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Processing and flexural properties of 3D, seven-directional braided $(SiO_2)_f/SiO_2$ composites prepared by silica sol-infiltration-sintering method

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Abstract

Three dimensional (3D), seven-directional braided $(SiO_2)_f/SiO_2$ composites were prepared by silica sol-infiltration-sintering (SIS) method. The flexural properties were evaluated by three-point bending test. The flexural strength and flexural elastic modulus were found to be about 107 MPa and 17.5 GPa, respectively. The results of stress vs. deflection curve and SEM examination revealed that the fracture mechanism of the $(SiO_2)_f/SiO_2$ composite was a mixed mode of brittle and ductile. The bond strength of fiber/matrix was weak at low temperature, causing the extensive fiber pull-out. On the other hand, the brittle fracture of some fibers maybe caused by the propagation of micro defect or crack, which existed in the as-prepared composites for the ten-cycle process.

Keywords: $(SiO_2)_f/SiO_2$ composites; Silica sol-infiltration-sintering method; Flexural properties; Fracture mechanism

1. Introduction

Continuous fiber reinforced ceramic matrix composites (CFCCs), such as C/SiC and SiC/SiC systems, are considered as potential materials to be used in advanced aero-engines, space and fusion power reactors due to surface strength at elevated temperatures and higher fracture toughness compared with metal and monolithic ceramics respectively [1–4]. Silica has high melting point, combined with high thermal shock resistance and excellent ablation resistance as well as low thermal conductivity, and has been received considerable attraction in the field of Radome materials [5]. However, low flexural strength and low fracture toughness have restrained the use of monolithic silica for many applications, include the random structures. For the purpose of improving the mechanical properties, several efforts [6–12] have been made to reinforce SiO_2 matrix composites by adding shorter silica fiber, long silica fiber, 2.5D silica braided and conventional 3D silica braided reinforcement. Glass and silica fiber reinforced silica matrix foams have been developed through slurry based processing, which was found to be beneficial for the stiffness property of the composite foam [8]. Unidirectional silica fiber reinforced SiO_2 composite was prepared by pressureless sintering method, and this improved mechanical properties and such improvements were attributed to the fiber pull-out [9]. 2.5D $(SiO_2)_f/SiO_2$ composites were fabricated via sol–gel infiltration method, and the flexural strength of the composites was improved from 31 to 80 MPa [10]. It can be seen that the mechanical properties of SiO_{2f}/SiO_2 composites were closely related to the braided structure of reinforcement and the preparation method. There are only a few reports on 3D, seven-directional braided $(SiO_2)_f/SiO_2$ composites; the only study reported till date points to higher out-of-plane stiffness, strength and high damage tolerance [13].

In this paper, the preparation and processing details as well as flexural properties (strength and modulus) as well as mode of fracture of 3D, seven-directionally braided $(SiO_2)_f/SiO_2$ composites are reported and discussed.

2. Experimental procedures

The 3D, seven-directional braided silica reinforcements were provided by Sinamo Science and Technology Company Limited, Nanjing, China. The yarn specification was B type

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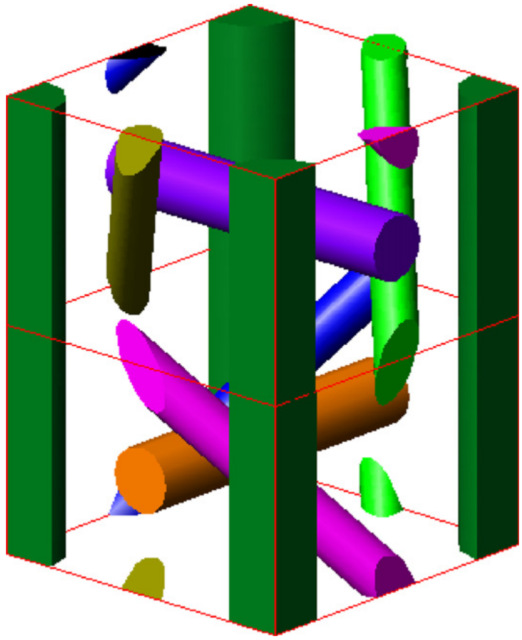

Fig. 1. Representative unit of 3D, seven-directional braided silica structure.

silica fiber (190Tex) and the fiber volume fraction was about 47%. Fig. 1 shows the representative unit structure of 3D, seven-directional braided silica.

