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Si_3N_4-BN-SiO_2 Based Microwave-Transparent Materials

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Abstract. The Si-B-O-N microwave-transparent materials were prepared by gas pressure sintering (GPS).The effect of BN and nano-SiO_2 contents on the mechanical and dielectric properties of the composites was studied. The microstructural characteristic and reinforced mechanism of the composites were also investigated. The results showed that a series of Si-B-O-N wave-transparent materials could be obtained by controlling the contents of raw materials and technological parameter. The bending strength of composites is from 74.7MPa to 174.83MPa, the dielectric constant is from 3.5 to 4.2 and the tangent of loss angle is from 0.5×10^{-3} to 4.5×10^{-3}.

Introduction

Radome as a vector of radar is a pivotal assembly for missile and radar system. With the development of aerospace technology, radome with high temperature resistance, erosion resistance, wide frequency band and high microwave-transparent rate is the study orientation. In recent years, high performance nitrides microwave-transparent ceramics gradually hold absolutely predominance in the field of missile radome of supersonic for the good dielectric performance, high strength and excellent high temperature properties.

Si_3N_4-BN-SiO_2 prepared by GPS was chosen as a basic system. Nano-SiO_2 and BN were added as second phase. BN powder can decrease dielectric constant and dielectric loss, improve dielectric stabilization and performance of thermal shock resistance of Si_3N_4 materials. The performance of the series of Si-B-O-N wave-transparent composites can be further improved by adding nano-SiO_2 as reinforced phase.

Experimental Procedure

Raw materials. The starting powder is commercially available α-Si_3N_4 with an average particle size of 2μm and a purity of 99.30wt%. The *h*-BN powder is commercial with an average particle size of 0.5μm and a purity of 99.14wt%. The amorphous nano-SiO_2 powder was supplied by Kiln Nanometer Technology Development Co., Ltd, with an average particle size of from 30 to 50nm and a purity of 99.90wt%. The yttrium oxide and alumina powders are analytical reagent and the average particle size is 1.1μm and 0.8μm respectively.

Specimen fabrication and tests. The nano-SiO_2 powder, varying from 0, 5, 10, 15, 20, 25 to 30wt%, and the BN powder, varying from 10, 20 to 30wt%, were ball-milled respectively with Si_3N_4 and sintering aids, then dried, granulated and molded by cold isostatic pressing. The green bodies were sintered by GPS at 1870°C for 2hrs at a pressure of 8MPa nitrogen gas.

Density, water absorption and apparent porosity of the sintered composites were measured by the Archimedes method. The flexural strength was measured by three-point method using MTS-810 at room temperature. The microstructure was analyzed by SEM. The crystalline phased were identified by XRD analyses. The dielectric performance of materials was tested by short circuit waveguide method in 2cm frequency wave band at 1MHz at room temperature.

Results and Discussion

Effect of nano-SiO_2 content on the physical and mechanical properties. Fig.1 shows the effect of

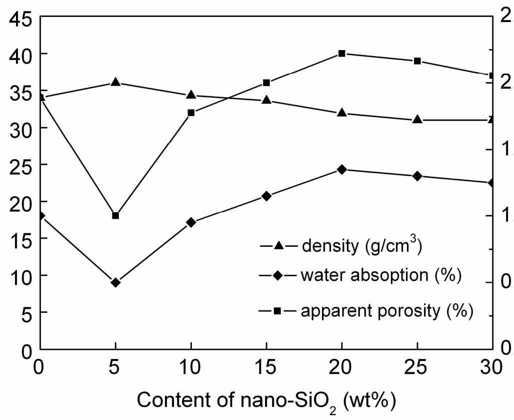


Fig. 1 The relationship between density, water absorption porosity, and content of nano-SiO_2

