# MICROSTRUCTURE OF 3D TEXTILE C/PYC/SIC COMPOSITES PREPARED BY CHEMICAL VAPOR INFILTRATION

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## Introduction

Ceramic matrix composites (CMCs) is one of the most promising candidates for high temperature structural material, in which SiC matrix exhibits an excellent oxidation resistance because of the formation of a protective layer of silica which limits further oxidation [1]. CMCs with continuous fibers exhibit attractive properties such as low densities, high elastic moduli, and high strengths at elevated temperatures. They are considered as materials with a high potential in various fields of applications including engines and reentry thermal protection for spacecrafts. Chemical vapor infiltration (CVI) processed SiC matrix composites reinforced with carbon or SiC fibers have been designed and developed to improve the damage tolerance of inherently brittle SiC monolithic ceramics while retaining their high stiffness, high-temperature strength, and oxidation resistance [1-3]. The 3D-C/SiC composites with pyrolytic carbon (PyC) interfacial layer exhibit good mechanical properties and a typical failure behavior involving fibers pull-out and brittle fracture of sub-bundle [2]. In this paper the microstructure of the 3D textile C/PyC/SiC composites prepared by chemical vapor infiltration had been investigated.

## Experimental

The 3D textile C/PyC/SiC composites were fabricated by chemical vapor infiltration. Chemical vapor infiltration is a good route for the densification of high-temperature composites primarily ceramic-matrix composites [3]. It is well known that the mechanical properties of CMCs depend upon the fibers/matrix bonding. So the interposition of a thin layer of a compliant material such as carbon or BN is deposited on the fibers [1-3]. In this study the carbon layer was employed as the interphase. An isothermal CVI furnace (shown in Figure 1) was used to deposit pyrolytic carbon layer (interphase) and silicon carbide matrix. The carbon fibers utilized were T300 carbon fibers from the Nippon Toray Corporation. The three dimensional (3D) fabric preform was braided by four-step

processing and supplied by Nanjing Institute of Glass Fiber in China. The structure of the preform is illustrated in Figure 2. The fiber volume fraction was 40%. A thin pyrolysis carbon (PyC) layer (0.2± μm) was firstly deposited on the surface of carbon fibers as the interfacial layer with $C_3H_6$ at 850℃ and 0.1MPa. Methyltrichlorosilane ($CH_3SiCl_3$ or MTS) was used for the deposition of the silicon carbide matrix because it contains the same number of silicon and carbon atoms in one MTS molecule and thus can easily prepare stoichiometric silicon carbide. MTS, used as the precursor, is a liquid at room temperature (boiling point at 1 atm, 66.4℃). It was kept in a temperature-regulated container through which hydrogen was bubbled, carrying the MTS into a mixing chamber and then into the CVI furnace. The conditions used for SiC deposition are 1100 ℃, a hydrogen to MTS ratio of 10 and a pressure of 0.1MPa. Argon was used as dilute gas to slow the reaction rate.

The fracture surface morphology and cross-sectional microstructure of the 3D-C/SiC composites were observed using scanning electronic microscopy (SEM, Model JEOL 840). X-ray diffraction (XRD) measurements were made with a Rigaku D/Max-B diffractometer unit using Ni-filtered CuK$_\alpha$ radiation at a scanning rate of 0.5°·sec$^{-1}$ and scanning from 15° to 80° of 2θ.
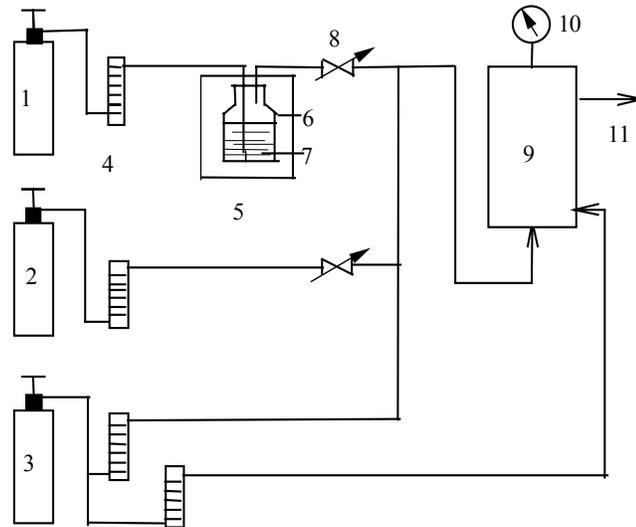


Figure 1. Schematic diagram of isothermal chemical vapor infiltration furnace
1. $H_2$; 2. $C_3H_6$; 3. Ar; 4. flow meter; 5. thermostatic box; 6. container of MTS; 7. MTS; 8. valve; 9. CVI furnace; 10.pressure/vacuum meter; 11. waste gas

