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Short communication

The fabrication of 2D C_f/SiC composite by a modified PIP process using active Al powders as active filler

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ABSTRACT

2D C_f/SiC composites were fabricated by a modified polymer infiltration and pyrolysis (PIP) process. The effect of multilayered PyC on the mechanical and physical properties of the composites was investigated. The space in the fibrous preform was first filled with SiC and Al powder by a pressure infiltration process to enhance the ceramic yield. The results indicate that Al incorporation decreased the chances of micro-crack formation in the matrix. The multilayered PyC interphase was more effective in property modification, in comparison with the single pyrocarbon interphase of the same thickness.

1. Introduction

Continuous fiber reinforced ceramic matrix composites (CFCCs) show superior performance to monolithic ceramics under severe conditions such as high-temperature and high-stress applications in aerospace, hot engine and energy conversion facilities. In particular, the C_f/SiC composites have been investigated in previous studies. They have several unique advantages such as relatively low cost, large scale production and good thermal stability at elevated temperature.

The characteristics of fiber/matrix bonding has significant influence on the mechanical properties of the composites [1]. A third phase is usually deposited on the fiber surface before composite fabrication to obtain sound interfacial characteristics. Pyrocarbon (PyC) is the most commonly used interphase material and is deposited by chemical vapor deposition (CVD) for the layered microtexture parallel to the fiber axis. However, the PyC microstructure is rather disorganized [2], which logically results in difficulty for crack deflection inside the single PyC interphase. The nanoscale PyC/SiC multilayered interphase deposited by pressure-pulsed chemical vapor infiltration (PCVI) has been previously investigated by Bertrand and coworkers [2,3]. Crack deflection which is beneficial for the toughening mechanism, was observed in the PyC sublayers. To our knowledge, no studies on multilayered PyC interphase have been reported in the literature, although crack deflection can also be expected in such an interphase.

Recently, studies on CFCCs fabricated by polymer infiltration and pyrolysis (PIP) have been widely reported [4,5]. A major drawback for the process is the large volume shrinkage of up to 60% and ceramic yield of usually lower than 70% during pyrolysis process. A new approach of active-filler-controlled

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polymer pyrolysis (AFCOP) by the incorporation of active filler into the preceramic polymer has been raised [6]. The volume expansion caused by nitridation and carburization of the active filler on polymer pyrolysis can compensate for the volume shrinkage of the polymer to some extent, decreasing the density of microcracks formed in the matrix.

The present research work involved the fabrication of 2D C_f/SiC composites by a modified PIP process, using SiC as inert filler and Al as active filler. Single and multilayered PyC interphases were deposited to investigate the effect on the mechanical properties of the composites.

2. Experimental Procedure

2.1. *Composite Manufacturing*

2D 0°/90° woven C fiber fabrics (Xinka Carbon Co., Shanghai, China) were stacked to prepare the fiber preforms which were used for composite fabrication. The fiber volume fraction was controlled at about 30%.

The preforms were PyC and then SiC coated by isothermal chemical vapor infiltration (ICVI). More details about the apparatus used for PyC and SiC deposition can be found in [1]. Two kinds of PyC interphases were deposited. The first type consisted of a single PyC interphase of 200 nm. The second type was made out of four layers of PyC interphases, the thickness of each layer being about 50 nm. The curve of deposition temperature and time for multilayered PyC interphase is shown in Fig. 1. Hexamethyldisilazane (HMDS) was employed as precursor for deposition of SiC coating around PyC interphase. The thickness was about 200 nm.

The polymer precursor employed for the matrix was polycarbosilane (PCS). SiC powders (Germany) with an average grain size of 0.5 μm and Al powders (Liaoning, China) with an average grain size of 4 μm were utilized as the filler material. The coated preforms were first dipped into a slurry containing 20 wt.% SiC powders, 20 wt.% Al powders and 10 wt.% polycarbosilane (PCS) in vacuum. After drying, the samples underwent a high-temperature pyrolysis process in nitrogen atmosphere.

Subsequently, the samples underwent PIP process using PCS as precursor. The pyrolysis process was conducted at 1200 °C to convert the polymer to ceramic matrix. The above process was repeated six times in order to increase the final density of the material.

