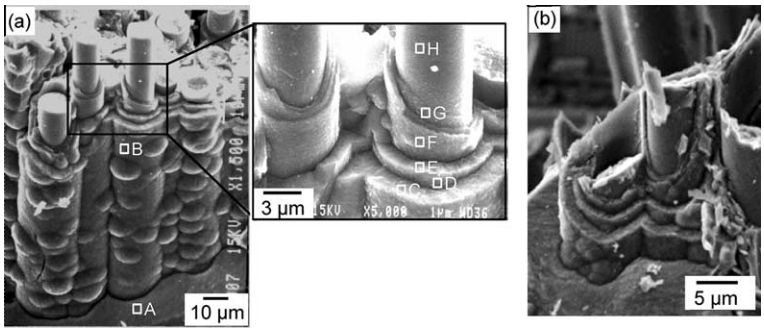

Fig. 4. SEM micorophotographs of fracture surface after (a) flexural test and (b) tensile test.

much smaller than that of SiC matrix. Cao reported that the flexural strength of SiC increased with decreasing the grain size [16]. The strength of inner SiC layer might be higher than that of SiC matrix. By the crack deflection within the outer C layer, the apparent thick fibers, which consisted of SiC fiber and inner SiC/C layer, were formed. These apparent thick fibers were able to retain higher load compared to the SiC fibers. Furthermore, the pull-out length might be increased because of the apparent thick fibers. From above reasons, the mechanical strength of composite with SiC/C multi-layer was higher than that of composite with single C layer.

The chemical composition of fiber surface on the fracture surface was slightly large amount of C comparing to the amount of Si by EPMA evaluation. The chemical composition of fracture surface after flexural test evaluated by EPMA is summarized in Table 2. The analyzed points of EPMA are also given in Fig. 4. This result indicates that the crack passed within the C layer although the very thin C existed between SiC fiber and first SiC layer. The reason is that the bonding strength between SiC fiber and C layer was high enough since the surface of Tyranno SA fiber was as rough as the SiC layer (Fig. 2).

The results of EPMA reveal that the crack deflection occurred at almost every C layer. The SEM observation reveals that the measured thickness of each C in the multi-layer was around 50 nm (Table 1). Results indicate that 50 nm-thick C layer was thick enough for the crack deflection. The particular SiC/C multi-layer configuration in this study operated efficiently to improve the mechanical properties.

Table 2
Chemical composition of the fracture surface within SiC/C multi-layer evaluated by EPMA

Analyzed points	Si (at.%)	C (at.%)
A	51	49
B	33	67
C	47	53
D	32	68
E	41	59
F	27	63
G	30	70
H	39	61

The analyzed points were shown in Fig. 4.

4. Conclusions

SiC/SiC composite with SiC/C multi-layer interphase coated on Tyranno SA SiC fibers was fabricated by F-CVI process. The characterization of SiC/C multi-layer and the effect of SiC/C multi-layer on the mechanical properties of the composite were investigated.

(1) The SEM and TEM observation verified that SiC/C multi-layer interphase was formed on SiC fibers in this process.
(2) Both flexural and tensile strengths of SiC/SiC composite with SiC/C multi-layer interphase were approximately 10% higher than that with single C interphase.
(3) Pull-out of fiber bundles as well as pull-out of single fiber occurred in the composite with SiC/C multi-layer.
(4) Crack deflections occurred at almost every 50 nm thick C layer. The particular SiC/C multi-layer configuration used in this study operated efficiently to improve the mechanical properties of SiC/SiC composite.

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