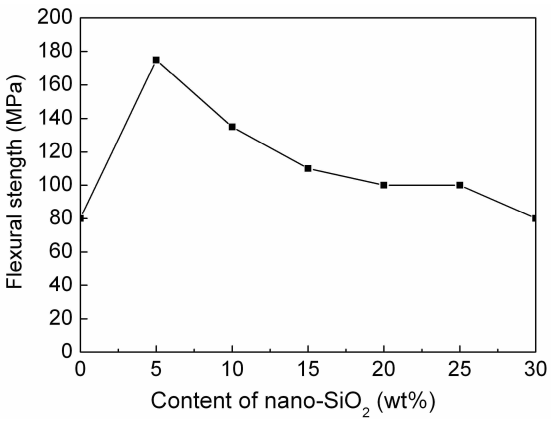


Fig.2 The relationship between flexural strength and content of nano-SiO_2

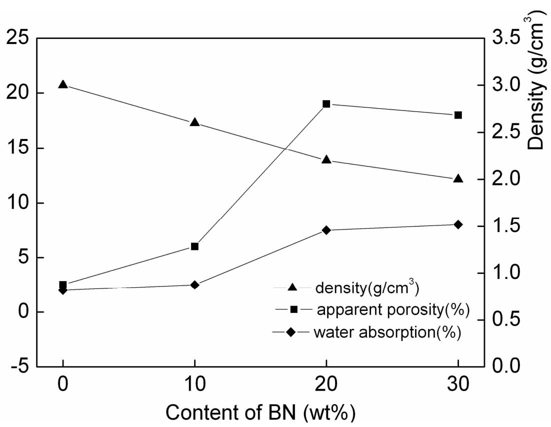


Fig.3 Relationship between density, porosity, water absorption and BN content

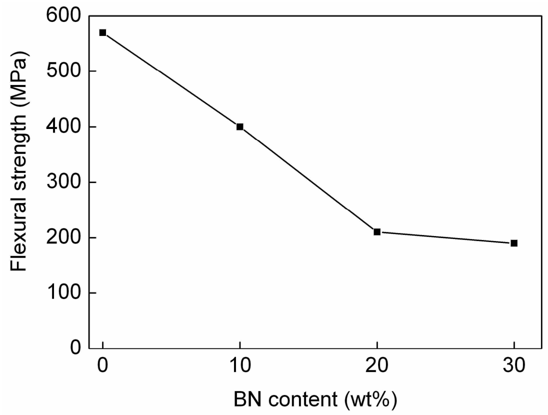


Fig.4 Relationship between bending strength and BN content

SiO_2 content on the density, water absorption and porosity of the Si-B-O-N composites. The samples have the highest density, the least water absorption and porosity with 5wt% SiO_2. The sintered density is decreasing with the content of SiO_2 increasing, while water absorption and porosity are increasing. Addition of 5wt% nano-SiO_2 in the composites effectively improves the relative density (69.5%) compared to those without nano-SiO_2 addition (60.8%). From above analysis it can be concluded that nano-SiO_2 can sharply promote sintered densification with 5wt% nano-SiO_2.

Fig.2 shows the effect of SiO_2 content on the bending strength. It can be seen that the bending strength reaches 174.83MPa with 5wt% nano-SiO_2, which is 2.34 times that of without SiO_2. So nano-SiO_2 powder has well reinforcing effect on the Si-B-O-N composites.

Effect of BN content on the physical and mechanical properties. Fig.3 shows the effect of BN content on the density, water absorption and porosity. With the BN content increasing, the density decreases, while water absorption, porosity increases respectively. And as shown in Fig.4, with the increasing of BN content, the bending strength decreases gradually.

Effect of components on dielectric properties of composites. The dielectric properties of composites will change with the difference proportion of various components. The theoretical dielectric constant of composites can be induced according to the Lichtencker logarithmic mixture principle.

$$\ln \varepsilon = \sum_i V_i \ln \varepsilon_i \tag{1}$$

$$\ln tg\delta = \sum_i V_i \ln tg\delta_i \tag{2}$$

Here ε, tgδ are the dielectric constants and tangent of loss angle of composites. ε_i, tgδ and V_i are the dielectric constant, tangent of loss angle and volume fraction of the *i* component respectively.

