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Thermal stability of three-dimensional carbon fiber reinforced Si–O–C composites fabricated by polysiloxane pyrolysis

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Continuous fiber reinforced ceramic matrix composites (CFRCMCs) show promise for overcoming the brittleness of monolithic ceramics and are attractive materials for applications requiring specific weight and high strength and toughness at elevated temperatures. Generally, there are several methods to fabricate CFRCMCs, such as chemical vapor infiltration (CVI), slurry infiltration combined with hot-pressing, reaction bonding, and polymer-infiltration-pyrolysis (PIP), etc.

The PIP route is gaining increasing attention in recent years for its advantages such as low processing temperature, controllable ceramic compositions, and near-net-shape technologies. Many different preceramic polymers have been discovered since their original development by Yajima *et al.* [1]. Of all the precursors for PIP, polysiloxane (PSO) has attracted increasing interest in recent years [2]. It is not only commercially available but also very cheap, and its derived silicon oxycarbide (Si—O—C) (in Ar or N_2) and silicon oxynitride (Si—N—O) (in NH_3) ceramics have been demonstrated to possess improved properties such as creep resistance and microstructural stability over those of many conventional silicate ceramics [3, 4]. Therefore, PSO is considered as a desirable precursor for low-cost CFRCMCs with high performance.

In our previous studies [5, 6], the curing and pyrolysis behaviors of PSO were investigated and three-dimensional braided carbon fiber reinforced Si—O—C (3D-B C_f/Si—O—C) composites with high mechanical properties were fabricated by PIP route. The present work focuses on the thermal stability of 3D-B C_f/Si—O—C composites under vacuum.

The raw materials used to fabricate 3D-B C_f/Si—O—C composites and the fabrication process were described in detail in Ref. [5], and the three-point bending strength at room temperature of the composites is shown in Table I. The flexural strengths of the composites at different temperatures under vacuum were tested by a three-point bending method, with the span/height ratio of 15 and a cross-head speed of 0.5 mm/min. The specimen bars with a size of $3 \times 4 \times 60$ mm were heated under vacuum at a heating rate of 40 °C/min to the preseted temperatures. Load was applied to the specimen bars after they were soaked at the preseted temperatures for 10 min to make temperature uniform throughout the specimen bar. The results are presented in Table I. Each data point was an average over three values.

As shown in Table I, the flexural strength of 3D-B C_f/Si—O—C composites can be maintained to 1400 °C under vacuum. The flexural strength of 3D-B C_f/Si—O—C composites at 1300 °C is 1.21 times of that at room temperature.

Figs 1–4 show the fracture surfaces of 3D-B C_f/Si—O—C composites after tested at different temperatures. In Figs 1–3, samples A, B, and C illustrate extensive fiber pullout and smooth surfaces of pulled-out fibers are observed. After tested at 1500 °C, it can be seen from Fig. 4 that carbon fibers are coated by a layer of glassy matter. In addition, it is noted that there is little matrix among carbon fibers in sample D while dense matrix can be observed among fibers in samples B and C.

Previous researches [6–8] showed that the matrix derived from polysiloxane pyrolysis at 1000 °C consisted of free carbon and silicon oxycarbide structure, which was a three-dimensional random network of silicon–oxygen tetrahedral with some silicons bonded to one or two carbons substituted for oxygen, which were in turn tetrahedrally bonded to other silicon atoms. When the temperature was elevated above 1200 °C, the pyrolyzates of polysiloxane would gradually transform into SiO_2, SiC, SiO, and CO as the results of the redistribution reaction of silicon oxycarbide structure and the carbonthermal reduction reaction between free carbon and SiO_2. However, the redistribution and the carbonthermal reduction were mild below 1400 °C, creating little SiO_2, SiO, and CO. As a result, the density of 3D-B C_f/Si—O—C composites did not decrease much. At the same time, carbon fibers were hardly chemically corroded by the SiO_2 in matrix. Thus, the interfacial bonding did not become strong and the tensile strength of carbon fibers did not decrease. Therefore, the flexural strength of 3D-B C_f/Si—O—C composites

TABLE I The flexural strengths of 3D-B C_f/Si—O—C composites at different temperatures under vacuum

Sample	Testing temperature (°C)	Flexural strength (MPa)
A	Room temperature	502.0
B	1300	608.6
C	1400	527.0
D	1500	331.0