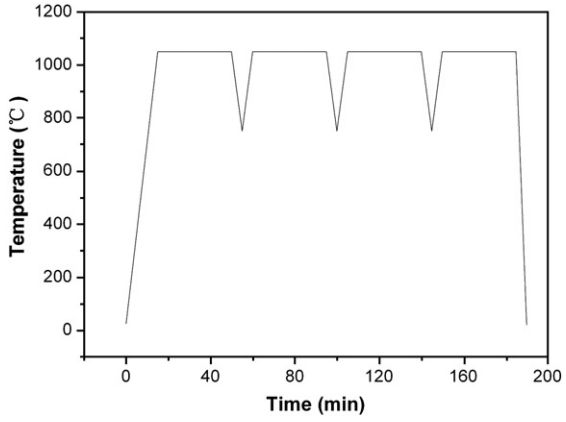

Fig. 1 – Curve of deposition temperature versus time.

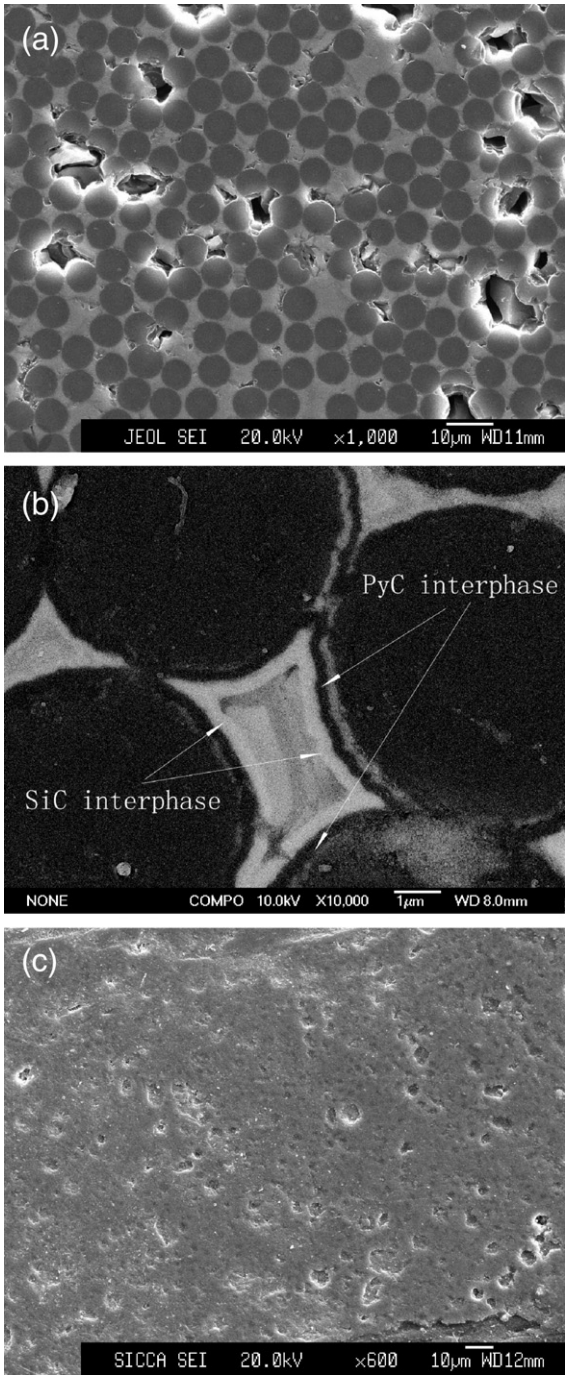

Fig. 2 – SEM micrographs of the polished cross sections.

2.2. *Characterization*

The fabricated composites were cut and ground into 2.5 mm × 4 mm × 36 mm samples for density, open porosity and three-point bending test. The density and open porosity of each sample were measured by the Archimedes method. Three-point bending test was performed on the Instron 5566 universal

testing machine, with a cross-head speed of 0.5 mm/min and a span of 24 mm. For each test, five samples were tested. The composite microstructure was investigated by scanning electron microscopy (SEM) of the polished cross sections.

3. Results And Discussion

Fig. 2 shows the cross-sectional micrographs of the fabricated C_f/SiC composite. As shown in Fig. 2(a), some isolated small pores could be observed in the intra-bundle areas even after several infiltration-pyrolysis cycles. This is a commonly observed phenomenon in the PIP-derived samples. These dispersed residual pores 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 thus hindering any further polymer infiltration. When the residual pores were small enough, the viscous PCS solution could not be effectively infiltrated into the consolidated body. It is advisable to stop the process at this point. In Fig. 2(b), SEM image of higher magnification shows the microstructure of the fiber boundary, the deposited PyC and SiC interphase, as well as the derived matrix. As shown in Fig. 2(b), the thickness of the deposited PyC and SiC interphase were about 200 nm. No obvious circular cracks around the fiber surface were observed, as commonly observed in the uncoated C_f/SiC composite [7]. These cracks result from the thermal expansion mismatch of the fiber and matrix. The PyC interphase helps to modify the thermal mismatch between the fiber and matrix. At the same time, the Al particles reacted with the carbon-containing volatile fragments from polymer decomposition and the reactive atmosphere, which led to formation of new phases such as Al_4C_3 and AlN. As reported by Greil [8], a specific volume expansion of 53% and 26% can be achieved for conversion of active Al with gaseous species to Al_4C_3 and AlN, respectively. Thus, the expansion of the infiltrated Al particles during pyrolysis process will compensate for the volume shrinkage caused by polymer-to-ceramic conversion and chances for micro-crack formation can be extensively reduced. The derived inter-bundle matrix is shown in Fig. 2(c). No obvious cracks were observed.