**Results and Discussion**

XRD pattern of the 3D textile C/PyC/SiC composites cross-sectional fracture surface is shown in Figure 3. Detailed analysis of the X-ray results indicated that the deposition are carbon and silicon carbide composed mainly of cubic (3C) type β-SiC with a small amount of 4H type α-SiC. It is clear that the diffraction angles of 35.6°, 60.1°, 72.1° and 75.5° correspond to β-SiC with cubic crystal structure and the diffraction angles of 33.7°

corresponds to $\alpha$-SiC with the hexagonal crystal structure [4]. According to the breadth of diffraction peaks, the crystallite sizes of silicon carbide was calculated from the Scherrer equation, $D = 0.89\lambda/\beta COS(\theta)$, Where $\lambda$ is the wavelength of characteristic X-ray, $\theta$ is the Bragg angle, and $\beta$ is the calibrated width of the half-height of diffraction peaks. The crystallite sizes of silicon carbide is 10~15nm.
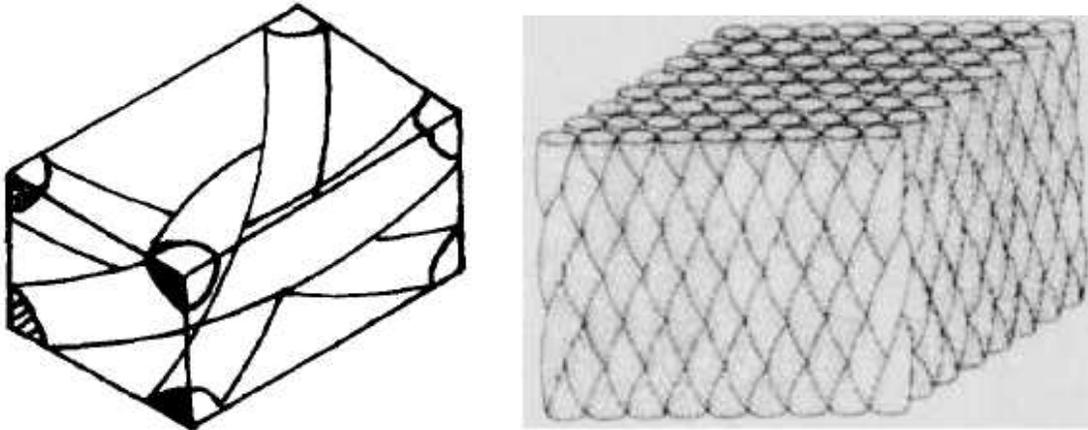


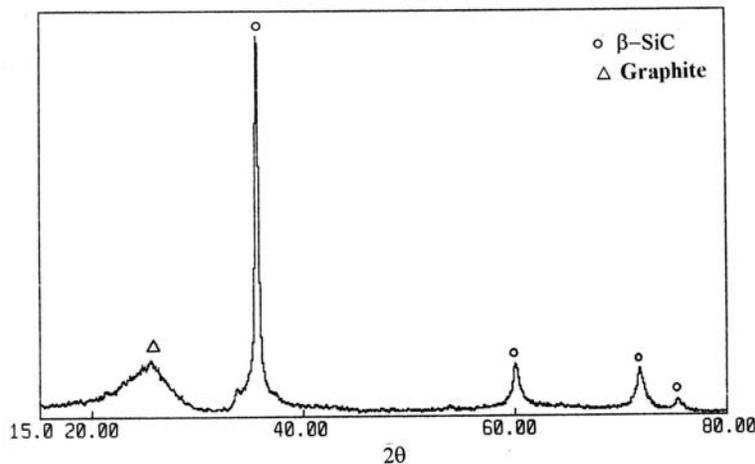Figure 2. Structure of three-dimensional braiding preform



Figure 3. XRD pattern of the C/PyC/SiC composites cross-sectional fracture surface

