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Effect of Temperature on the Synthesis of SiC Coating on Carbon Fibers by the Reaction of SiO with the Deposited Pyrolytic Carbon Layer

Hejun Li†, *Haibo Ouyang, Lehua Qi, Yulei Zhang, Zhengjia Li and Jianfeng Wei*
C/C Composites Research Center, Key Laboratory of Ultrahigh Temperature Composites, Northwestern Polytechnical University, Xi′an 710072, China

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The influence of reaction temperature on the preparation of SiC coating on carbon fibers by the reaction of silicon monoxide with the deposited pyrolytic carbon (PyC) layer has been discussed. With rising reaction temperature, the thickness of SiC layer increases and the SiC grain is coarsening. The apparent activation energy for the synthesis of SiC layer is about 103.3 kJ/mol. The oxidation resistance of carbon fiber can be improved by the SiC/PyC layers significantly. The initial oxidation temperature of the SiC/PyC coated carbon fiber is about 300°C higher than that of the uncoated carbon fiber. The oxidation of the SiC/PyC coated carbon fiber is owing to the diffusion of oxygen through the cracks generated by the mismatch of thermal expansion.

KEY WORDS: Carbon fiber; SiC coating; Pyrolytic carbon; Oxidation behavior

1. Introduction

Carbon fibers are the most significant reinforcement applied in advanced composites, due to their high specific strength, modulus, and low thermal expansion coefficient[1,2]. However, the sensitivity to oxidation restricts the wide application of carbon fibers in composites. In the development of the oxidation protection of carbon fibers, SiC coating has been considered to be one of the best candidates, due to its effectiveness in the oxidation resistance and the suppression of the inter-diffusion and possible chemical reactions between carbon fiber and matrix[3–5].

Various methods have been developed for preparing SiC coating on carbon substrates, such as chemical vapor deposition (CVD) and carbothermal reduction[6–9]. However, the reaction between silicon compound and carbon fiber would damage the fiber and deteriorate the mechanical properties of fibers[3,10]. To protect carbon fibers in the deposition of SiC, pyrolytic carbon (PyC) inner layer is commonly employed[11–13]. Moreover, PyC can act as an interlayer to compensate for the difference in thermal expansion between the ceramics coating and substrates, which is of benefit for improving the oxidation resistance of carbon fibers[14,15]. Therefore, PyC/SiC double layer coating is suggested to apply in the carbon fiber reinforced composites[3]. In our previous work, the preparation of SiC/PyC multilayer coated carbon by the reaction of silicon monoxide with the pyrolytic carbon layer was presented[16]. The effect of holding time on the synthesis of SiC coating and the formation mechanism of it have been discussed.

In this paper, the influence of reaction temperature on the microstructure, morphology of the SiC/PyC layers coated carbon fiber was discussed. The reaction kinetics for the synthesis of SiC layer was investigated. In addition, the oxidation behavior of the uncoated, PyC coated, and SiC coated carbon fiber were studied.

† Corresponding author. Prof., Ph.D.; Tel.: +86 29 88495004; E-mail address: lihejun@nwpu.edu.cn (H.J. Li).

2. Experimental

2.1 Synthesis of SiC/PyC *multilayer on carbon fibers*

Polyacrylonitrile-based short carbon fibers, with a length of 3–5 mm and diameter of 6 μm, were selected for the deposition of PyC. The PyC layer on carbon fibers was prepared by isothermal chemical vapor infiltration (ICVI) using methane as the precursor and nitrogen as the diluted gas at ambient pressure. The deposition of the PyC layer was carried out at 1100°C for 1 h with the methane partial pressure of 30 kPa. Subsequently, the carbon fibers deposited with PyC layer were put into a vacuum sintering furnace for the synthesis of SiC coating by the reaction of SiO and deposited PyC layer. A graphite crucible was used as a sample holder. The mixture of SiO_2 and Si powder was put on the bottom of the graphite crucible. Fibers were placed on a carbon felt over the mixture powder. At the elevated temperature, the mixture powders reacted and released SiO vapor. Then, the SiO reacted with the pyrolytic carbon layer to synthesize the SiC. The holding time for the experiments was 1 h. The pressure of the furnace was maintained 20 Pa in the heating process.