The $(SiO_2)_f/SiO_2$ composites were fabricated by silica sol-infiltration-sintering method, as shown in Fig. 2. First, the 3D, seven-directional braided silica reinforcements were placed in a closed container and the container was evacuated to 0.1 Pa. Then, the high pure silica sol (volume ratio 35%, average particle size 10 nm, pH9 obtained from, Ningbo Company, China) was sucked into the container, and during which entire process, the pressure was maintained at 0.1 Pa. The container was then pressurised to 10 atm and maintained at that high pressure for 1 h. The composites were then dried at 80 °C for 1 h and 110 °C for 1 h to gradually remove water content of the gel solution. Then it was sintered at 450 °C for 2 h to remove the coupling agent and bounded water. The process was repeated for ten cycles to obtain the final composite.

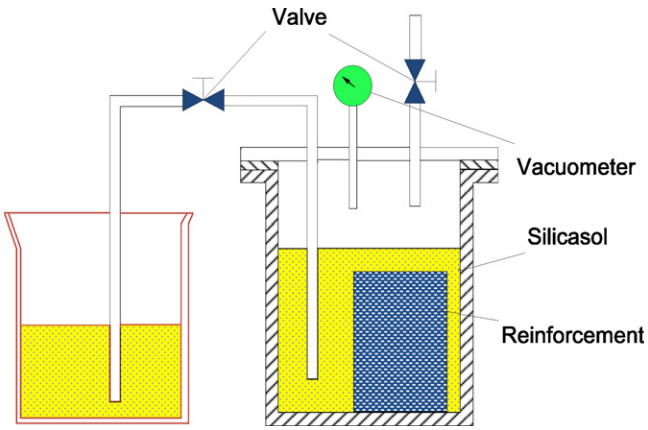

Fig. 2. Schematic diagram of silica sol-infiltration method.

Test specimens were cut parallel to the direction of fiber and then ground into bars of 40 mm in length, 5 mm in width and 3.5 mm in thickness. Three-point bend tests on an Iosipescu Universal tester (SANS CMT5105 electronic universal testing machine) were conducted with a displacement rate of 0.1 mm/min and 30 mm span according to British Standard Methods of Testing. The load vs. deflection data were recorded till fracture. The fracture strength and elastic modulus were computed from the load deflection data. Microstructure and fracture mechanism were examined by scanning electron microscopy (JEOL JSM-6360). The specimens for SEM were gold coated.

3. Results and discussion

Fig. 3 is stress vs. strain curve for a typical CFCC. The curve could be divided into three stages: elastic, damage and failure. At the elastic stage, the mechanical behavior of the composites is grossly linear elastic. The matrix cracks and other defects of manufacturing do not grow to cause any significant amount of damage at this stage and the composites attains a peak or maximum stress (σ_u) following this, damage in the form of a nonlinear region with increasing stress with deflection is mostly observed (please see the actual load/stress-deflection plots reported in the present CFCCs and also those reported in other studies) presented. During this damage stage, the matrix continuously cracks, and meanwhile the interface of fiber/matrix debonds. The matrix fails when the maxim stress is attained. At the failure stage, there are two different modes: A typical brittle/sudden fracture behavior, with no fiber pull-out and/or a typical ductile/gradual fracture behavior, with extensive fibers or bundles pull-out.

Fig. 4 shows the actual stress vs. deflection curve of 3D, seven-directional $(SiO_2)_f/SiO_2$ composite. The nature of deformation and fracture broadly correspond to the one described in the previous. The maximum stress the composite exhibits is about 87 MPa, indicating reasonably high strength of silica matrix. Fig. 5a shows the SEM fractographs of the initial silica matrix, presented large particles and good poly-crystallization indicating an effective SIS processing. At the damage stage, a nonlinear region, though of a limited extent is observed. The matrix failed when the maxim stress reached 107 MPa.

