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Fabrication and characteristic of non-oxide fiber tow reinforced silicon nitride matrix composites by low temperature CVI process

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Abstract

Non-oxide fiber tow reinforced silicon nitride matrix composite was fabricated by low temperature CVI process with PyC as interphase. The tensile strength of the C and SiC fiber tow composites were 547 MPa and 740 MPa, respectively. The difference in tensile strength was analyzed based on the length, amount of pull-out fiber and also interface bonding. The infiltration uniformity of CVI silicon nitride (SiN) matrix within SiC fiber tow was comparable with that of CVI SiC matrix. These results suggested that the low temperature CVI process is suitable for the fabrication of fiber reinforced SiN matrix composites with proper interface bonding and high strength.

Keywords: B. Composites; C. Mechanical properties; D. Si_3N_4; E. Structural applications; CVI

1. Introduction

The chemical vapor infiltration (CVI) is a well-known technological route which fabricates ceramics at relatively low temperature [1]. However the fabrication processes of fiber reinforced silicon nitride (Si_3N_4) composites were still based on sintering techniques such as reactive bonding sintering, high pressure and high temperature sintering and so on [2–4]. Most of current fiber reinforcements cannot sustain the demanding sintering condition of high temperature and pressure. Fiber damage and difficulty in the controlling of interface bonding between fiber and matrix are the main problems in sintering process. Although Si_3N_4 is one of the most important engineering ceramics, there were only several trials to fabricate fiber reinforced Si_3N_4 composites by CVI process. One of the main obstacles, preventing the development of CVI process, is the relatively high deposition rate of Si_3N_4 which results in the easy closure of the outside pores and hence blocking the reactive gases to move into fiber tows. Fitzer et al. [1] reported the CVI infiltration behavior of Si_3N_4 matrix by porous reactive bonding sintered Si_3N_4 preform. The results showed that the densification of porous preform was quite difficult when the CVI temperature was higher than 1000 °C due to the higher deposition rate of Si_3N_4 at elevated temperature. Kothari et al. [5] made carbon nanofiber reinforced Si_3N_4 composites by CVI process, but the nanofiber as reinforcement was not suitable for real applications. Veltri et al. [6] deposited Si_3N_4 matrix on C preforms at the temperature of 1450 °C. However, high process temperature resulted in too strong interface bonding and toughness was not observed. Chen et al. [7] also fabricated C fiber reinforced Si_3N_4 composites at 1250 °C and the ablation mechanism was investigated. However, there was no detailed information about the microstructure of as-fabricated composites. The higher CVI temperature normally accompanies with higher deposition rate, which quickly closes the gas path way and leaves close pores inside the fiber tows. The resulting high porosity and low infiltration efficiency definitely are not good for properties of the composites. Meanwhile, the fibers are prone to be damaged at high process temperature and it is also difficult to control the interface bonding condition between the fibers and matrix. Although research started several decades ago, there was no report about fiber reinforced Si_3N_4 composites by the CVI process with good properties. Due to high deposition rate, poor densification efficiency, and strong interface combination, the CVI Si_3N_4 did not attract much

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attention. Therefore it is necessary to study continuous fiber reinforced Si_3N_4 matrix composites at low temperature with lower deposition rate, proper interface bonding, and acceptable properties.

In our previous work, [8] Si_3N_4 particle reinforced Si_3N_4 composites were fabricated by CVI process. Here in this study, the C and SiC fiber tows reinforced SiN composites were produced at 800 °C and properties were characterized. The fracture toughness and other basic properties will be discussed by 2D bulk composites in the later reports.

2. Experimental procedure

Two types of non-oxide fiber tows, carbon (C, T300, Toray Carbon Fibers, Japan) and silicon carbide (SiC, Hi-Nicalon, Nippon Carbon Co., Ltd, Japan), were used as reinforcement. The fiber tows were first deposited by PyC interphase at 900 °C and then SiN matrix were prepared. The deposition of SiN matrix was conducted at 800 °C with the reaction of silicon tetrachloride ($SiCl_4$, 99.99 wt%) and ammonia gas (NH_3, 99.99%) at the pressure of 0.01 atm. The reactive gas ratio of $SiCl_4/NH_3$ was 0.47 with excess NH_3. The hydrogen (H_2, 99.999%) was used as carrier gas and argon (Ar, 99.9%) as dilution gas with a ratio of 5.