Table 1 lists the physical and mechanical properties of the two composites. Bulk densities of the two composites with multilayered and single PyC interphases were 1.72 and 1.71 g/cm^3, respectively, indicating that PyC microstructure has no effect on the infiltration process and final density. Bending test results at room temperature suggest that the composites with multilayered PyC interphases exhibited a relatively higher strength of 211 MPa, about 12% higher than that of composites with a single PyC interphase by calculation. This phenomenon demonstrates that the multilayered PyC interphase was more resistant to fracture behavior.

Table 1 – Effect of interphase microstructure on the physical and mechanical properties

Interphase	Density (g/cm^3)	Porosity (%)	Bending strength (MPa)	Failure displacement (mm)
Single	1.71 ± 0.02	27	188 ± 11	0.28
Multiple	1.72 ± 0.03	27	211 ± 13	0.33

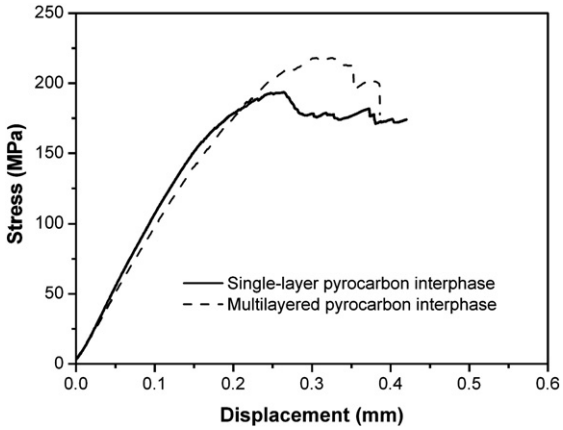

Fig. 3 – Stress/displacement curves for the fabricated composites with single-layer and multilayered PyC interphases.

Typical stress-displacement curves derived from the bending test for the two composites are shown in Fig. 3. It is observed that the composites with multilayered PyC interphase displays a higher strength but a slightly lower modulus of elasticity which can be observed from the slope of the linear stage in the stress-displacement curves. For the two composites, bending strength decreased gradually after the peak value. This characteristic might be ascribed to the relatively loose matrix and weak interphase material, providing the lower elastic modulus. Even though the weak interphase material is beneficial for fiber pullout and crack bridging, it is simultaneously detrimental for load transfer from the matrix to the fibers through such a weak interphase [9]. For the multilayered composites, a still weaker interfacial bonding was achieved, so a slightly lower slope was observed for the initial stage of linear deformation, demonstrating relatively lower elastic modulus. The matrix cracks might be deflected within the multilayered PyC interphase, and the shear stress in the crack tip can be reduced, which hampers the abrupt failure of the fiber bundles near the initial matrix cracks. At the same time, the lengthened cracks resulting from crack deflection have larger capability of deformation-energy absorption in the fracture process. Therefore, the composite with multilayered PyC interphase has a slightly higher bending strength than that with a single one. Considerable failure displacement was achieved for both composites.

4. Conclusions

2D C_f/SiC composites, with two kinds of PyC interphases were fabricated by slurry infiltration and PIP process. The high magnification micrograph of the fiber boundary confirmed that a homogeneous deposition of PyC and SiC interphases was achieved by the ICVI process. The microstructure of the PyC interphase has noticeable effect on the flexural strength

of the fabricated composites. The matrix cracks might be deflected in the multilayered PyC interphase. The deflected cracks result in reduced shear stress in the crack tip and lengthened cracks, which avoids abrupt failure of fiber bundles and facilitates absorption of deformation energy. As a result, the multilayered composites exhibited a slightly higher strength of 211 MPa. Volume expansion caused by conversion from active Al fillers to Al_4C_3 and AlN results in a reduction of matrix shrinkage during the pyrolysis process, which reduces the formation of pyrolysis-caused microcracks in the matrix and between the fiber surface and the matrix.

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