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Preparation and study on microwave absorbing materials of boron nitride coated pyrolytic carbon particles

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ABSTRACT

Boron nitride coatings were synthesized on pyrolytic carbon (BN-coated PyC) particles via chemical reaction of boric acid and urea in nitrogen. The results of Scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FI-IR) and X-ray photoelectron spectroscopy (XPS) show the formation of boron nitride coating. The TGA curves indicate that the oxidation resistance of the PyC particles is improved by incorporating BN coating on the surface. The mass of the BN-coated PyC particles remains over 60% at 1200 °C whereas the PyC particles are oxidized completely at 920 °C. The investigation of microwave absorbing property reveals that compared with the PyC particles, the BN-coated PyC particles have lower permittivity (ε', ε'') and better absorbing property. The BN-coated PyC particles show a strong absorbing peak at 10.64 GHz, where the lowest reflectivity −21.72 dB is reached. And the reflectivity less than −10 dB is over the range of 9.6–12 GHz.

1. Introduction

In recent years, materials that can absorb electromagnetic waves have been increasingly investigated for the explosive growth in the utilization of electrical and electronic devices for industrial and commercial applications [1–4]. In view of these utilizations, excellent microwave absorbing materials should exhibit strong microwave absorption properties over a wide frequency range and need to be thin and light weight, especially for aircrafts [5].

Carbon fiber reinforced carbon matrix composites (C/Cs) with low density and excellent high-temperature properties [6] are considered as candidates in the development of potentially revolutionary microwave absorbing materials. However, low electrical resistivity (10^{-2} Ω cm) and strong reflectors of radar-waves of C/Cs limit their applications as microwave absorbing materials. It is reported that the modified carbon could be a good microwave absorber to dissipate electromagnetic energy into heat by space charge polarizations and vibrational scattering [7]. Many treatments [8–12] have been used to enhance the microwave absorption property of carbon fibers. But to our knowledge there is limited information being reported on the modification of PyC matrix of C/Cs for microwave absorbing properties improvement.

Boron nitride which has low dielectric constant (5.16) and dielectric loss (0.0002) is suitable to serve as microwave-transparent material [13]. The impedance matching of PyC can be improved by coating BN on the surface, and thus enhances its microwave absorption property. In addition, by incorporating BN coating on PyC can also improve its oxidation resistance due to the higher initial oxidation temperature of BN than that of PyC and the formation of a protective B_2O_3 layer at higher temperatures [14,15].

In the present work, the BN coating was synthesized on the surfaces of PyC particles, which was expected to enhance the microwave absorption property of PyC particles and make C/Cs probable to be applied as good microwave absorbing materials. BN coatings were produced via chemical reaction of boric acid (H_3BO_3) and urea ($CO(NH_2)_2$) in a nitrogen-gas atmosphere. The microstructure, oxidation resistance and microwave absorption property of BN-coated PyC particles were investigated in detail.

2. Experimental

2.1. Sample preparation

PyC particles were crushed from bulk material in an agate mortar and then sieved (150 μm). The bulk PyC was prepared by chemical vapor infiltration (CVI) at 1000 °C under 1 kPa pressure. C_3H_6 and N_2 were used as carbon resource and dilute gas, respectively. Boric acid (reagent grade, Xilong Chemical Co., Ltd, Shan'tou China) and urea (reagent grade, Xilong Chemical Co.) were used to produce the BN precursor to coat the PyC particles. The experimental procedure is presented schematically in Fig. 1. Boric acid and urea with the mole ratio of 1:3 were dissolved in non-aqueous ethanol to form a solution. Then the PyC particles were dispersed into the solution. This mixture was milled by using the wet

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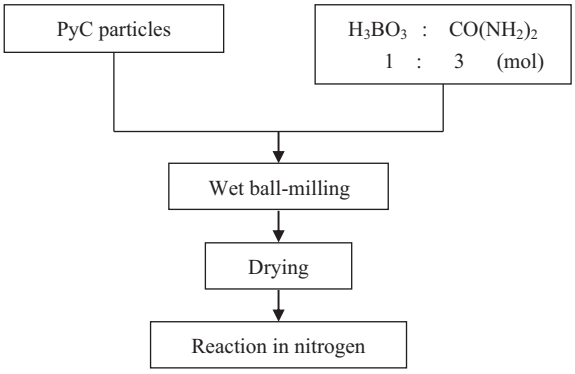

Fig. 1. Flow chart of fabrication process of BN-coated PyC particles.

ball-milling method with agate balls and ethanol in a plastic bottle for 24 h. After drying, the BN precursor-covered particles were heated at 850 °C in nitrogen gas for 6 h. Under this condition, the boric acid reacted with urea and BN-coated PyC particles were obtained. After reaction, the weight content of BN coating in composite particles is 31 wt% by adjusting the ratio of boric:urea:PyC.

