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Effects of random chopped fiber on the flexural strength and toughness of reaction bonded silicon carbide composite

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Abstract

Chopped carbon fibers were introduced into the SiC/C suspensions to enhance the mechanical behaviors of reaction bonded silicon carbide ceramics. The effects of fiber fraction on the flexural strength and fracture toughness were investigated. No preferred fiber orientation was observed on the fracture surface of the green body. This implies a homogeneous dispersion of chopped fiber in the composite. XRD analysis indicates a complete siliconization of both carbon particles and carbon fiber during liquid silicon infiltration. The flexural strength and fracture toughness increase with chopped fiber ranging from 10 vol.% to 30 vol.%. This improvement derives from the fiber pullout, fiber debonding as well as crack deflection. The consequent decline at fiber fraction of 40 vol.% is resulted from the siliconization of chopped fiber and the increase of residual silicon.

Keywords: A. Slip casting; B. Fibres; C. Mechanical properties; E. Structural applications

1. Introduction

Reaction bonded silicon carbide (RBSC) ceramic is a typical structural material characterized by full densification, low sintering temperature, short sintering time and near-net shaping [1–3]. Due to the merit of being near-net shaping, RBSC ceramic has been used as the large scale and shape complicated structural components including cutting tools, gas turbine engine and light armor [4–6]. In view of its high stiffness, low coefficient of thermal expansion (CTE) and high thermal conductivity, RBSC is also believed to be a potential material for optical mirrors of the next generation, key parts of high resolution space telescopes [7,8]. Typical fabrication process for RBSC optical mirrors involves sintering, joining, machining, coating and polishing. Such a complicated process requires excellent flexural strength and fracture toughness of the RBSC ceramic. But RBSC ceramic is known as a material inherently with low flexural strength and toughness. To improve the flexural strength and toughness, attempts have been made by fabricating RBSC ceramic with fine starting SiC particles, B_4C alternative, and silicon alloy infiltration [9–12]. Yet, however, the requirements for large scale components have not been fully met.

An alternative approach for RBSC ceramic reinforcing is to introduce fibers. The continuous fiber reinforced composites show high reliability and superior mechanical strength, and thus has been used in aerospace and energy conversation techniques [13,14]. However, the high cost, complex processing route and long production period limit its broader applications [15]. Krenkel [14] developed C/C–SiC composite with various carbon fibers by the liquid silicon infiltration (LSI) process, which shows evident superiority compared with the chemical vapor infiltration (CVI) and polymer infiltration and pyrolysis (PIP) processes. But, the complex route is still an obstacle in the fabrication of large scale, shape complicated components. In addition, continuous fibers can only improve the mechanical strength along the direction of the fiber aligned, while not in other direction [16]. This anisotropy of mechanical strength will bring down the reliability of the large scale components made of RBSC ceramic.

Homogeneously dispersed chopped fibers generally can avoid the anisotropy of mechanical properties. Research attention has been paid to the chopped fiber reinforced structural ceramics that high mechanical strength, convenient

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process method and low process cost are demanded [17,18]. The chopped carbon fiber reinforced ceramic matrix composite has been widely used in wear, thermal control, and electrical systems, e.g. SiC, ZrB_2 [19–21] in hot pressing sintering, due to its simple process route and low process cost. However, the high sintering temperature and pressure reduced the reinforcing effect of chopped carbon fiber. Here we focused on the random chopped carbon fiber reinforced RBSC composites that were prepared by LSI.

In the present work, the random chopped carbon fiber was homogeneously dispersed into SiC/C suspensions to improve the mechanical properties of RBSC ceramic. In view of the harmful effect of residual silicon on the flexural strength and toughness at the high fraction of chopped fiber, the chopped fiber fraction was controlled in the range of 10–40 vol.%. We presented the orientation of random chopped fiber in the composite. After LSI, both flexural strength and fracture toughness of the composite were enhanced in the RBSC composite. Further, the toughening mechanisms of the random chopped fiber in RBSC composites were evaluated.

