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SciVerse ScienceDirect
Journal of the European Ceramic Society 32 (2012) 4025–4029
www.elsevier.com/locate/jeurceramsoc

Short communication

Fabrication of Si_3N_4–SiBC composite ceramic and its excellent electromagnetic properties

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Received 12 May 2012; received in revised form 27 June 2012; accepted 1 July 2012
Available online 24 July 2012

Abstract

To obtain better electromagnetic wave absorbing property, it is vitally necessary to develop novel ceramics with not only high dielectric loss but also low dielectric constant. Si_3N_4–SiBC, a composite ceramic with such dielectric properties, was fabricated by infiltrating SiBC into porous Si_3N_4 ceramic via low pressure chemical vapor infiltration. The high dielectric loss and the low dielectric constant are attributed to the unique microstructure of SiBC, which also leads to a very excellent wave-absorbing property of Si_3N_4–SiBC ceramic, attaining a minimal reflection coefficient of −28 dB. Besides, the Si_3N_4–SiBC ceramic also shows a high mechanical property. Therefore, the Si_3N_4–SiBC ceramic exhibits great potential as an excellent functional and structural ceramic.

Keywords: Si_3N_4; SiC; Carbides; Chemical vapour deposition; Functional applications

1. Introduction

SiC ceramic is an attractive thermostructural material because of its superior properties such as excellent high-temperature mechanical properties and good oxidation resistances.[1] On the other hand, SiC is also a wide bandgap semiconductor material with relatively stable dielectric characteristics at high temperature.[2] Therefore, many works have focused on SiC-based ceramic for electromagnetic wave (EMW) absorption.[2–7] Some SiC matrix composites have exhibited good EMW absorbing property when a porous EMW-transparent Si_3N_4 ceramic or ZrO_2 fiber is coated with SiC matrix by chemical vapor infiltration (CVI).[4–6] Although these composites show high dielectric loss of greater than or equal to 0.8, the dielectric constants are not low enough, still higher than 10 at 8.2 GHz. Higher dielectric constant leads to the impendence mismatch and enhances the surface reflection of the EMW,[8,9] which hinders the considerable improvement of EMW absorbing property. To obtain better EMW absorbing property, it is vitally necessary to develop novel ceramics with not only high dielectric loss (≥0.8) but also low dielectric constant (<10).

In the present work, Si_3N_4–SiBC composite ceramic was fabricated by infiltrating SiBC into porous Si_3N_4 ceramic via low pressure CVI (LPCVI). Both the permittivity and reflection coefficient of the Si_3N_4–SiBC ceramic were investigated. The aim of this work is to develop a novel Si_3N_4-based ceramic with high dielectric loss and low dielectric constant in order to improve the EMW absorbing properties of Si_3N_4-based structural ceramic, compared with the previous work.[4,5,10]

2. Experimental

The fabrication process of porous Si_3N_4 ceramic was described elsewhere.[5] SiBC was infiltrated into Si_3N_4 by LPCVI from methyltrichlorosilane (CH_3SiCl_3, 98.0% purity) and boron trichloride (BCl_3, 99.99% purity). Hydrogen (H_2, 99.999% purity) and argon (Ar, 99.999% purity) were used as the carrier and the dilution gas, respectively. The processing conditions were as follows: $\alpha = [H_2]_0/[MTS]_0 = 10$, $\beta = [MTS]_0/[BCl_3]_0 = 15$, $T = 1100\,°C$, $P = 1$ kPa. After LPCVI

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http://dx.doi.org/10.1016/j.jeurceramsoc.2012.07.002

Table 1
Mechanical property, density and porosity of the Si_3N_4 ceramic and Si_3N_4–SiBC composite ceramic.

Materials	Density/g cm^{-3}	Porosity/%	SiBC/vol.%	Flexural strength/MPa
Si_3N_4	1.78	46	0	189
Si_3N_4–SiBC	1.96	40	6	260

for 15 h, the content of SiBC in the composite ceramic was 6 vol.%.

The composite ceramic was machined into specimens with two kinds of dimensions: 3 mm × 4 mm × 40 mm for flexural strength measurement[5] and 2.8 mm × 10.16 mm × 22.86 mm for dielectric property measurement. The dielectric property testing was performed by a vector network analyzer (VNA; MS4644A, Anritsu) using the waveguide method. The relatively complex permittivity ε_r (ε', ε'') and permeability μ_r (μ', μ'') on the frequency range of 8.2–12.4 GHz (X-band) were obtained and reflection coefficient (RC) used to characterize the EMW absorbing property of material was determined using ε_r and μ_r according to the following equations:

$$\mathrm{RC} = 20\ \log_{10}|\Gamma| \tag{1}$$

$$\Gamma = \frac{\sqrt{\mu_r/\varepsilon_r}\tanh(\gamma t) - 1}{\sqrt{\mu_r/\varepsilon_r}\tanh(\gamma t) + 1} \tag{2}$$

$$\gamma = \frac{j\omega\sqrt{\mu_r\varepsilon_r}}{c} \tag{3}$$

where γ is the EMW propagation factor in the material, t is the material's thickness, ω is the angular frequency, and c is the light speed. The smaller the value of RC is, the better the EMW absorbing property of material. If RC is smaller than −10 dB, it means more than 90% of EMW is absorbed.

