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Microstructure and mechanical properties of boron nitride nanosheets-reinforced fused silica composites

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ABSTRACT

In order to overcome intrinsic brittleness and poor mechanical properties of fused silica (FS), boron nitride nanosheets (BNNSs) as a novel reinforcement were employed for fabrication of BNNSs/fused silica composites. BNNSs with micron lateral size were homogeneously dispersed with FS powder using a surfactant-free flocculation method and then consolidated by hot pressing. The flexural strength and fracture toughness of the composite with the addition of only 0.5 wt.% BNNSs increased by 53% and 32%, respectively, compared with those of pure FS. However, for higher BNNSs contents the improvement in mechanical properties was limited. Microstructural analyzes have shown that the toughening mechanisms are combinations of the pull-out, crack bridging, and crack deflection mechanisms.

1. Introduction

Two-dimensional nanosheets, such as graphene, boron nitride nanosheets (BNNSs) possess a large specific surface area, two dimensional high aspect ratio sheet geometry and outstanding mechanical properties; therefore, it is promising as a strengthening agent [1–4]. Graphene has been considered as an attractive reinforced nanofiller to polymers [5,6], metals [7] and ceramics [8–11] to produce composites with enhanced mechanical properties.

Due to a structural analogy with graphene, BNNSs share common properties with it, like high mechanical strength, excellent thermal conductivity, and good lubrication. It should be noted that BNNSs have some advantages over graphene. Firstly, the oxidizing temperature of BNNSs is higher than 800 °C, and graphene can be easily oxidized around 450–500 °C in an air atmosphere, so BNNSs are suitable for high-temperature applications [12–14]. Secondly, BNNSs can retain the good dielectric properties of composites, while graphene ruins the electrical insulation of the matrix [15]. Lastly, the white appearance of BNNSs makes it feasible to keep the intrinsic colors of the matrix. Thus it is not surprising that an increasing interest has been focused on BNNSs.

The aforementioned advantages make BNNSs more suitable for reinforcing composites applied at some special environment. A number of studies with polymer-based matrices have demonstrated that BNNSs can significantly improve the mechanical properties of polymers [16–19]. However, to the best of our knowledge, only relatively few reports have so far been published on the use of BNNSs additives to improve the mechanical properties of ceramic matrix composites [20]. It should be noted that boron nitride nanotubes (BNNTs) have been used in several ceramic matrices as reinforcement showing good improvement in mechanical properties [21–23]. But the difficulty in large-scale synthesis of BNNTs with high purity and quality significantly restricts the research on BNNTs-reinforced ceramics. In contrast, BNNSs can be synthesized by facile top-down processes from *h*-BN. Furthermore, BNNTs are utilized only at the outer surface of their structure when they are dispersed in the matrix, whereas BNNSs have a two-dimensional structure which can enhance the toughness by maximizing the interfacial area in contact with the matrix [6,24]. As a result, small amounts of BNNSs in a ceramic matrix may lead to large improvements in the composite properties.

Fused silica is an irreplaceable electromagnetic wave transparent material with low and stable dielectric constant (3–4), but the relatively low mechanical strength limits its reliability [25]. For this reason, it is important to incorporate a nanofiller to improve the mechanical properties without worsening the wave-transparent efficiency. BNNS with excellent mechanical strength and relatively low dielectric constant is an ideal nanofiller to reinforce fused silica based electromagnetic wave transparent materials.

Homogeneous dispersing of nanofillers is important in terms of efficient stress transfer between different components. Graphene and BNNSs have a tendency to agglomerate and restack due to their

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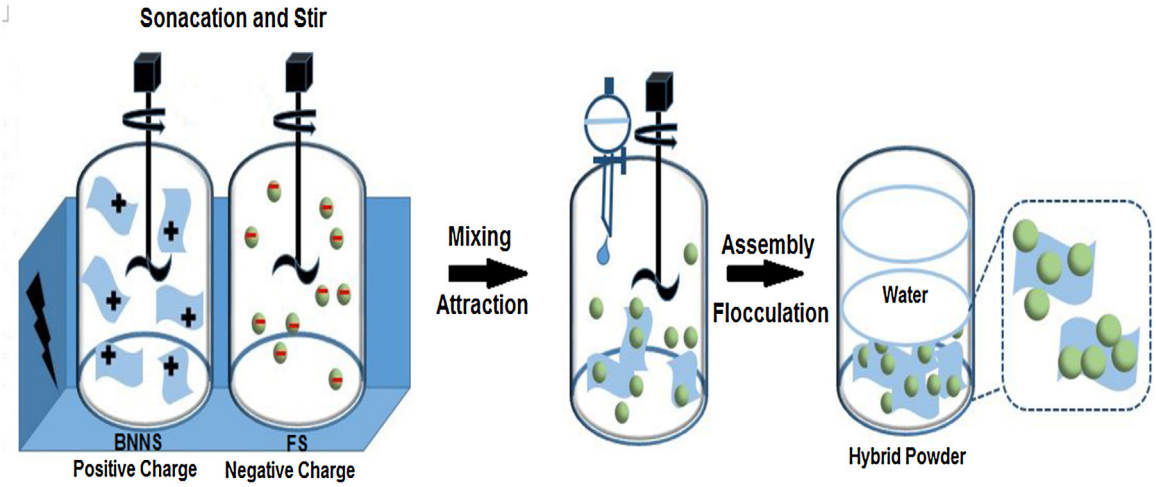