2. Experimental procedure

Continuous carbon fiber (IM400-6K, TOHO, Japan) was cut into short segments of 3 mm in length. The diameter, elastic modulus and density of the continuous carbon fiber were 6 μm, 294 GPa and 1.74 g/cm^3, respectively. To remove the organic binder, the chopped carbon fibers were desized with 0.2 M dilute nitric acid for 24 h and then washed with deionized water. Then the chopped carbon fibers were pre-dispersed with ultrasound vibration. Here carboxymethyl cellulose (CMC, molecular weight 20000, GONSO, Shanghai, China) was chosen as dispersant. Two commercial α-SiC (purity 98.5%, Huanyu, Zibo, China) powders of 10 μm and 60 μm, and amorphous carbon were chosen as the matrix. The raw powders together with dispersant and plasticizer were ball milled with silicon carbide balls in water for 10 h. Then the as-treated chopped carbon fibers were added into the SiC/C suspension and a further ball milling for another 2 h was conducted. The homogeneous slurry was evacuated for 15 min, and then poured into the plaster mould. After drying for 72 h, the green body was obtained with near-net shaping. Finally, the obtained green body was sintered by LSI at 1700 °C for 90 min. Four specimens were prepared, and the volume ratios of random chopped fiber to the starting SiC particles were 10 vol.%, 20 vol.%, 30 vol.% and 40 vol.%.

Microstructure of the polished and fracture surfaces was observed with scanning electron microscopy (QUANTA 200, FEI, USA). Flexural strength was tested with crosshead speed of 0.5 mm/min and span of 30 mm. The tested bars were in dimension of 3 mm × 4 mm × 36 mm and were polished with diamond slurry of 3.5 μm. Fracture toughness was evaluated using a single edge notched beam (SENB) with crosshead speed of 0.05 mm/min and span of 20 mm. The tested bars were in dimension of 2 mm × 4 mm × 22 mm and tested with a notch of 2 mm in depth and 0.02 mm in width. The data for each specimen was averaged over five tests. Crystal structure of the composite was analyzed by X-ray diffraction (XRD, Rigaku, Dmax-rb).

3. Results and discussion

3.1. Orientation of chopped fibers

Typical fracture surface of the green body with 30 vol.% random chopped fiber is shown in Fig. 1(a). The chopped fibers were pulled out from the matrix in the flexural strength test. No preferred fiber orientation is observed in the image, implying the homogeneous dispersion of the chopped fibers in the matrix. This homogeneous dispersion is ascribed to a series of treatments, including acid impregnation, ultrasound vibration, ball milling as well as the CMC dispersant. However, due to the impact between the fibers and the SiC balls, the fiber length is reduced, which is inevitable in the ball-milling method. The morphology of polished surface is presented in the BSE image, which is shown in Fig. 1(b). The dark gray, light gray and white phases correspond to, respectively, the starting α-SiC, the formed β-SiC and the residual silicon. The carbon particle phase, which is often a black phase, is not observed in the graph. This means the carbon particles are completely converted into SiC during LSI. The linear, light gray phase is the chopped fibers that were oriented in the polished plane. As reinforcement, the random orientation of chopped fiber can effectively resist crack propagation towards any direction. This isotropy is superior to the anisotropic reinforcing of the continuous carbon fiber reinforcing. However, during LSI the carbon fiber reacted

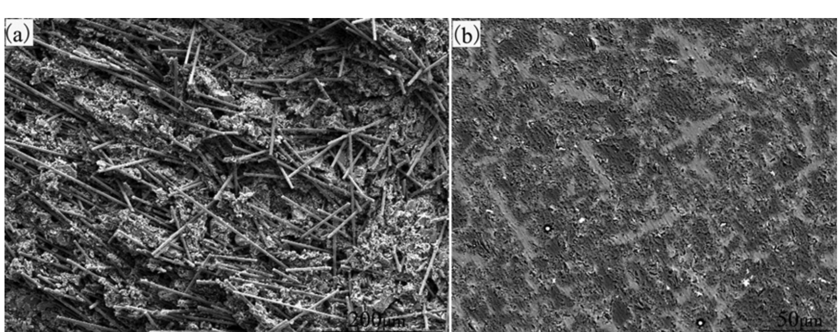

Fig. 1. Distribution of chopped carbon fibers in the matrix: (a) indicates microstructure of the green body with 30 vol.% chopped fiber, and (b) indicates morphology of the polished surface prepared by LSI at 1700 °C.

with molten silicon, resulting in a reduction of tensile strength and a variation of diameter in the composite.

