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Manufacturing isotropic carbon fibre preforms for multilayered silicon carbide composites with a pyrolytic carbon interphase

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ABSTRACT

In the present work, isotropic carbon fibre preforms with self-supporting porous structures and random orientation were fabricated through a wet papermaking filtration method. Then pyrolytic carbon (PyC) and silicon carbide (SiC) were alternatively infiltrated into the as-laminated preforms to obtain the multilayer composites (C_{sf}/SiC-C) by the chemical vapor infiltration method. The effects of fibre content and soft-hard multilayer matrix on mechanical properties were investigated. With the increase of fibre content, the bending strength and fracture toughness of composites without PyC inside matrix (C_{sf}/SiC) increase while the porosity decreases. When the fibre content is 10.2, 11.8, and 13.9 vol%, the bending strength of C_{sf}/SiC composites reaches the values of 170, 182, and 203 MPa, respectively. Furthermore, the composites exhibit good isotropy in mechanical properties. A model related to random orientation fibres was established to illustrate the relationship between bending strength, fibre content and porosity, which shows good accordance with the experimental data. Compared with C_{sf}/SiC, C_{sf}/SiC-C shows maximum increases of 17.3% and 18.3% in bending strength and fracture toughness, respectively. It is indicated by micrographs that the energy is consumed when cracks deflect by random orientation fibres and in multilayer of PyC and SiC, which provides a major contribution for improvement in mechanical properties.

1. Introduction

Silicon carbide (SiC) has been extensively studied in view of its low density, chemical stability, excellent high temperature mechanical properties and self-passivating behavior in oxidizing environment [1–4]. It shows gratifying application prospect in wear components such as mechanical face seal rings, journal bearings, valves, nozzles, rotors etc [5]. However, like other ceramics, its low fracture toughness remains a major concern for its wide application in severe environment.

Introduction of secondary phases is an effective method to ameliorate fracture toughness. Main secondary phase reinforcements include particulates, whiskers, continuous fibre and short fibre [6]. Continuous carbon fibre is a prevailing selection because of its promising mechanical properties and has been employed to reinforce SiC matrix. However, fabrication process of continuous fibre reinforced SiC composites tends to be time-consuming and very expensive [7]. Compared with continuous fibre reinforced composites, the short carbon fibre reinforced SiC composites (C_{sf}/SiC) have excellent adaptability to conventional manufacturing techniques and cost less to fabricate, and therefore are increasingly studied [5]. Hot pressing (HP), liquid silicon infiltration (LSI) and tape casting followed by pressureless sintering have been adopted to fabricate C_{sf}/SiC composite [8]. Hyuga et al. [9] obtained 98% densified C_{sf}/SiC by HP, in which the fracture toughness was enhanced, but the bending strength was weakened to some extent. Li et al. [10,11] used the method of LSI to produce C_{sf}/SiC composites. The fracture toughness reached a peak at 30 vol% of carbon fibres, but due to the existence of residual silicon, the mechanical properties worsened as the volume content of fibres was above 30%. Biamino et al. [1–14] obtained multilayered ceramics by tape casting followed by pressureless sintering. The fibres tended to align fairly well along the tape casting direction, so the properties of the multilayered ceramics were not isotropic. Moreover, interphase delamination between adjacent tapes was found in 20 vol% fibre content C_{sf}/SiC multilayer composites, which led to a constant decrease of bending strength with the increase of carbon fibre content.

Chemical vapor infiltration (CVI) is an effective method to produce carbon fibre reinforced SiC composites, where methyltrichlorosilane (MTS, CH_3SiCl_3) is used to infiltrate SiC matrix on porous preforms. It has several significant advantages including near-net final shapes, minimization of the mechanical damage of the fibres due to much lower pressures and temperatures in the range of 900–1100 °C, low mechanical stress, and higher purity of the matrix [15]. However, since self-

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supporting structures are indispensable to form unobstructed channels for vapor infiltration in the CVI process, the preforms of CVI method are mostly 2D continuous carbon fibre cross-woven fabrics or 3D continuous woven fabrics [16]. Although continuous woven fabrics reinforced composites possess excellent strength and toughness, their mechanical properties are anisotropic. Zhang et al. [17,18] found that the mechanical properties of 2.5D SiC matrix composites reinforced by continuous woven fibres are highly anisotropic. The interlaminar shear strength, modulus and ultimate tensile strength in the weft direction are 76.3%, 55.2% and 29.1% lower than those in the warp direction. Yang et al. [19] observed the anisotropy of 2D C/SiC woven composites. While the modulus along 45° off-axis is 24.5% less than that along axis, the tensile strength along 45° off-axis is 16.6% less than that along 15° off-axis. It is hopeful to overcome such anisotropy through using chopped carbon fibres to fabricate CVI preforms, because they could be well distributed with random orientation. However, few studies have been done to fabricate preforms for CVI process with chopped carbon fibres. Mathur et al. [20] prepared carbon paper for porous electrode in a fuel cell. The preparation process involved a fine dispersion of chopped carbon fibres in an aqueous medium followed by the use of paper-making technology to obtain a highly porous carbon mat. It was found that the carbon fibre paper made by the above method possesses high porosity and sufficient handling strength. Yang et al. [7] separate fibres from bundles to single fibres with association of several dispersants and found out that X-100 have the best efficiency. Therefore, on the basis of former experience which partly solved the problem of dispersing fibres, preforms qualified for CVI process with self-supporting structures were obtained by laminating the as-prepared fibre papers in this study.