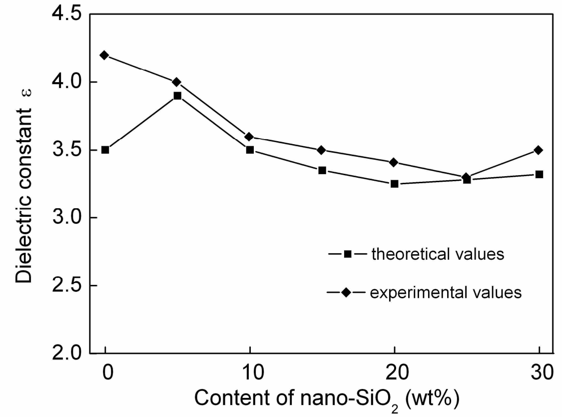


Fig.5 The relationship between dielectric constant and nano-SiO_2 content

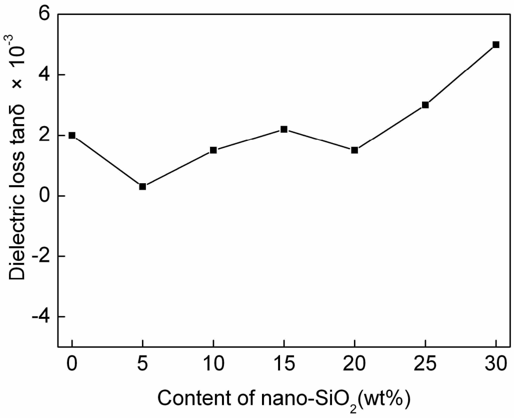


Fig.6 The relationship between dielectric loss and nano-SiO_2 content

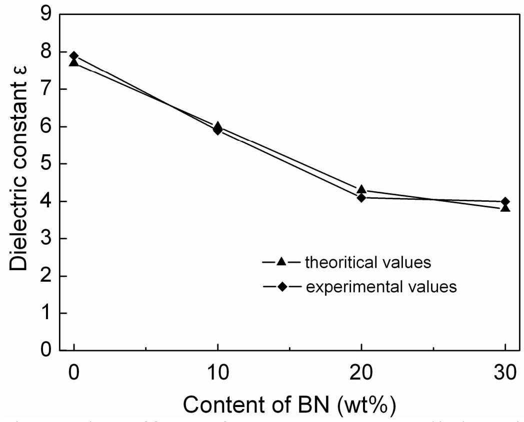


Fig.7 The effect of BN content on dielectric constant

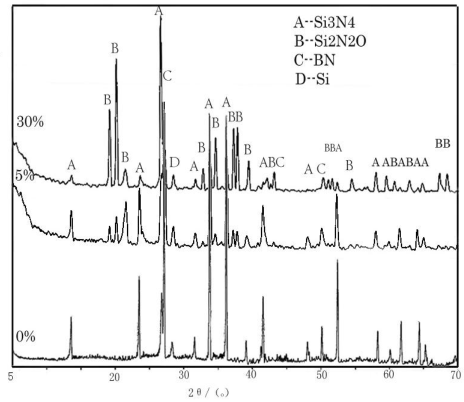


Fig.8 XRD diagram of Si-B-O-N composites

The Si-B-O-N composites with different dielectric performance can be made by varying the constant of components. At the same time, the theoretical dielectric constants of Si-B-O-N composites were calculated by Lichtenecker logarithmic mixture principle and compared with the experiment value. The effect of the content of components on the dielectric performance of Si-B-O-N composite is shown in Fig.5, Fig.6 and Fig.7 respectively.