3.2. Composition of the composite

XRD analysis was carried out to determine the crystal structure of the composites. Fig. 2 exhibits the XRD patterns for composites with 20 vol.% and 40 vol.% chopped fibers. The composites consist of α-SiC, β-SiC and silicon. Peaks assigned to carbon are not determined, which indicates that carbon had been consumed completely by the siliconization reaction. This is in agreement with the results of the BSE image in Fig. 1(b). A slight development of silicon can be observed in the pattern of 40 vol.% chopped fiber, implying an increase of residual silicon in the composite. This increase originates from the porosity increase of the green body; during LSI, the pores were filled by the molten silicon so as to realize complete densification of the sintered body. In the mechanical test, the increase of residual silicon leads to the reduction of both flexural strength and fracture toughness.

3.3. Flexural strength and toughness of the composite

In Fig. 3 the flexural strength and fracture toughness as a function of chopped fiber volume fraction are given. Noteworthy is the similar varying trends of flexural strength and fracture toughness with increasing of chopped fibers.

A maximum value of 5.1 MPa $m^{1/2}$ is obtained with 30 vol.% chopped fiber. The consequent decline originates from the increase of residual silicon, which is a deleterious phase in RBSC composites because of the high brittleness and low melting point. The toughening mechanism of the composite will be discussed in the following part. Similarly, the flexural strength of the composites is enhanced by increasing of the chopped fiber. The value of flexural strength related to chopped fiber fraction of 30 vol.% (416 MPa) is approximate twice than that related to 10 vol.% (213 MPa) chopped fiber. This increase is related to the microstructure change of the composites. The siliconization of carbon fiber produced fine β-SiC particles of several micrometers, which

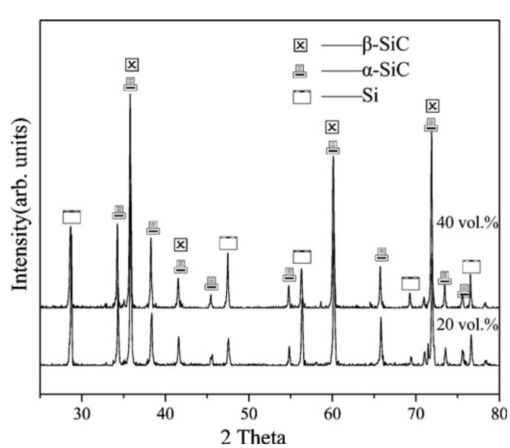

Fig. 2. Composition of the random chopped fiber reinforced RBSC composites with fiber fraction of 20 vol.% and 40 vol.%.

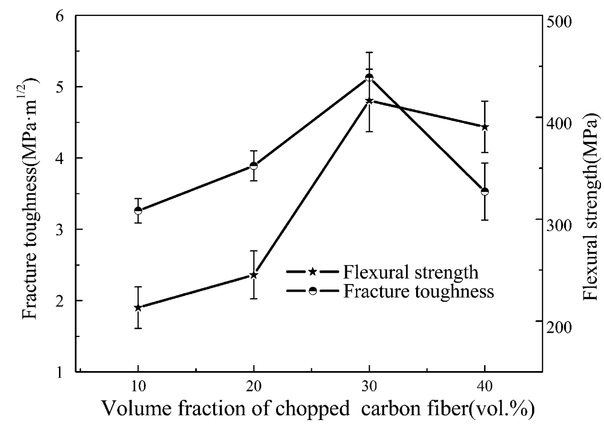

Fig. 3. Variation of flexural strength and fracture toughness with chopped fiber ranging from 10 to 40 vol.%.

facilitated the densification of the composite. Besides, crack propagation path was changed at the fiber region.

Compared with the C/C–SiC composite by LSI [14,22], the product in this study shows higher flexural strength due to the complete siliconization of the starting carbon; however, the fracture toughness is lower, resulting from the siliconization of chopped fiber filaments and the higher fraction of residual silicon. From the reported work of Krenkel and Berndt [22], the fiber bundles were protected from the siliconization reaction by the dense carbon matrix, so the toughening performance of carbon fiber was excellent. With regard to the fabrication process, the route here presents a simple and low cost method.