In the present work, a wet papermaking method was taken to fabricate random orientation carbon fibre papers. The method includes fibre dispersion and vacuum filtration, which ensures both homogeneous dispersion and integrity of fibres. Preforms with self-supporting porous structures for vapor infiltration were obtained after lamination of the random orientation carbon fibre papers. Pyrolytic carbon (PyC) interphase was deposited on the as-prepared preforms and then SiC and PyC were alternatively deposited to obtain multilayer composites with PyC inside matrix (C_{sf}/SiC-C). Accordingly, C_{sf}/SiC composites without PyC inside matrix were also obtained. The effect of fibre content on mechanical properties of C_{sf}/SiC composites was investigated. A related fibre distribution model was established to illustrate the numerical relationship among bending strength, porosity and fibre content. The difference between the mechanical properties of C_{sf}/SiC-C and C_{sf}/SiC composites was discussed.

2. Experimental

2.1. Raw materials and fabrication

Preforms were fabricated with a wet papermaking technique followed by the lamination process. The method of preparing preforms involves four steps: solution preparation; fibre dispersion; vacuum infiltration and lamination, as illustrated in Fig. 1. Commercially available carbon fibres (T300) were used as the additive reinforcement, with a diameter of 7 μm and an average length of 5 mm. It is reported that T300 fibre has good thermal stability and high temperature mechanical properties [21], which is qualified for the CVI process. Initially, as-received fibres were dispersed and stabilized in carboxymethyl cellulose (CMC, 0.2 wt. %) by aid of ultrasonication using Triton X-100 (1 wt. %) as nonionic surfactant. Triton X-100 has been reported to own an excellent capacity to distribute fibres [7]. Then the obtained highly homogenous chopped carbon fibre suspension was vacuum infiltrated through a polytetrafluoroethylene filter membrane with a 0.22 μm pore size under −0.1 MPa. Single random orientation carbon fibre paper with diameter of 100 mm and thickness of ca. 320 μm was obtained in this process. After being dried in an oven at 70 °C for 30 min, approximately 10 layers of fibre paper were laminated to produce preforms. It is reported [22] that addition of short carbon fibres seems to worsen the densification process in the Csf/SiC multilayer composites obtained through a tape casting and pressureless sintering method. In this work, we chose different carbon fibre contents (10.2 vol%, 11.8 vol%, 13.9 vol%) to study the influence of fibre content on mechanical properties.

In order to relieve the thermal stress between the carbon fibre and the SiC matrix and make the composite have the similar fracture behavior of metal materials, it is common to deposit a layer of PyC interphase with a certain thickness and proper weak bonding between the carbon fibre and the SiC matrix. Moreover, this layer can protect the carbon fibre from corrosion of harmful gas. Then PyC interphase (about 200 μm) was initially deposited on the as-prepared preforms by CVI at a temperature of approximately 900 °C. Subsequently, multilayer matrix of SiC and PyC was alternatively infiltrated into the preforms at around 1000 °C by CVI to obtain C_{sf}/SiC-C composites. Also, C_{sf}/SiC composites without PyC inside matrix were fabricated. The detailed CVI conditions and parameters can be found in the previous works [22,23].

2.2. Observation and measurement

Microstructure of composites before and after fracture was observed by scanning electron microscopy (SEM, Hitachi S-2700, Hitachi Ltd., Tokyo Japan). Archimedes principle was employed in the measurement of bulk densities and open porosities of the composites while pore size distribution was determined by mercury injection. Fracture toughness was evaluated using the single edge notched beam (SENB) method with crosshead speed of 0.05 mm min^{-1} and span of 20 mm. Bending strength was tested with crosshead speed of 0.5 mm min^{-1} and span of 30 mm. All the samples were with dimensions of about $2 \times 3 \times 36\ mm^3$. The data for each specimen was averaged over five tests to ensure the accuracy of the data. The fracture toughness was obtained by the calculation formulas in ASTM E399-74.

$$K_{IC} = \sigma_c \sqrt{a}\, Y = \frac{P_c}{B} \frac{S}{W^{\frac{3}{2}}} f\left(\frac{C}{W}\right) \tag{1}$$

$$f\left(\frac{C}{W}\right) = 2.9\left(\frac{C}{W}\right)^{\frac{1}{2}} - 4.6\left(\frac{C}{W}\right)^{\frac{3}{2}} + 21.8\left(\frac{C}{W}\right)^{\frac{5}{2}} - 37.6\left(\frac{C}{W}\right)^{\frac{7}{2}} + 38.7\left(\frac{C}{W}\right)^{\frac{9}{2}} \tag{2}$$

where P_c is critical load, S is span, C is incision depth, B is the width of the sample and W is the thickness.

3. Results and discussion

3.1. Microstructures

Compared with ball-milling method where fibre length reduction is inevitable [11], the wet papermaking method in this paper can obtain homogeneous fibre dispersion and avoid fibre length reduction simultaneously. The average fibre length after dispersion is still around 5 mm, indicating no length reduction during the carbon fibre paper fabrication process. Moreover, Fig. 2a shows that the surface of the carbon fibres after dispersion is quite smooth. The evidence above indicates that the method in this paper not only maintains the original fibre length, but keeps the integrity of carbon fibre surface. Fig. 2b indicates the image of the preform fabricated by laminating over 10 layers of carbon fibre papers. It can be observed that the preform with self-supporting structures has sufficient handling strength.

SEM images of the carbon fibre paper after about 100 h SiC deposition are shown in Fig. 3. The chopped fibres are homogeneously dispersed and joint with each other, forming a self-supporting network. It can be seen that the overall structure of carbon fibre paper is well maintained during lamination and the following CVI process. Fig. 3b