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Effects of SiC contents on the dielectric properties of SiO_{2f}/SiO_2 composites fabricated through a sol-gel process

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ABSTRACT

SiC powders have been synthesized through a combustion process using Si powders and carbon black as starting materials. As a C-enriched α-SiC solid solution, the prepared SiC powders contain antisite defects of C_{Si} and graphite state of sp^2C, revealing good real part ε′ and imaginary part ε″ of permittivity. Subsequently, SiO_{2f}/SiO_2 composites are fabricated via a sol-gel process employing three-dimensional braided quartz fiber fabrics as a skeleton and a mixture of high-purity silica sol and the synthesized SiC powders as slurry. The effects of the SiC contents on the dielectric properties of the composites are investigated. The real and imaginary parts of the complex permittivity (ε′ and ε″) of the composites increase first before they decrease with an increasing SiC content from 0 to 40 wt.%. The maximum ε′ and ε″ are obtained at the SiC content of 20 wt.% in the range of 8–18 GHz, and the bandwidth with the reflectivity below −10 dB of the SiO_{2f}/SiO_2 composite with 20 wt.% SiC powders is 5 GHz, which can well meet the requirements as good microwave absorbing materials.

1. Introduction

Microwave absorbing materials, a kind of functional materials widely used in civil and military domains, have seen higher and higher demands nowadays due to the progressive uses of microwaves. Ideally, microwave absorbing materials should feature low density, favorable mechanical property, and strong microwave absorbing properties over a wide frequency range, and they can serve at high temperature environment, especially for uses in the stealth technology of aircrafts such as battle plan [1–4]. However, it is extremely difficult to meet all requirements using a single material. The further optimization of the desirable properties in the microwave absorbing materials is still in great need [1]. Meanwhile, silica fiber reinforced silica (SiO_{2f}/SiO_2) composites, which exhibit low density, good ablation resistance, excellent dielectric properties, high temperature stability, fine thermal shock damage resistance, and low cost, are reportedly suitable for applications at high temperature and electromagnetism environments [5–8].

Hence, applying SiO_{2f}/SiO_2 composites to the microwave absorbing materials may be a daring and interesting attempt, which can widen the family of high-temperature microwave absorbing materials.

However, a challenge still presents that SiO_{2f}/SiO_2 composites have rather low dielectric constant and loss tangent [5,7], none of which meets the requirement for microwave absorbing applications. As it is well-known, to make desirable microwave absorbing materials, the reflection coefficient should be as low as possible so that the electromagnetic waves could enter the materials with the maximum intensity, which generally requires certain permittivity parameters, especially a low imaginary part of permittivity. On the other hand, the electromagnetic wave entering the materials should be absorbed with maximum intensity, which also requires appropriate permittivity parameters, especially a relatively high imaginary part of permittivity that enhances absorption in terms of dielectric loss. The degree of dielectric loss can be evaluated using the dielectric loss tangent, i.e., $\tan\delta = \frac{\varepsilon''}{\varepsilon'}$, where δ is the dielectric loss angle [9,10]. It is thus important to adopt a feasible approach to overcome the above mentioned challenge and to meet the microwave absorbing properties as needed.

Silicon carbide (SiC), as a wide band gap semiconductor, features many attractive properties such as high breakdown field strength, high saturated carrier drift velocity, high temperature stability, high thermal conductivity, and good microwave absorbing property [11–13]. With these excellent characteristics, it is usually considered a promising candidate for microwave absorbing materials at both cryogenic and elevated temperatures [14,15]. M.S. Cao et al. fabricated a kind of Ni-decorated SiC by an improved solution chemical method and reported high-temperature dielectric properties of the core-shell SiC-Ni powders in a temperature range from 373–673 K at frequencies

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of 8.2–12.4 GHz (X-band). It was found that the Ni-decorated SiC showed great microwave absorption coupled with wide broadband characteristic, specifically excellent performance at higher temperatures [16].

In the present work, SiO_{2f}/SiO_2 composites are fabricated through a conventional sol-gel process. In order to improve the electromagnetic properties of the obtained SiO_{2f}/SiO_2 composites, a special kind of C-enriched α-SiC powders is synthesized through a combustion process and then incorporated into the composites as fillers. To our delight, it is found that the SiO_{2f}/SiO_2 composites with certain amount of the C-enriched SiC fillers actualize well with microwave absorbing properties. Subsequently, the correlation between the SiC contents and dielectric properties of the SiO_{2f}/SiO_2 composites is investigated.

2. Experimental procedures

2.1. Preparation of SiC powders

Commercial available silicon powder (99.995% pure, mean particle size 5 μm, the Second Gear Plant of Zhengzhou, China) and carbon black (99.9% pure, particle size 20–40 nm, Jiaozuo Chemical Co. Ltd., China) were used as sources of silicon and carbon, respectively. Mixtures of the Si and C powders with a molar ratio of 1:1 were ball-milled for 15 h in a SiC jar using SiC balls as the milling media (the ball/powder weight ratio was 10:1) with ethanol as a solvent. The ethanol was subsequently evaporated at 60 °C, and the milled powders were poured into a graphite crucible and heated at 1900 °C for 4 h with a heating rate of 15 °C/min in a 0.5 MPa Ar atmosphere in a graphite furnace (diameter 60 cm, depth 70 cm). After the heating, the obtained powders were fired at 650 °C under air atmosphere for 1 h to remove the excess carbon. The above powers were ground and sieved through a 200 mesh sieve, and the finer fraction of the powders (<200 mesh) was collected.