3.4. Toughening mechanism of the composite

The toughening performance of chopped fiber is controlled by the fiber distribution, interface phase and matrix properties. Especially, the interface strength is a key factor in determining mechanical properties of the fiber reinforced ceramic matrix composites. The introduction of chopped fiber can effectively improve the toughness of the RBSC composite as shown in Fig. 3. The main toughing mechanism in this composite consists of fiber debonding, fiber pullout and crack deflection.

Morphologies of chopped fibers on the fracture surface are illustrated in Fig. 4(a–c). The LSI process generally leads to a strong bonding strength between fibers and matrix; however, the fiber debonding is observed on the fracture surface, which is shown in Fig. 4(a). It is measured that diameter of the chopped fiber is 9.5 μm, which is higher than the starting chopped fiber. This is explained by two causes: (1) attachment of nano SiC particles formed from reaction between carbon particles and molten silicon; (2) volume expansion of chopped fiber due to the siliconization reaction. Fiber pullout and fiber breakage are shown in Fig. 4(b) and (c), respectively. The fracture surface formed around the fiber during the fiber pullout, which evidently consumed the fracture energy. So, fiber pullout is considered the most effective mechanism of fiber toughening. The fiber breakage suggests the strong bonding between the fiber and the matrix, which should be avoided in the following study.

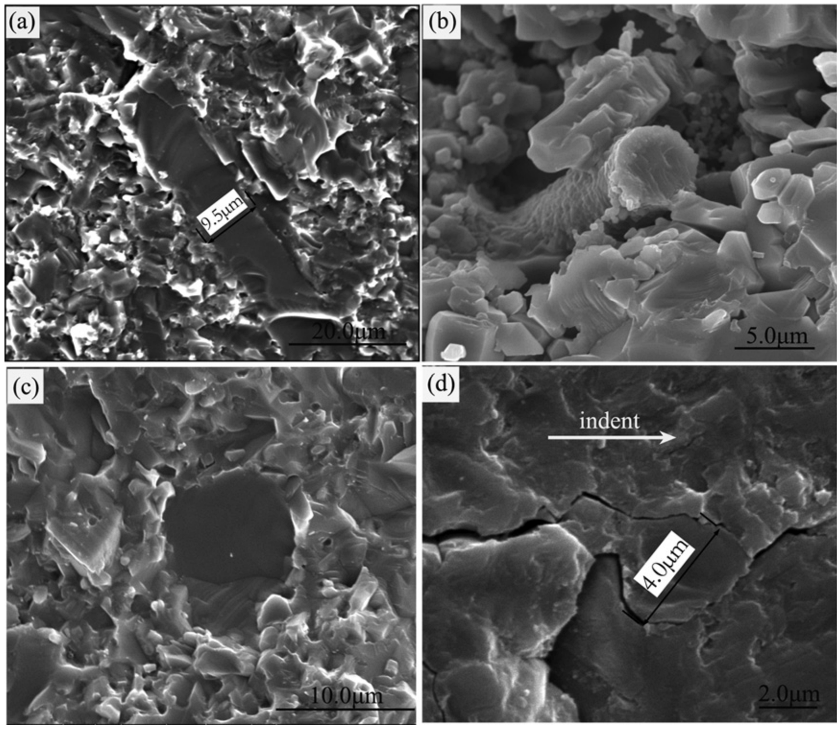

Fig. 4. Morphologies of chopped fibers in the RBSC composites: (a), (b) and (c) correspond to fiber debonding, fiber pullout and fiber breakage, respectively, on the fracture surface; and (d) is the fiber debonding on the polished surface.

The fiber debonding on the polished surface was examined by SEM, and the result is shown in Fig. 4(d). The horizontal arrow indicates the general direction of crack propagation. From the micrograph, the crack propagates along the interface between the chopped fiber and the matrix. This implies a moderate bonding strength between chopped fiber and the matrix. As is well known, the moderate bonding strength can effective impels crack deflection along the fibers. The diameter of the chopped fiber in Fig. 4(d) is lower than the starting fiber. It is speculated that the carbon on the fiber surface dissolved into the molten silicon during LSI.