The flexural strength and flexural elastic modulus of the composites were measured using the three-point-bending method. The flexural strength of the 3D textile C/PyC/SiC composites was 453Mpa, flexural fracture strain, 0.58%, and work of fracture, 25.2kJ•m$^{-2}$. The microstructure of the 3D textile C/PyC/SiC composites is shown in Figure 4-5. The fracture surface of the C/PyC/SiC composites is shown in Figure 4. Figure 5(a) and Figure 5(b) show the microstructure of fracture surface. From the XRD pattern, it is easy to determine that SiC matrix consists of a large number of nanometric SiC crystallites. The failure behavior of the composites was attributed to the interfacial bonding between fiber and matrix. It was the PyC interface layer that ensured the

proper interfacial bonding between carbon fiber and matrix, as well as the load transfer from silicon carbide matrix to the carbon fiber. Because of the difference of the thermal expansion coefficients between the silicon carbide matrix and the carbon fiber, a tensile stress was generated cross the PyC interfacial layer. As a result, it was easy for the fiber to be pulled out from the silicon carbide matrix (Figure 5b). For the 3D textile CMCs, the bundle/bundle interfacial bonding is usually considered as a kind of weak interfacial bonding because of the pores residual in the composites, caused by the "bottom neck effect" during the chemical vapor infiltration process. Accordingly fiber bundle pullout was always observed (Figure 4 and Figure 5a).



Figure 4. Images of fracture surface of 3D textile C/PyC/SiC composites
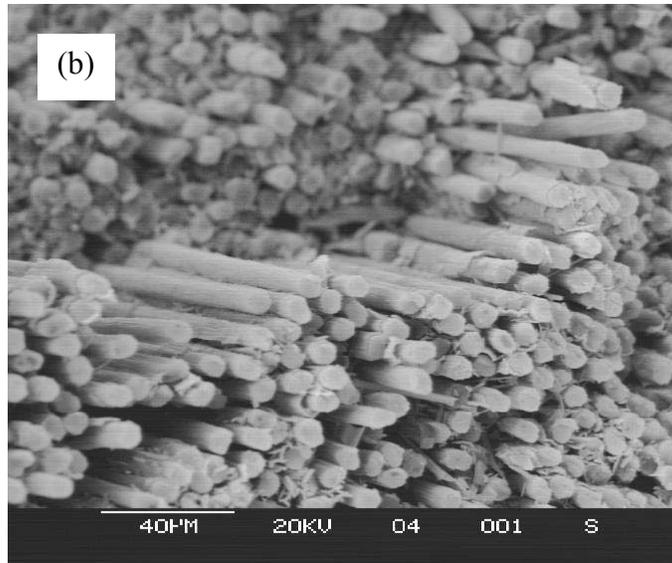


(a)

1MM    19KV    04    001    S
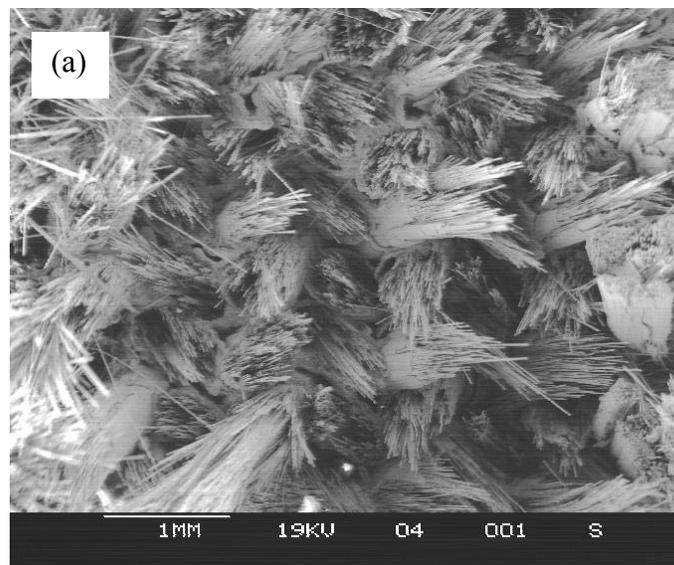
Figure 5. Images of cross-sectional fracture surface of 3D textile C/PyC/SiC composites

**Conclusions**

(1) The 3D textile C/PyC/SiC composites with pyrolytic carbon interfacial layer were fabricated by chemical vapor infiltration. A thin pyrolysis carbon layer was firstly deposited on the surface of carbon fibers as the interfacial layer with $C_3H_6$ at 850℃ and 0.1MPa. Methyltrichlorosilane ($CH_3SiCl_3$ or MTS) was used for the deposition of the silicon carbide. The conditions used for SiC deposition are 1100℃, a hydrogen to MTS ratio of 10 and a pressure of 0.1MPa.

(2) The flexural strength of the 3D textile C/PyC/SiC composites was 453Mpa, flexural fracture strain, 0.58%, and work of fracture, 25.2kJ•m$^{-2}$.

(3) Because of the difference of the thermal expansion coefficients between the silicon carbide matrix and the carbon fiber, a tensile stress was generated cross the PyC interfacial layer. It was easy for the fiber to be pulled out from the silicon carbide matrix. Fiber bundle pullout was always observed because of the pores residual in the composites, caused by the "bottom neck effect" during the chemical vapor infiltration process.

**References**

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