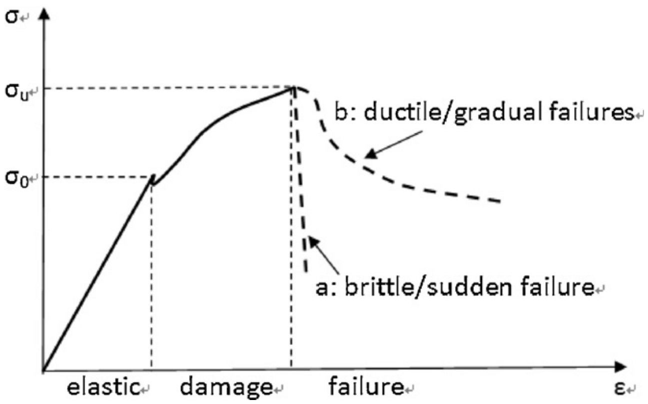

Fig. 3. Typical stress vs. strain curve of CFCCs.

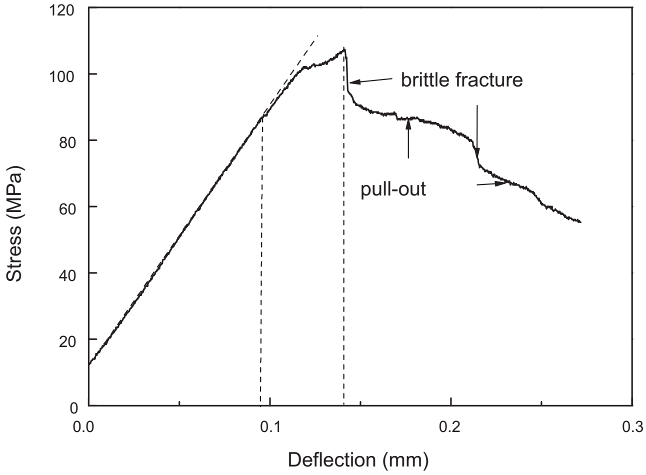

Fig. 4. Stress vs. deflection curve of $(SiO_2)_f/SiO_2$ composite.

After the attainment of maximum stress, the stress dropped suddenly, caused by the fibers fracture, indicating the mechanism was a brittle mode. However, further stress drop is found to be gradual. Then it dropped gradually, caused by the fibers pull-out, indicating the mechanism changed to a ductile mode. And the fibers were fractured and pulled out in turn until failure. It can conclude that the fracture mechanism of the composite was a combination of ductile and brittle. The flexural strength and elastic modulus of three-point bending test was about 107 MPa and 17.5 GPa, respectively.

In general, the microstructure governs mechanical properties and mode of fracture. The fracture surfaces of the composites were examined by SEM after three-point bending test. Fig. 5b was a SEM image of the fracture structure of the $(SiO_2)_f/SiO_2$ composite, which showed extensive fibers pull-out at low magnification. The improved toughness was obtained from a microstructure where the fibers can absorb significant energy extent of by bridging crack surfaces and dissipation energy as the matrix disintegrates during fibers pull-out. Long fibers pull-out were also observed on the high magnification SEM, as shown in Fig. 5b and c, indicating the composites exhibited a certain high extent toughness.

However, many flat fracture surfaces of fibers were also observed, as shown in Fig. 5c indicating that these fibers were fractured in a brittle mode. In the present braided composite, both fibers fracture and pull-out have been observed indicating the combination of brittle and ductile fracture, which is in accordance with the results of stress vs. deflection curve.

The purpose of the fiber/matrix interfacial materials is to provide a proper fiber/matrix interface. The functions of the interface are (1) to prevent damage matrix cracking from penetrating the fibers; (2) to maintain a good load transfer between the fibers and the matrix; (3) to act as a diffusion barrier and relaxation of residual stress [14]. If the bonding strength of fiber/