The crack propagation on the polished surface is presented in Fig. 5. The cracks were introduced from Vickers indentation of 19.6 N. The crack path is indicated by arrows. A deflective path is observed in the image, which consumed more fracture energy in comparison with the straight path. Also, grain bridging occurs on the polished surface due to the platy SiC grains, as is shown by the long arrows.

Fracture surface of the Si–SiC matrix is shown in Fig. 6. Based on the SEM micrograph, the intergranular fracture is the main fracture mode, coupled with the brittle fracture of residual silicon. Under loading, the crack propagated along the boundary

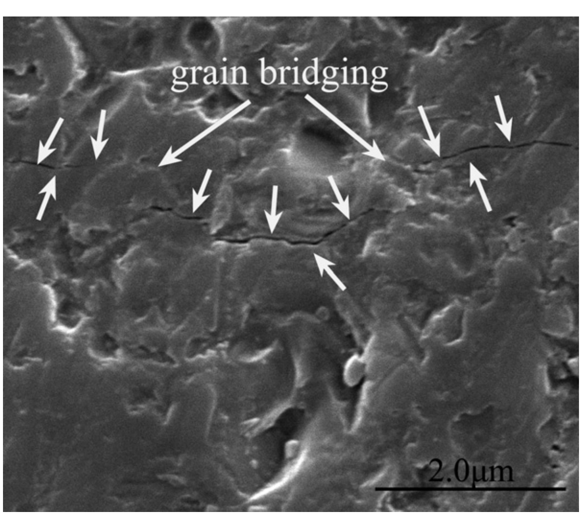

Fig. 5. Crack deflection and grain bridging on the polished surface of the composite.

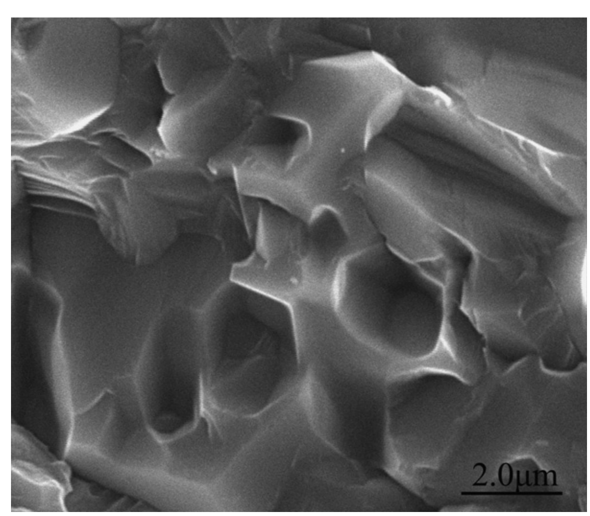

Fig. 6. Morphology of the fracture surface of the RBSC matrix sintered at 1700 °C.

of micron, faceted SiC particles, which were produced from the siliconization reaction of the starting carbon particles.

4. Conclusions

In this investigation, the random chopped fibers with volume fraction ranging from 10 vol.% to 40 vol.% were dispersed into SiC/C suspensions to reinforce the RBSC ceramic. The influence of random chopped fiber on the flexural strength and toughness was investigated. To overcome the aggregation of chopped fibers, the chopped fibers were pre-treated with dilute nitric acid, followed by homogeneous dispersion into the SiC/C suspensions with dispersant. The fracture surface of green body showed no preferred fiber orientation. During LSI, the chopped carbon fibers as well as the carbon particles were converted to SiC. Although the siliconization of carbon fiber was a deleterious reaction for the mechanical properties of the composite, both flexural strength and fracture toughness of the composite increased significantly in the range of 10–30 vol.% chopped fiber. When further increasing the volume fraction of chopped fiber, a decrease was observed due to the increase of residual silicon. The XRD analysis confirmed the increase of residual silicon at 40 vol.% chopped fiber. The predominant toughening mechanisms of chopped fiber were fiber debonding and fiber pullout, although siliconzation reaction suppresses the reinforcing performance of chopped fiber. The crack deflection and grain bridging also improve the fracture toughness of the RBSC composites.

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