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Letter to the Editor

Moisture behavior and effects on the mechanical properties and the microstructures of SiO_{2f}/Si_3N_4–BN amorphous composites

Y.G. Jiang *, C.R. Zhang, F. Cao, S.Q. Wang, H.F. Hu, G.J. Qi

Key Laboratory of Advanced Ceramic Fibers and Composites, National University of Defense Technology, Changsha 410073, PR China

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Abstract

Water diffusion in fiber reinforced ceramic composites could reduce the flexural strength as a result of weakened fiber/matrix binding. In the paper, the braided silica fiber reinforced silicon nitride and boron nitride amorphous composites (SiO_{2f}/Si_3N_4–BN) was prepared through repeated infiltration of hybrid preceramic precursor and pyrolysis at high temperature in ammonia atmosphere. The moisture behavior of the SiO_{2f}/Si_3N_4–BN composites and moist effects on the mechanical properties and the microstructures of composites were studied. The results showed that the water absorption characteristic of amorphous composites could be described by using the Fick' law. The flexural strength could be adjusted and the maximal value of that reached 161.7 MPa by controlling moderate relative humidity, which is 58.8% higher more than that of the as-received composites. SEM indicated that good mechanical properties is on the ground of the interface structure change between fiber and matrix and more fibers were pulled out which absorbing much more fracture energy.

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Keywords: Ceramics; X-ray diffraction; Fracture; Mechanical, stress relaxation; Strength; Defects

1. Introduction

The experience of recent years, especially in aeronautics, shows the importance of environmental effects on the mechanical properties of composites as well as on their long-term behavior. The most important environmental factors are temperature, humidity and radiation. Among the above phenomena, water diffusion in composites can cause plasticization and hydrolysis of the polymeric matrix and, later, a possible decohesion of the fiber/matrix interface [1–3], resulting in loss of microstructural integrity. The net effect of moisture absorption is the deterioration of matrix-dominated properties such as compressive strength, interlaminar shear strength, fatigue resistance and impact tolerance [4–6].

In the paper, the SiO_{2f}/Si_3N_4–BN composites have been prepared through repeated infiltration of hybrid preceramic precursor and pyrolysis at high temperature in ammonia atmosphere. Study showed that it is an amorphous ceramic composite and has high flexural strength. However, according to the literature [7] the silica fiber was susceptible to water and cannot be removed even in high temperature due to group –OH in the surface of the fiber, because of the hydrogen bonding between groups –OH and H_2O. When exposed to moisture environment the BN will absorb the water vapor in air and hydrolyze into boron oxide [8]. Cofer and Economy [9] found that the sensitivity to moisture was dominated by the crystalline structure, specimens which have large $d(002)$ spacing are significantly more sensitive than those that are close to the theoretical hexagonal BN structure.

Since moisture absorption may reduces badly the flexural strength through decreasing decohesion of the fiber/matrix interface and dielectric properties owing to water

* Corresponding author. Tel./fax: +86 731 4576433.
E-mail address: jygemail@yahoo.com.cn (Y.G. Jiang).

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absorption, it is urgent to understand the moisture behavior of the SiO_{2f}/Si_3N_4–BN composites in order to predict long-term material and structural performance.

Up to now, there is little available literature regarding on the moisture absorption behavior of the fiber reinforced ceramic composites [10]. This paper focuses on the moisture diffusion behavior of SiO_{2f}/Si_3N_4–BN amorphous composites, and the relationship between water absorption characteristics and changes in mechanical properties and microstructures over time.

2. Experimental procedures

2.1. Raw materials

The silica fibers, produced by JingZhou Feilihua Quartz Glass Corporation (China), had a density of 2.2 g/cm^3, a tensile strength of 1700 MPa and an elastic modulus of 78 GPa braided silica fiber fabric (SiO_{2f}), with the fiber volume fraction 46%, was woven by Beijing Fiberglass Research & Design Institute (China).