2.2. Preparation of SiO_{2f}/SiO_2 composites

Commercial available high-purity silica sol (Zhejiang University, Hangzhou, China) and the above synthesized SiC powders were used as raw materials. Mixtures of the silica sol and SiC powders with different weight ratios were fully stirred to form a slurry. Three-dimensional braided quartz fiber preforms with a fiber volume fraction about 44.7% were used as the reinforcements in this study. SiO_{2f}/SiO_2 composites were fabricated through a sol-gel process that included three stages. In the first stage, quartz fiber preforms were infiltrated with the slurry in vacuum. In the second stage, these preforms filled with slurry were dried at 150 °C for 4 h in vacuum. In the third stage, the dried preforms were pyrolyzed at 750 °C for 40 min with a heating rate of 3 °C/min under Ar atmosphere. These above steps were repeated nine cycles to densify the SiO_{2f}/SiO_2 composites.

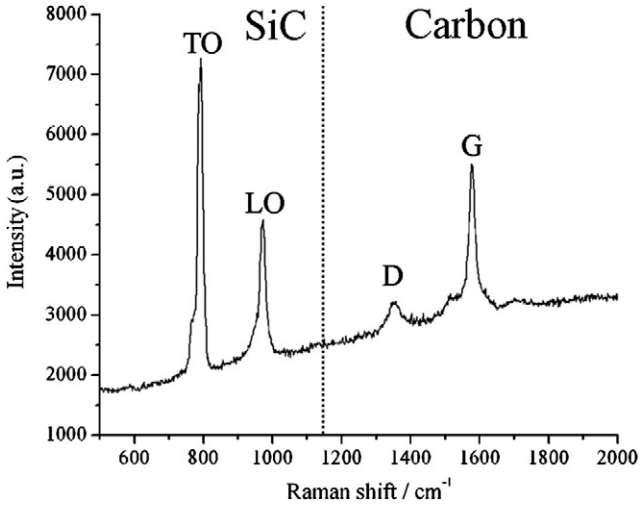

Fig. 2. Raman spectra of the synthesized silicon carbide powders.

2.3. Analytical methods

X-ray diffraction (XRD) was used to verify the crystallite characteristics of the synthesized SiC powders. XRD patterns of the powders were obtained from a D8 ADVANCE diffractometer using Cu Ka radiation ($\lambda = 0.15418$ nm) as the source. The scanned angles (in 2θ scale) were ranging between 20 and 80° with a step of 0.02° and a 13 s exposure time. X-ray diffractograms were indexed using the DIFFRAC+ software (Socabim), which contained JCPDS files database. The 99.99% Si ($a = 0.543088$ nm) was used as an inner standard. Raman spectra were obtained using a confocal laser micro-Raman spectrometer (Jobin Yvon LABRAM-HR) with a 514.5 nm line of laser. X-ray photoelectron spectroscopy (XPS) was used to investigate the surface chemical state of the synthesized SiC powders. The data were obtained with an ESCALab220i-XL electron spectrometer from VG Scientific using 300 W Al Kα radiation, with the base pressure about 3×10^{-9} mbar. The survey spectrum was collected from 0 to 1100 eV. The relative amounts of different atoms were estimated from respective areas of assumed Gaussian–Lorentzian curves. Transmission electron microscopic (TEM) analysis was performed on a JEOL JEM-2010 (Tokyo, Japan) operating at an acceleration voltage of 200 kV to analyze the microstructure of the SiC powders. The dielectric parameters of the SiC powders were measured with a network analyzer Agilent 8720ET. The sample for dielectric parameter measurements was prepared through dispersing

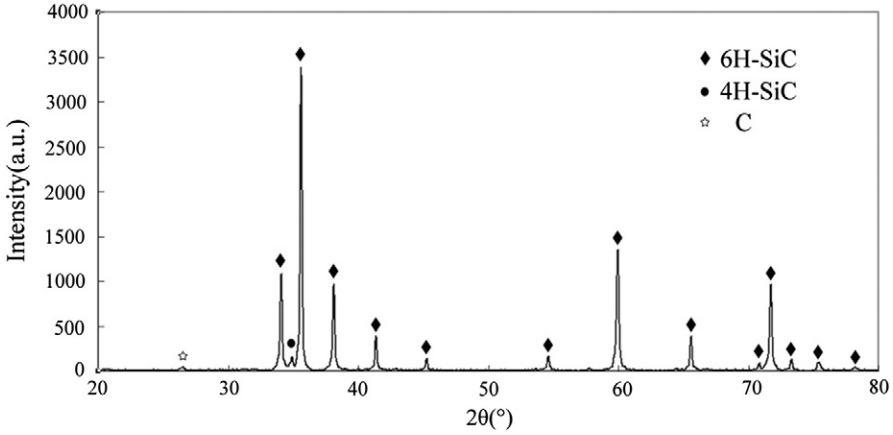

Fig. 1. X-ray diffraction pattern of the synthesized silicon carbide powders.