2.2. Characterization

The surface morphologies of the BN coated and uncoated PyC particles were investigated using scanning electron microscopy (SEM, Nova NanoSEM 230). The chemical compositions of BN coating were analyzed by Fourier transform infrared spectroscopy (FI-IR, IS10, Thermo scientific) and X-ray photoelectron spectroscopy (XPS, K-Alpha 1063) with Al Kα radiation. The oxidation resistance of the BN coated and uncoated PyC particles were studied in air up to 1200 °C in a DTA/TGA instrument (STA 409PC, NETZSCH). The complex permittivity (ε', ε'') and complex permeability (μ', μ'') of the BN coated and uncoated PyC particles were measured by the coaxial line method in the frequency range of 2–18 GHz using a network analyzer (AV3618). For permittivity and permeability measurement, the samples were prepared by mixing the particles with molten paraffin (mass ratio: 1:4), using a mould with 7.0 mm outer diameter and 3.0 mm inner diameter. The reflection loss was calculated by using the measured values of the complex permittivity (ε', ε'') and the complex permeability (μ', μ'') of the BN coated and uncoated PyC particles based on the transmission line theory.

3. Results and discussion

The surface morphologies of the BN coated and uncoated PyC particles investigated by SEM are shown in Fig. 2. As can be seen from Fig. 2, the surface morphology of the BN coated PyC particles becomes rough while compared with the smooth surface of the uncoated PyC particles. The PyC particles are covered by blocky-shaped BN particles interconnected with each other. The size of the BN particles ranges from tens of nanometers to hundreds of nanometers (shown in Fig. 2(c)).

The chemical compositions of the as-received BN-coated PyC particles were identified by FI-IR spectra and XPS, as shown in Figs. 3 and 4, respectively. For the BN precursor-covered PyC particles, the FI-IR spectrum shows the N—H and O—H bands in the range of 2800–3600 cm^{-1} and the N—C=O band at 1620 cm^{-1} (Fig. 3(b)). After reaction at 850 °C, the bands detected near 800 and 1380 cm^{-1}

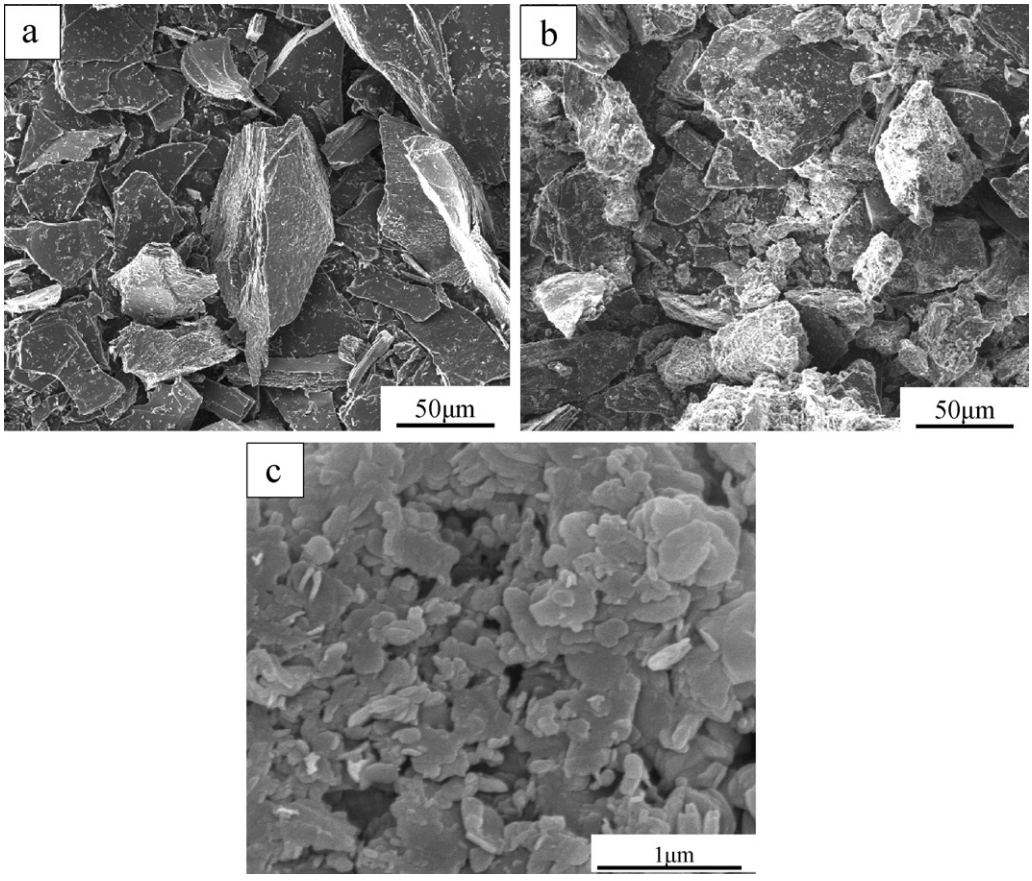

Fig. 2. SEM images of (a) the PyC particles, (b) the BN-coated PyC particles and (c) BN coating.