Preparation and Properties of Boron Nitride Fiber Reinforced SiBON Composites

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Abstract. In this paper, BNf/SiBON composites were prepared by adding short boron nitride fibers and in-situ inorganic solid state reaction. The relationships among hot-pressing sintering temperature, boron nitride fiber content, density and flexural strength of the composites were studied. The phase composition and microstructure of the composites were also analyzed and characterized. The results show that the bending strength of BN/SiBON increases with the increase of sintering temperature when the fiber content is constant, and decreases with the increase of fiber content when the sintering temperature is constant. XRD diffraction patterns also showed that BN had higher crystallinity and larger grain size when the sintering temperature was 1700°C and the content of BN fiber was 40%. Therefore, the best content of boron nitride fibers is in the range of 20% to 40%, and the hot pressing sintering temperature is about 1700°C.

1. Introduction

Thermal wave-transmitting materials is a kind of special dielectric materials with good dielectric properties at high temperature and even ablation. They have wide and important applications in the electromagnetic transmission windows of various high speed spacecraft. Thermal wave-transmitting materials are typical multifunctional integrated materials and have excellent and stable dielectric properties in a wide temperature range, as well as good mechanical properties, heat resistance, ablation resistance, insulation and thermal shock resistance. The performance and application not only directly affect the communication and guidance system, but also affect the aerodynamic characteristics and structural reliability of aircraft[1].

At present, SiO2f/SiO2 composites can meet the requirements and have been already applied. However, the strength of SiO2 fibers decreases rapidly when the temperature is over 800°C and too low to be measured when the temperature exceeds 1200°C, which limits its wider application[2,3]. In contrast, BN fibers and BN matrix composites produces heat-resistant, excellent mechanical properties at high temperature and dielectric properties. The oxidation resistance temperature of BN fibers is higher than that of carbon fibers and boron fibers and can be used in oxidation atmosphere at 900°C for a long time, and the strength is not decrease in inert atmosphere below 2000°C. SiBON ceramics combine the advantages of SiO2 and BN, such as excellent dielectric, mechanical, anti-diffusion and bonding properties with other materials and has significant value [4-8].
In this paper, BNf/SiBON composites were synthesized in-situ inorganic solid-state reaction by adding short-cut boron nitride fibers and the properties were studied.

2. Experimental

2.1. Preparation
The raw materials are short BN fibers, H$_3$BO$_3$ powder (analytical purity), Si$_3$N$_4$ powder and n-hexane (analytical purity). B$_2$O$_3$ is dehydrated from H$_3$BO$_3$. B$_2$O$_3$ powder, Si$_3$N$_4$ powder, short BN fibers and grinding balls were grinded with n-hexane as solvent for 16-20 hours at a mass ratio of 2:1:0.6-1.2:1.5, and then dried. The dried powder was packed into the graphite mould coated with boron nitride on the inner wall, sintered at 1600°C and 1700°C for 2hours under argon protection and then hot pressed with 10MPa for 15minutes.

2.2. Characterization
The density was determined by Archimedes drainage method. The bending strength of specimens was measured by three-point bending method on universal testing machine. The size of specimens was 3mm×4mm×36mm, the span was 30mm, and the moving speed was 0.5mm/min. The surface morphology was observed by scanning electron microscopy. The phase composition of the powder product was analyzed by D/max 2200 X-ray diffractometer.

3. Result and Discussion

3.1. Effect on Density
The density of BNf/SiBON composites is affected by both hot pressing sintering temperature and fiber content. In this paper, the sintering temperature is 1600°C and 1700°C, and the content of boron nitride fiber is 20%, 40% and 60%.

Figure 1 shows the density of samples prepared at 1600°C and 1700°C when the content of boron nitride fiber is 20%, 40% and 60%, respectively. It can be seen from Figure 1 that the density increases with the increase of sintering temperature when the fiber content is 20%. When the fiber content is 40%, the density decreases with the increase of sintering temperature, but the density value are still higher than 2.0g/cm$^3$. Compared with the sample of 20% fiber content, the density of 40% fiber content is more stable. The density of BNf/SiBON composites decreases gradually from 2.09 g/cm$^3$ to
1.91 g/cm³ with the increase of BN fiber content at 1700°C. It is concluded that when the content of BN fibers is in the range of 20% to 40%, the effect is most ideal.

### 3.2. Effect on bending strength
The bending strength of composites prepared by hot pressing sintering at 1600°C and 1700°C with 20%, 40% and 60% BN fibers are shown in Figure 2.

![Figure 2. Bending strength at different temperatures and fiber contents.](image)

With the increase of sintering temperature, the bending strength of samples increases gradually, and the bending strength at 1700°C is better than that at 1600°C. When the sintering temperature is constant, the bending strength decreases with the increase of boron nitride fiber content, so the content of fibers should not be too much, and had better to be controlled in the scope of 20 to 40 percents.

### 3.3. Analysis of Phase Composition
The phase analysis of BNf/SiBON composites with BN fibers contents of 20% and 40% prepared by hot pressing sintering at 1600°C and 1700°C is carried out. The XRD diffraction patterns are shown in Figure 3 and Figure 4.

![Figure 3. XRD patterns of 20% BN fibers.](image)
From Figure 3 and Figure 4, it can be found that the BN/SiBON composites mainly consist of BN and Si₃N₄. When the fiber content is constant, the intensity of BN diffraction peaks sintered at 1700°C is stronger than that sintered at 1600°C. When the sintering temperature is 1700°C, the crystallization of BN is more sufficient, and the grain size is larger. There is no obvious difference in the Si₃N₄ intensity of diffraction peaks at 1600°C and 1700°C. Therefore, the properties of the composites prepared by hot pressing sintering are better when the sintering temperature is 1700°C.

3.4. Microstructure analysis

The SEM photographs of 20% BN fibers at 1600°C are shown in Figure 5. It can be seen that SiBON ceramic matrix accounts for the majority of the samples. When brittle fracture occurs, the boron nitride fibers adhere to the ceramic matrix and are pulled out. SEM photos of 20% BN fibers at 1700°C are shown in Figure 6. Comparing with Figure 5, it can be concluded that when brittle fracture occurs, less BN fibers were pulled out and the density is higher.

The SEM photographs of 40% BN fibers at 1600°C are shown in Figure 7. Compared with Figure 5(a), the content of boron nitride fibers in the samples increased and the fibers are not uniformly dispersed, but the bonding between matrix and fibers is a close connection. When brittle fracture occurs, the fibers are pulled out from the fracture surface, as shown in Figure 7(b).
The SEM photographs of 20% BN fibers at 1700°C are shown in Figure 6. When brittle fracture occurs, no fibers are pulled out from the surface as shown in Figure 8 (b). The bonding between fibers and matrix is closer, and better than that of 40% BN sintered at 1600°C.

4. Conclusion

(1) When the content of boron nitride fiber is 20% and 40%, the density is 2.09g/cm³ and 2.04g/cm³ respectively, which were more stable than 1600°C when the sintering temperature was 1700°C.
(2) The bending strength of BN/SiBON increases with the increase of sintering temperature when the fiber content is constant, and decreases with the increase of fiber content when the sintering temperature is constant. It goes for a maximum (134MPa) at 20% BN fiber and 1700°C.

(3) The BN crystallization is more sufficient and the grain size is larger with the hot pressing sintering temperature increase. There is no obvious difference in the intensity of diffraction peaks of Si₃N₄ phase at 1600°C and 1700°C.

(4) When the sintering temperature is 1700°C, the matrix is more closely bound to the BN fibers.

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