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Manufacturing 2D carbon-fiber-reinforced SiC matrix composites by slurry infiltration and PIP process

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Abstract

Sub-micrometer SiC particles were firstly added to the preceramic solution in the first infiltration step to enhance the mechanical properties of 2D C_f/SiC composites fabricated via polymer infiltration and pyrolysis (PIP) process. The effects of pyrolysis temperature and SiC-filler content on microstructures and properties of the composites were systematically studied. The results show that the failure stress and fracture toughness increased with the increase of pyrolysis temperature. SiC filler of sub-micron scale infiltrated into the composites increased the mechanical properties. As a result, for the finally fabricated composite infiltrated with a slurry containing 40 wt.% SiC filler, the failure stress was doubled compared to that without SiC filler addition, and the fracture toughness reached ≈10 MPa $m^{1/2}$.

Keywords: B. Porosity; B. Composite; C. Mechanical properties; Preceramic polymer

1. Introduction

Continuous fiber reinforced ceramic matrix composites (CFCMCs), such as C_f/SiC, SiC_f/SiC composites have been widely recognized as the most promising candidates for brake disks, heat exchangers, advanced aero-engines, fusion power reactors and space usage for their outstanding characteristics including high toughness, low density, thermal and chemical stability, radiation tolerance and so on [1–4]. Especially, the C_f/SiC composites, for the relatively lower cost, larger-scale production and better thermal stability at elevated temperature of C fiber, have been extensively investigated. This kind of composites can be prepared by chemical vapor infiltration (CVI), liquid or vapor silicon infiltration (LSI or VSI) and hot pressing (HP) [5–8]. CVI process is difficult to operate and requires complex equipment. The HP process limits the components to simple plate shapes and the consequent machining is of high cost. During LSI or VSI process, the residual silicon in the composite leads to material strength degradation at elevated temperatures [9]. In recent years, with the advent of preceramic polymer with high ceramic yield, polymer infiltration and pyrolysis (PIP) process has been developed for the relatively lower processing temperature, more convenient operation and relatively simple equipment. By this process, the components even with complex shape and large dimension can be easily fabricated. Slurry containing SiC particles is usually employed in the first infiltration step to enhance infiltration efficiency. Thus, particle content in the slurry should be optimized to obtain better mechanical properties.

The present research work involved the fabrication of 2D C_f/SiC composites by slurry infiltration and PIP step, using polycarbosilane (PCS) as preceramic precursor and SiC particles as inert fillers. The effects of pyrolysis temperature and SiC filler content on the physical and mechanical performances of the 2D C_f/SiC composites were investigated.

2. Experimental procedure

2.1. Sample preparation

The 2D woven C fiber fabrics (Xinka Carbon Co., Shanghai, China) were used to prepare the fiber preforms which were

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Table 1
Properties of the Xinka C fiber

Fiber type	Density (g cm^{-3})	Strength (MPa)	Modulus (GPa)	Filament	Diameter (μm)
Xinka	1.74–1.77	2000–3000	175–215	1k	5

employed as reinforcement for composite fabrication in this paper. Typical properties of the fiber are listed in Table 1. The preforms were obtained by stacking the 2D 0°/90° woven fabrics. The fiber volume fraction of the preform was about 30%.

In the first infiltration step, slurries containing various SiC filler (0.5 μm, Germany) content were used. To enhance the infiltration efficiency, the vessel containing the fiber preforms was first evacuated by a vacuum pump. Then, slurries were added to immerse the preforms and a pressure of 2 MPa was applied by gas to facilitate the infiltration process. After drying, the samples were pyrolyzed at 800–1100 °C in nitrogen atmosphere with a heating rate of 5 °C/min. Subsequently, the pyrolyzed composites underwent seven PIP process, using 50 wt.% (PCS) solution (with no filler).

2.2. Sample characterization

The open porosity of each sample was measured by the Archimedes method. The flexural strength was obtained by three-point-bend testing on the Instron 5566 universal testing machine, with samples of 2.5 mm × 4 mm × 36 mm dimensions, a cross-head speed of 0.5 mm/min and a span of 24 mm. The fracture toughness (K_{IC}) was determined by the single edge notched beam (SENB) method, with samples of 2.5 mm × 5.0 mm × 36 mm dimensions, a cross-head speed of 0.05 mm/min, a notch depth of 2.40 mm and a span of 20 mm. Five individual samples were tested at each point. The polished cross sections and fracture surfaces after the bending test were observed by scanning electron microscope (SEM).