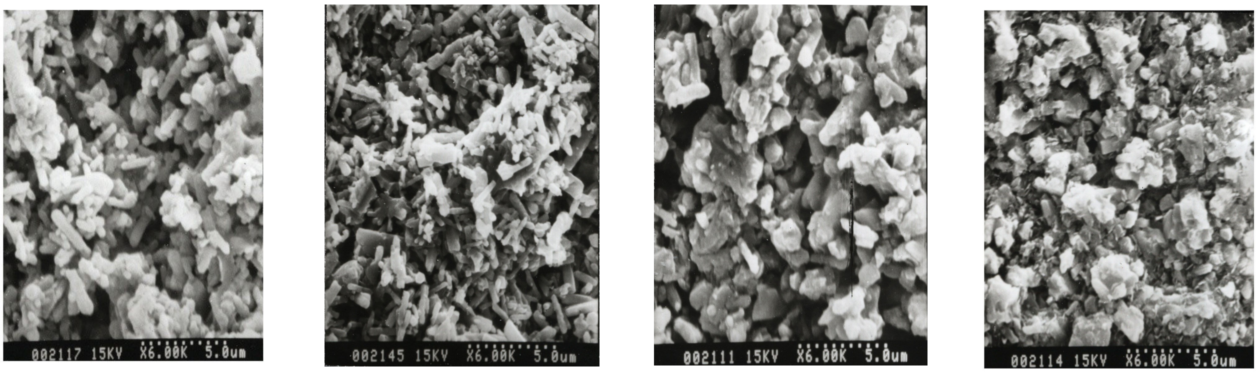
Fig.5 shows the comparison of dielectric constants of experimental and theoretical value with different SiO_2 content. With the increasing of the SiO_2 content, the dielectric constants decrease and the difference between experimental and theoretical value becomes smaller. It is proved that the dielectric constants of this series of Si-B-O-N composites are in accord with logarithmic mixture principle well and can direct to designing dielectric constants of material series.

Fig.6 shows the relationship of tangent of loss angle and SiO_2 content. When the content of SiO_2 is 5wt%, the tangent of loss angle of composites is the least. With the content of SiO_2 increasing, the tangent of loss angle increases gradually.

Fig.7 shows the influence of BN content on dielectric constants of composites. With the content of BN increasing, the dielectric constants of Si-B-O-N composites decrease gradually and experimental value approximately conforms to the theoretical value.

Phase analysis of composites. Fig.8 is the XRD diagram of Si-B-O-N composites, which shows, if no SiO_2 was added, there are Si_3N_4, BN and little free silicon in composites. Adding SiO_2 from 5wt% to 30wt%, there are Si_3N_4, BN, Si_2N_2O and little free Si, and no SiO_2 in the composites. With the content of SiO_2 increasing, the content of Si_2N_2O will be increasing. That is to say, nano-SiO_2 reacts with Si_3N_4 completely and transforms into a new phase, Si_2N_2O, as equation 3. Si_2N_2O has excellent thermal shock resistance, anti-oxidized and good high temperature strength.

$$Si_3N_4 + SiO_2 = 2Si_2N_2O \tag{3}$$



(a) no SiO_2(n) (b) adding 5wt% SiO_2(n) (c) adding 10wt% SiO_2(n) (d) adding 25wt%SiO_2(n)

Fig.9 SEM micrographs of the Si-B-O-N ceramic composites

Analysis of microstructure. Significant columnβ-Si_3N_4 grains were observed in the Fig.9 (a), and the ratio of length to diameter is about from four to five. Furthermore, there is higher porosity in the composites without nano-SiO_2 and much looser among the grains. With the content of nano-SiO_2 being up to 5wt% (Fig.9(b)), the β-Si_3N_4 becomes acicular crystals, the ratio of length to diameter being about seven or eight, and the β-Si_3N_4 grains grow interlacedly each other. The porosity is lower than other composites in Fig.9. These make the composites with 5wt% nano-SiO_2 have excellent mechanical properties. But if the content of SiO_2 is much more, the thickset crystals become more and more in Fig.9 (c) and Fig.9 (d), and the synthesized Si_2N_2O in the composites is too much comparing to the normal level. These make the mechanical properties of composites decrease.

Conclusions

Series of Si-B-O-N wave-transparent materials were prepared by GPS. And attained high mechanical properties, low dielectric constant and low tangent of loss dielectric angle composites successfully through adjusting the content of nano-SiO_2 and BN. Introducing nano-SiO_2 can reinforce the Si-B-O-N composites by reacting with Si_3N_4 to form Si_2N_2O and control the proportion of Si_2N_2O at the same time. Adding BN powder as the second phase can effectively decrease the dielectric constant of Si_3N_4, but BN can block obviously the sintering process of Si_3N_4. Because of the superior properties, practical uses of the series of Si-B-O-N composites may be expected in a wide range of transmitting fields.

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