The starting preceramic precursor is synthesized by mixing perhydropolysilazane (PHPS) and poly[2,4,6-tri(methylamino)borazine] (PolyMAB) [11], the ratio between them is no more than 1/10. The precursor for silicon nitride (Si_3N_4) ceramic matrix, PHPS was synthesized by the ammonolysis of dichlorosilane–pyridine adduct [12]. PolyMAB, the precursor for boron nitride (BN) ceramic matrix, prepared by thermal condensation of 2,4,6-tri(methylamino)borazine (MAB) according to the literature [13]. The precursor was a transparent liquid with a low viscosity of 25–40 MPa s (25 °C), a density of 0.86 g/cm^3, and a high ceramic yield ($\geqslant$80%).

2.2. Composites preparation

The preparation of SiO_{2f}/Si_3N_4–BN composites included three stages. Firstly, SiO_2 fabrics were infiltrated with hybrid precursor in vacuum. Then, the performs filled with precursor cured at 100–200 °C for 3–6 h in an inert atmosphere. Finally, the cured performs were pyrolyzed in ammonia at 800 °C. The infiltration-cure-pyrolysis cycles were repeated for three times in order to density the composites. In the paper the composite density reached 1.67 g/cm^3 after three infiltration-pyrolysis cycles.

2.3. Composites characterizations

SiO_{2f}/Si_3N_4–BN composites were placed in four different atmosphere with different relative humidity (RH) from 5%, 18%, 55% and 95%, donated as A1–D1, and A2–D2, respectively, controlled by environment instrument (HS-100, China) at room temperature, the experiment period is 26 days and 54 days (Tables 1 and 2). The weight gain was measured by analytical balance with measurement precision 0.1 mg (TG328-A) at regular intervals. The moisture content (M) at any time t was calculated according to the following equation:

$$M = \frac{m - m_0}{m_0} \times 100\% \quad (1)$$

Where M represents moisture content, m_0 and m are weights of dry and wet specimens, respectively.

XRD patterns were recorded at room temperature by using a Siemens equipment and Cu Kα1 radiation ($\lambda = 0.154$ nm), XRD was used to assess the phase compose of the composites. Three-point bending tests machine (WDW-100) were used to evaluate flexural strength of the dry and wet specimens with the span of 30 mm and a cross-head speed of 0.5 mm/min carried out at a test piece 4 mm wide and 3 mm thick. For mechanical properties tests, at least three specimens were measured for each composite. After the flexure tests, Fracture surfaces of the specimens were examined on the scanning electron microscopy (JSM-5600LV).

Table 1
Flexural strength of the composites for different relative humidity after 26 days

Specimen	Relatively humidity RH (%)	Moisture content M (%)	Flexural strength σ_f (MPa)
A1	5	0.601	114.1
B1	18	1.798	117.5
C1	55	10.87	143.3
D1	95	24.15	154.2

Table 2
Flexural strength of the composites for different relative humidity after 54 days

Specimen	Relatively humidity RH (%)	Moisture content M (%)	Flexural strength σ_f (MPa)
A2	5	1.332	121.8
B2	18	2.503	161.7
C2	55	13.13	143.7
D2	95	30.31	130.6

3. Results and discussion

3.1. X-ray diffraction spectra of the composites

As is revealed in Fig. 1, XRD pattern of the SiO_{2f}/Si_3N_4–BN composites, three phase SiO_2, Si_3N_4, and BN, were detected for the as-received composites pyrolyzed at 800 °C. No crystalline BN was observed for the composite except the weak but sharp peak at 42.5° and 77.8° due to the low pyrolysis temperature. No crystal silica fiber was found, which is important in maintaining low coefficient of thermal expansion and good mechanical properties as a reinforcement for the silica fibers and the same as to Si_3N_4. Silica fibers and Si_3N_4 still remain amorphous at pyrolysis temperature corresponding to the broad peaks in the figure, which is consistent with the literature [14–16]. Therefore, the present composites should be amorphous.