3. Results and discussion

3.1. Effect of pyrolysis temperature on the physical and mechanical properties

The open porosity and mechanical properties of the composites pyrolyzed at 800–1100 °C are plotted as functions of pyrolysis temperature in Figs. 1 and 2. As seen from Fig. 1, a decrease of open porosity is observed with an increase of pyrolysis temperature. As reported in [10], the preceramic polymer would be pyrolyzed completely and closed pores in the composites opened at elevated temperature with the result that the composites may be refilled in the following infiltration steps. Hence, the open porosity gradually decreased with increase of the pyrolysis temperature. In Fig. 2, the composite pyrolyzed at 800 °C exhibited the lowest failure stress of only ≈97 MPa. When the pyrolysis temperature was enhanced to 1100 °C, the failure stress increased to ≈232 MPa, more than two times higher than that of composite pyrolyzed at 800 °C. With the increase of pyrolysis temperature, the bonding strength between the matrix and fiber increased, which is more efficient in load transfer from the matrix to the fiber. However, the measured failure stress is lower than the calculated strength which can be calculated by the following formula:

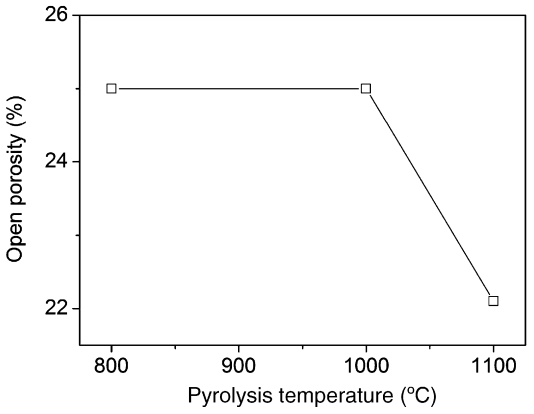

Fig. 1. Open porosity as a function of pyrolysis temperature.

$$\sigma \approx \frac{\sigma_f^R V_f}{2}$$

σ is the calculated strength of the composites, σ_f^R the average strength of the fiber and V_f is the fiber volume fraction, 1/2, for the 2D C_f/SiC composite. The calculated value is ≈375 MPa for the as-fabricated composites. The reason is probably that formation and propagation of the microcracks in the matrix result in early failure of the matrix and the reinforcing fiber in such fiber-reinforced ceramic matrix composites. When the pyrolysis temperature increased to 1100 °C, the fracture toughness increased to ≈10 MPa m$^{1/2}$. Because rather low fracture toughness can be expected at 800 °C, no test of fracture toughness was conducted for such a low processing temperature.

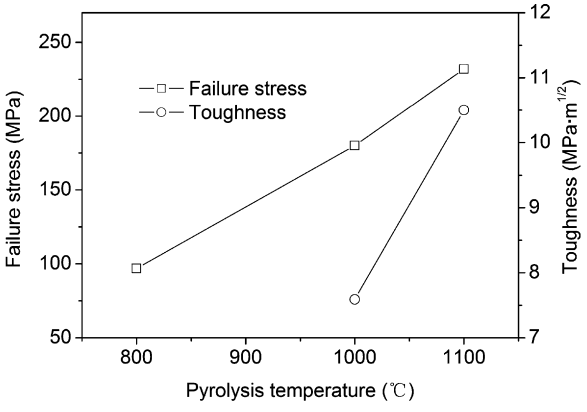

Fig. 2. Failure stress and toughness as functions of pyrolysis temperature.

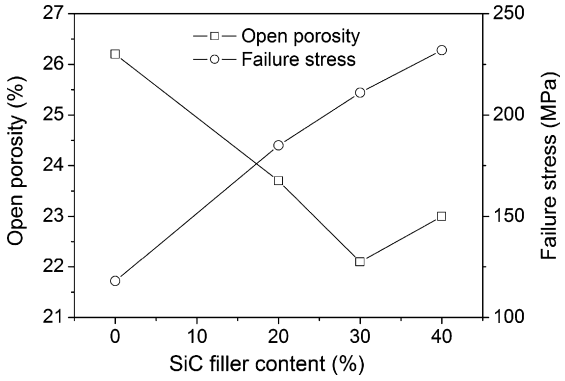

Fig. 3. Open porosity and failure stress as functions of slurry concentration.