2.2 Characterization of SiC/PyC *coated short carbon fibers*

X-ray diffraction (XRD) was used for phase analysis of the samples. Prior to XRD, the carbon fibers were ground into powder by ball milling. XRD measurements were performed on PANalytical X-Pert Pro diffractometer (Netherlands) with Cu$K\alpha$ monochromatic radiation (λ=0.15406 nm). The character of the crystalline phases presented in the sample was checked using the data base of the Joint Committee on Powder Diffraction Standards (JCPDS). The morphology of the SiC/PyC coated carbon fibers were characterized by field emission scanning electron microscopy (FESEM, JSM-6700F, JEOL, Japan).

The oxidation behavior of uncoated and coated fibers was investigated by thermo gravimetric analysis with differential scanning colorimetry (TGA/DSC, Mettler Toledo, Switzerland) system. About 10 mg of fibers were placed in alumina crucibles and heated to 1250°C. The elevating rate of the temperature is about 10°C/min. The synthetic air with a flow rate of 50 mL/min was employed as oxidation agent.

3. Results and Discussion

3.1 Phase analysis

Figure 1 shows the XRD patterns of the SiC/PyC coated carbon fibers prepared at different reaction temperatures. It is obvious that only carbon and β-SiC diffraction peaks can be observed in these patterns. The peaks at approximately 26 and 42.6 deg. correspond to (002) and (100) peaks of carbon.

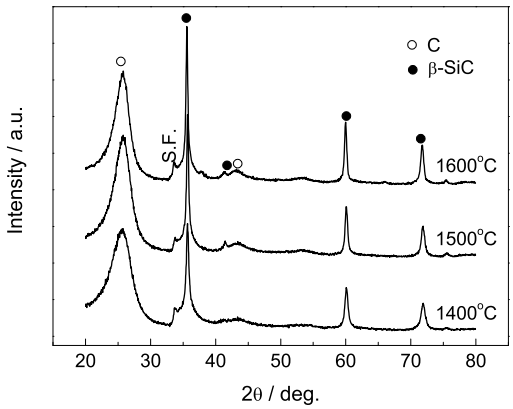

Fig. 1 XRD patterns of the SiC/PyC coated carbon fibers prepared at different reaction temperature

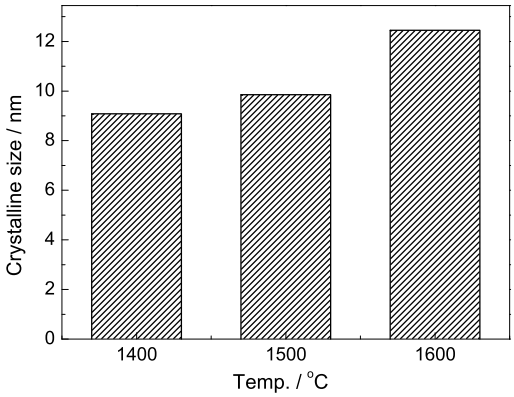

Fig. 2 The variation of crystalline sizes of SiC with reaction temperature

The peaks at approximately 35.8, 41.3, 60.0 and 72.0 deg. correspond to (111), (200), (220), and (311) peaks of β-SiC, which agrees with the standard value for β-SiC (JCPDS Card No. 0029-1129). The diffraction peak of 33.5 deg. marked as S.F. is due to stacking faults in β-SiC layers, which was also observed in the work of Yang *et al.*[17] and Wu *et al*[18]. The relative intensity of the characteristic peaks of β-SiC increases with the elevation of the reaction temperature. It is indicated that the conversion of PyC into β-SiC increases with the elevation of the reaction temperature.

Moreover, the full width at half maximum of diffraction peak of the characteristic peaks of β-SiC (111) becomes narrow with the elevation of the reaction temperature. The crystallite sizes of silicon carbide are calculated from the Scherrer equation[19,20],

$$D = \frac{K\lambda}{\beta \cos\theta} \tag{1}$$

where D is the crystallite size, λ (0.15406 nm) is the wavelength of characteristic X-rays, θ is the Bragg angle, K is the Scherrer constant, and β is the calibrated full width at half maximum of diffraction peaks. The variation of crystalline sizes with reaction temperature is shown in Fig. 2. With the deposition temperature increasing from 1400 to 1600°C, the crystallite