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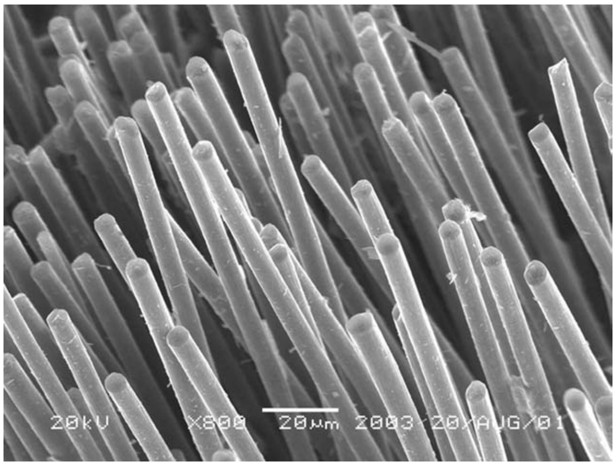

Figure 1

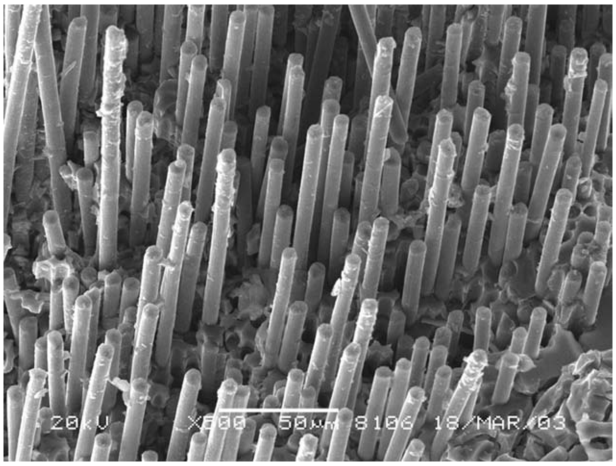

Figure 2

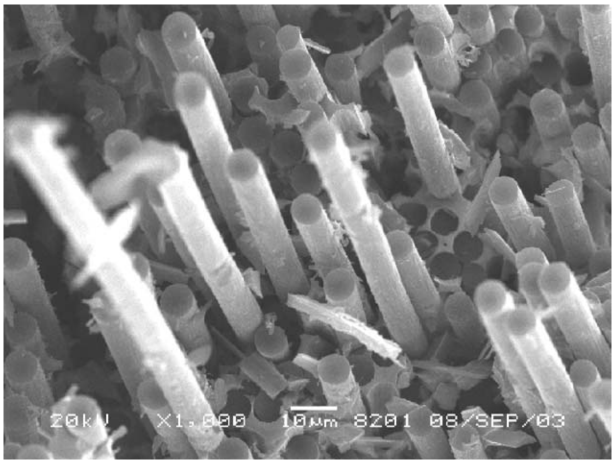

Figure 3

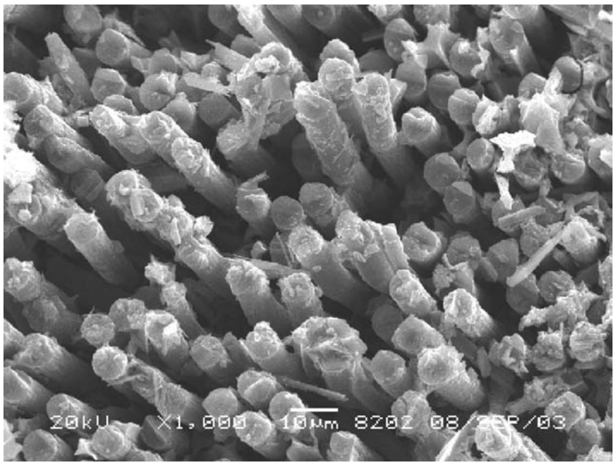

Figure 4

was maintained to 1400 °C. That the flexural strength at 1300 °C is higher than that at room temperature needs further investigation. When temperature was elevated from 1400 to 1500 °C, the redistribution and the carbon-thermal reduction became fierce, creating much SiO_2, SiO, and CO. The evolution of much gaseous SiO and CO remarkably decreased the density of composites. At the same time, the created viscous SiO_2 adhered to carbon fibers and the reaction between carbon fibers and SiO_2 occurred, lowering the tensile strength of carbon fibers remarkably. Because of the remarkable decrease in density and tensile strength of carbon fibers, the flexural strength of 3D-B C_f/Si–O–C composites at 1500 °C was only 65.9% of that at room temperature.

In summary, the flexural strength of 3D-B C_f/Si–O–C composites fabricated by polysiloxane pyrolysis can be maintained to 1400 °C under vacuum. With increasing temperature from 1400 °C, the flexural strength will decrease sharply.

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