The morphology of the Si_3N_4–SiBC ceramic was examined by scanning electron microscopy (SEM, S4700, Hitachi). Phase composition analysis was conducted by X-ray diffraction (XRD, X'Pert Pro, Philip). Transmission electron microscopy (TEM) analysis was performed using a JEM 2100 operated at 200 kV. The ultra-high resolution micro-chemical analysis was carried out by the attached energy dispersive spectrometer (EDS, Oxford).

3. Results and discussion

Table 1 shows that the density of porous Si_3N_4 ceramic increases and the porosity decreases after infiltrating SiBC. The flexural strength of the Si_3N_4–SiBC ceramic increases by 38% compared to that of the Si_3N_4 ceramic. Fig. 1 shows the Si_3N_4 ceramic consists of rod-like β-Si_3N_4 and many three-dimensional network pores among the grains. After infiltrating SiBC, the ceramic becomes uniform and dense in microstructure, which results in the increase in the flexural strength. The previous studies[4,5] revealed that the flexural strength of Si_3N_4–SiC(BN) with 19.8 vol.% SiC(BN) and Si_3N_4–SiC with 11 vol.% SiC was 14% and 37% above that of Si_3N_4 ceramic, respectively. In this study, 6 vol.% SiBC presents more remarkable strengthening effect, which may be relative to the unique microstructure of CVI SiBC.

TEM investigations (Fig. 2a and b) show that there are two crystalline phases existing and dispersing uniformly in SiBC and they form a dense network structure. EDS analysis for the micro-area of center (P1) and wall (P2) in the network structure shows the phases in P1 and P2 are B_4C and SiC, respectively. XRD pattern of the Si_3N_4–SiBC composite ceramic can further prove this identification (Fig. 2c). It is worth noting that both phases are nano-sized and B_4C phase is wrapped by SiC nanograins, which is dispersed between B_4C phases, accompanied by abundant interfaces. Such structures, especially nano-sized SiC particles and SiC/B_4C interfaces, can produce positive impacts on mechanical and electromagnetic properties of the composite ceramic.[2]

The most important factors to the electromagnetic interaction of a dielectric material are the complex permittivity ($\varepsilon_r = \varepsilon' - j\varepsilon''$) and the dissipation factor ($tg\delta = \varepsilon''/\varepsilon'$).[11] According to the theory of complex permittivity, the increase in ε' can be mainly ascribed to the bound charges, i.e., dielectric relaxation and space charge polarization effect, and the increase of

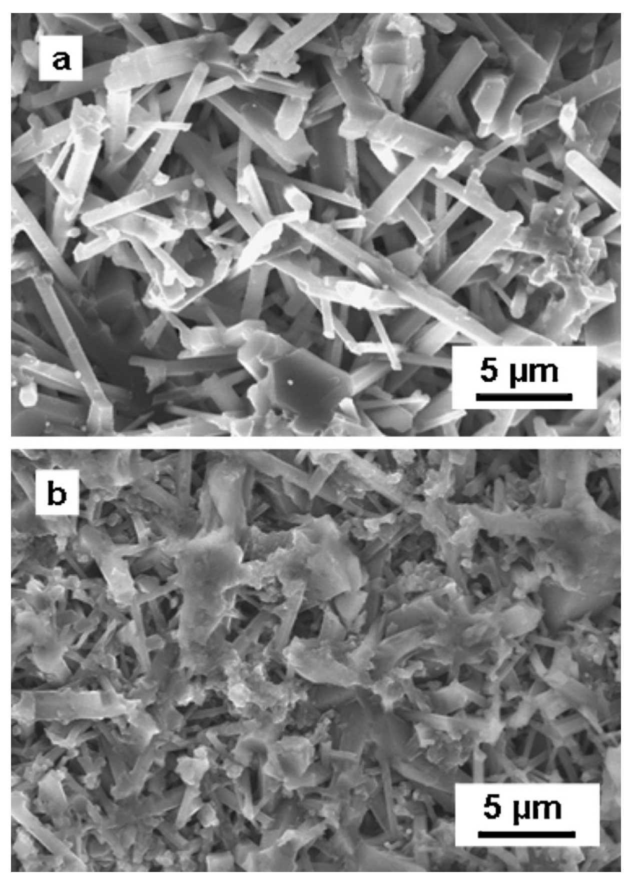

Fig. 1. SEM images of surface morphology of the Si_3N_4 ceramic (a) and Si_3N_4–SiBC composite ceramic (b).