The room temperature tensile strength was measured in a fiber tensile test machine (INSTRON-3345). Briefly, fiber tow composites were fixed in central line of two pieces of the aluminum plates by epoxy and cured at room temperature. The composites had 50 mm length between the two aluminum plates. For more details about the fixture please refer to our previous publication [13]. Tensile speed of 0.2 mm/min was used in our experiments and at least 20 samples were tested for the statistic analysis. The distribution of the tensile strength of composites was analyzed by two-parameter Weibull distribution [9–12]. The microstructures were examined using a scanning electron microscopy (SEM, S-4700, Hitachi, Tokyo, Japan) along with attached EDS software. The porosity of the as-received fiber tow and their composites were examined by the mercury intrusion porosimeters (Poremaster-33, Quantachrome Instruments, Florida, USA).

3. Results and discussion

The C and SiC fiber tows composites eventually called as C/SiN and SiC/SiN composites. The initial porosity of as-received C and SiC fiber tows was 33% and 83% which decreased to 28% and 44% for C/SiN and SiC/SiN composites, respectively. The SiC/SiN composites exhibited higher porosity as compared to C/SiN composites, however, it revealed higher infiltration efficiency (the ratio of porosity after and before matrix deposition) which indicated higher percentage of matrix in SiC/SiN composites. The difference in infiltration further affected the properties of the fabricated composites. The two-parameter Weibull distribution of the room temperature tensile strength of C/SiN and SiC/SiN was shown in Fig. 1. Most of the experimental data coincided with the fitting line. The statistical average tensile strengths of C/SiN and SiC/SiN composites were 547 MPa and 740 MPa, with Weibull parameters of 8.1594 and 8.6453, respectively, as shown in Table 1. The tensile strength of C and SiC fiber was 3.1 GPa and 2.8 GPa, respectively, as reported by the suppliers [14–15]. The tensile strength of C/SiN composite was lower than that of SiC/SiN composites which will be discussed later on the basis of microstructure analysis.

The C/SiN composite was mechanically fractured and the cross-section was observed under the microscope as shown in Fig. 2. Long and large amount of fiber pull-out was observed in the cross-sectional image, Fig. 2(a), which indicated that the deposition of SiN matrix did not damage the C fiber. If damaged, the fibers were intended to break at the damage place resulting in the composites with shorter or no fibers pull out. The long fiber pull-out also suggested that the interface bonding between the fiber and matrix was not very strong which was further demonstrated by the debonding phenomena, as shown in Fig. 2(b) and (c). The thickness of the matrix was ~2 μm at outside of the fiber tows, whereas, it was less than 1 μm at the central part of the fiber tows.

Table 1
Parameters of Weibull distribution of room temperature tensile strength of C and SiC fiber tow reinforced silicon nitride composites.

Fibers	m	σ_0	$\overline{\sigma}_{comp}$	C_V
C	8.1594	579.9533	547	0.1455
SiC	8.6453	782.8749	740	0.1371

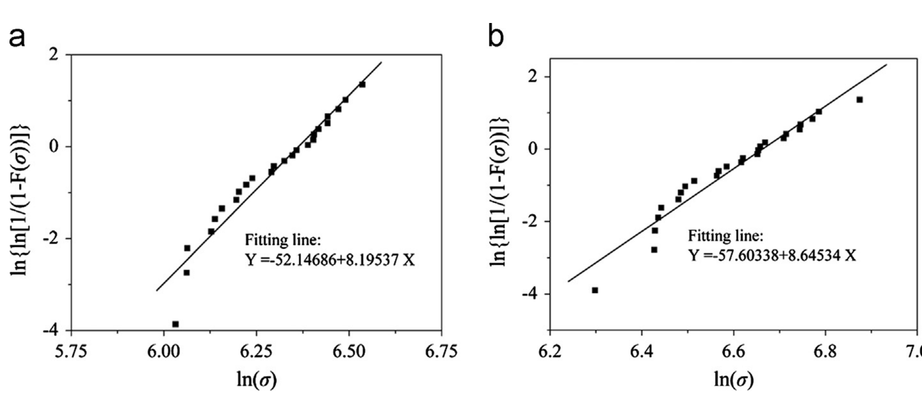

Fig. 1. Two-parameter Weibull distribution of the tensile strength of (a) C and (b) SiC fiber tow reinforced silicon nitride composites at room temperature.