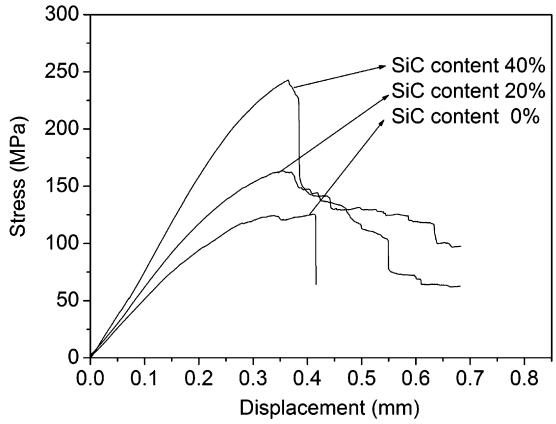

Fig. 5. Stress–displacement curves of the composites pyrolyzed at 1100 °C.

3.2. Effect of slurry concentration on the physical and mechanical properties

The effect of SiC filler content on the open porosity and failure stress of the composites was also investigated in this paper. The results are shown in Fig. 3. It can be easily seen that the open porosity decreased with an increase of filler content, indicating addition of SiC filler enhanced the infiltration efficiency for higher ceramic yield and smaller volume shrinkage can be achieved during pyrolysis. A slight enhancement of open porosity was observed when SiC filler content increased from 30 to 40 wt.%, which may be ascribed to the formation of more closed pores in the composite surface, which hampered the further polymer infiltration process. This feature suggested that high filler loading might have a negative influence, impeding further densification [11]. Therefore, it was considered that the effect of the filler loading on the physical and mechanical properties should be clarified in order to optimize the infiltration process. The failure stress also gradually increased with the increase of SiC content. The infiltrated SiC particles also act as a reinforcement, which leads to increased mechanical properties of the PIP-derived matrix and more tight bonding between the matrix and the fibers, which is benificial for load transfer from the matrix to the fibers. Thus, the failure stress of the final composite was enhanced.

Fig. 4 shows the cross-sectional micrographs of the 1100 °C processed C_f/SiC composite infiltrated with 40 wt.% slurry. As shown in Fig. 4(a), some isolated large pores could be observed

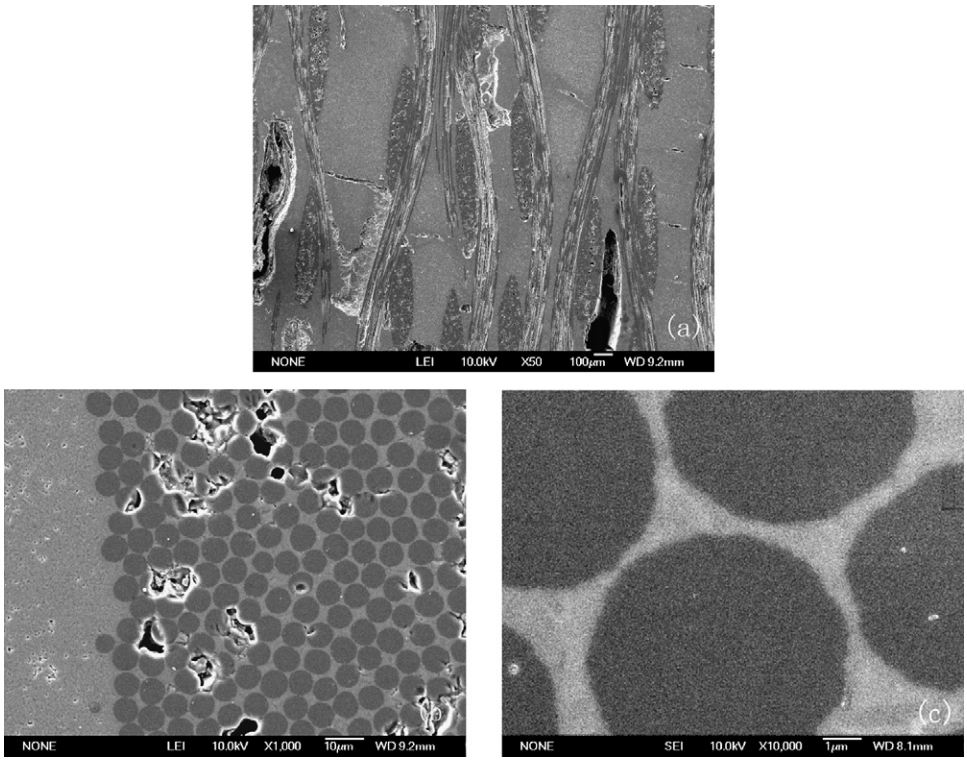

Fig. 4. Typical SEM micrograph of polished cross section of the composite infiltrated with slurry containing 40 wt.% SiC: (a) inter-bundle matrix and large pores, (b) intra-bundle matrix and small pores and (c) fine physical compatability between matrix and fiber.