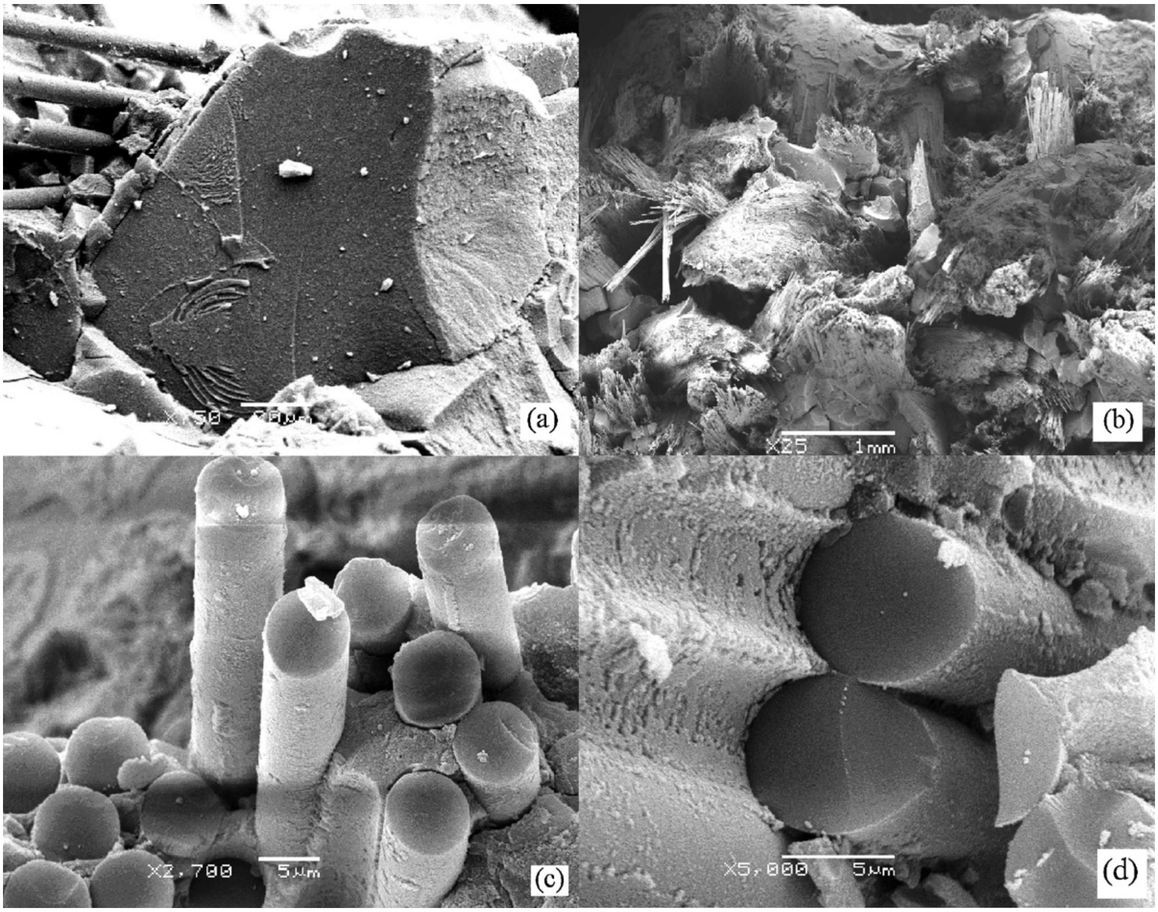

Fig. 5. SEM images of $(SiO_2)_f/SiO_2$ composite: (a) silica matrix; (b) fracture surface; (c) fibers pull-out; and (d) fiber peel-off.

matrix is weak, the fiber will be easy to pull-out, causing a good toughness and low fracture work. On the other hand, if the bonding strength of fiber/matrix is strong, the fiber will be difficult to pull-out, causing a brittle fracture and high fracture work.

The sintering temperature of the samples was maintained only at 450 °C, much lower than the values suggested in References [9] and [10], because of the silica fiber tends to get damaged after high temperature treatment [15]. The bonding strength of fiber/matrix would then be weak at low temperature, causing the extensive fibers pull-out as shown in Fig. 5a and b. Hence, the fabrication process of $(SiO_2)_f/SiO_2$ composites was repeated for ten-cycle process. During these repeated cycles, micro defects or cracks of the fibers were found to accumulate, causing a fiber peeling on further increase in stress, as shown in Fig. 5d.

4. Conclusions

3D, seven-directional braided $(SiO_2)_f/SiO_2$ composites were fabricated by silica sol-infiltration-sintering method. The flexural strength and flexural elastic modulus were found to be 107 MPa and 17.5 GPa, respectively. The results of stress-deflection data and SEM examination revealed that the fracture mechanism was a combination of brittle and ductile fracture. The bond strength of fiber/matrix was weak at low temperature, causing the extensive fibers pull-out. On the other hand, the extent of brittle fracture of some fibers may have increased due to fiber peeling that was found to occur with each step of ten-cycle process.

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