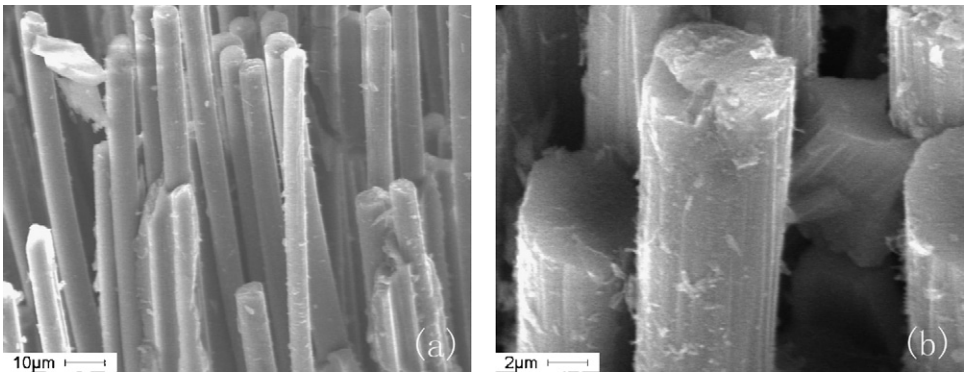

Fig. 6. SEM micrograph of fractured surface for composite pyrolyzed at 1100 °C: (a) sound fiber pullout and (b) fine-integrity of the fiber surface.

in the inter-bundle areas even after several infiltration-pyrolysis cycles, which is a commonly observed phenomenon in the PIP-derived samples for the low PIP efficiency in filling such large inter-bundle pores. In Fig. 4(b), dispersed residual pores were easily observed in the intra-bundle regions, which were ascribed to the shrinkage of the infiltrated PCS on pyrolysis and the difficulty for achieving effective polymer infiltration after the matrix was formed in the first cycle slurry infiltration. Generally, during PIP process, the size and number of residual pores left in the inter- and intra-bundle areas would gradually decrease when the PIP cycles proceeded and then hindered further polymer infiltration. When the residual pores were small enough, the viscous PCS solution could not be effectively infiltrated into the consolidated body. At this time, the process should be stopped. It seems difficult to achieve a fully dense matrix using the present PIP process because of the difficulty in penetrating the polymer into small pores that exist in the converted matrix. A higher magnification of the fiber boundary was shown in Fig. 4(c). Fine physical compatability between the fibers and the matrix were achieved, for no obvious circular cracks were observed for the thermal mismatch of the two phases.

Typical stress–displacement curves derived from the bending test for the composites infiltrated with slurries containing various SiC content are shown in Fig. 5. The composite infiltrated with high-concentration slurry displays not only higher failure stress but also higher elastic modulus, as observed from the linear stage of the curves. These mechanical properties may be attributed to the relatively densely formed matrix and tight bonding between the fibers and matrix, so that higher failure stress and elastic modulus could be achieved [12,13]. Stress–displacement curves for composites infiltrated with SiC filler demonstrate a peseudo-ductile fracture behavior. After reaching the maximum value, the load decreases gradually. This characteristic might be ascribed to loosely formed matrix compared to that of composites prepared by HP or CVI. Even though weak interface is beneficial for fiber bridging and fiber pullout. It is simultaneously detrimental for strength because of the low load transfer ability from the matrix to fibers through the weak interface.

Fracture surface after bending test for composite infiltrated with 40 wt.% slurry is shown in Fig. 6. The fracture surface demonstrates sound fiber pullout and fine-integrity of the fibers, indicating no serious damage to the fiber occured during the pyrolysis process. In the PIP-derived composite, since the density is relatively low and the matrix is loosely formed, cracks easily propagate along the weak region, which leads to fiber debonding from the matrix and then fiber pullout.

4. Conclusions

2D C_f/SiC composites were fabricated by PIP process, using slurries containing various contents of SiC filler in the first infiltration step. The SiC filler content has a significant influence on the physical and mechanical properties of the composites. The maximum failure stress, fracture toughness of the composites with particulate loading have reached $\approx$232 MPa and $\approx$10 MPa $m^{1/2}$, respectively. The composite pyrolyzed at 1100 °C has relatively weak interface and long fiber pullout dominated the fracture surface. Further work will be performed on preforms with increased fiber fraction to enhance the mechanical properties to fit the requirements for preparing strong and/or tough composites by the present PIP process.

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