Fig. 1. Schematic illustration of the fabrication process of BNNSs/FS hybrids.

high surface energy and typically weak interactions with solvents or matrix components [26]. As a result, one of the main obstacles in processing BNNSs-reinforced ceramic matrix composites lies in the difficulty of dispersing BNNSs homogeneously throughout the matrix since agglomerate would deteriorate the mechanical properties of composites. Surface modification is a common method to tackle this problem. Graphene can be oxidized or modified with different active groups to form chemical bond with the dispersion solution or matrices. In comparison with graphene which can utilize many well-established chemical reactions to realize effective surface modifications, the chemical reactions for B and N elements are much more limited [27]. Surfactant-assisted colloidal method is another effective method to obtain homogenously dispersed reinforcements in the matrix. However, there have been no systematic studies into surfactant removal or the effects of residual surfactant on the properties of composites. In view of the present scenario, the homogeneous dispersing of BNNSs is still a challenging research topic.

In this paper, hence, for the first time, we report on the development of fused silica based composites reinforced by BNNSs with micron lateral size. The objective of the present study is to fabricate BNNSs/FS composites with improved BNNSs dispersion and investigate its microstructure and mechanical properties. For the homogeneous dispersion of BNNSs in FS powder, a surfactant-free flocculation method was adopted. Then dense bulk composites were prepared from the powder mixtures by hot pressing. Toughening mechanisms for enhanced mechanical properties of BNNSs/FS composites are presented.

2. Experimental procedure

2.1. Fabrication of BNNSs/fused silica composites

BNNSs were prepared by the liquid-exfoliation method reported elsewhere [28]. Typically, 3 g *h*-BN powder (Alfa Aesar) was added to 2000 mL beaker. 1000 mL of ethanol/water with ethanol volume fractions of 55% was added as dispersion solvent. The sealed beaker was sonicated for 12 h, and then the dispersion was centrifuged at 3000 rpm for 20mins to remove aggregates.

The BNNSs/FS hybrids were deagglomerated and uniformly dispersed to obtain a stable aqueous colloidal suspension through a combination of surface modification through pH adjustment process and agitation in conjunction with sonication treatments, as shown in Fig. 1. The commercially available-FS powder (Donghai Fucai Co., Ltd., China), with a purity of 99.6%, was used in this study. The as-received BNNSs (1 g/L) and FS powder (12 g/L) were separately dispersed in deionized water by stirring in a high speed in conjunction with ultrasonic treatment for 6 h. The pH value of BNNSs colloid and FS colloid were then adjusted to around 3 using HCl solution (1 M). Zeta potential of BNNSs colloid (12.6 mV) and FS colloid (-13.4 mV) are opposed, which gives effective electrostatic interactions between them. The suspensions were gently mixed by introducing the BNNSs colloid to the FS colloid drop by drop while vigorously stirred. The electrostatic interactions process was considered completed once the supernatant became transparent. The precipitate was separated by vacuum filtration followed by drying at 80 °C. Then the mixtures were placed into a graphite die with a diameter of 30 mm, and hot pressed at 1300 °C in a multipurpose high-temperature furnace (Fujidenpa Kogyo Co., Ltd., Osaka, Japan) under a pressure of 30 MPa in argon atmosphere for 1 h. Different amounts of BNNSs were added into the batches (0, 0.5, 1.0, 1.5 and 2.0 wt.%) and the corresponding composite was denoted as 0.0BNNS/FS, 0.5BNNS/FS, 1.0BNNS/FS, 1.5BNNS/FS and 2.0BNNS/FS, respectively.

2.2. Characterization

The relative density of the composites and pure ceramic was measured by Archimedes' method in deionized water, where the theoretical densities for FS and BNNSs were 2.3 and 2.1 g/cm^3, respectively. The density of BNNSs are assumed based on the density of bulk *h*-BN.

A three-point bending test with a load speed of 0.5 mm/min was performed to record the flexure strength responses of the hot-pressed samples ($3 \times 4 \times 25$ mm^3 with precisely parallel flat surfaces, four specimens each) had been attached to their polished, scratch-free tension surfaces. And three-point bending test at a load speed of 0.05 mm/min was performed to directly measure the fracture toughness (K_{IC}) of four notched specimens ($3 \times 4 \times 25$ mm^3) via single-edge notched beam (SENB) method with a notch of 2×0.3 mm^2 at the center.

Microstructural analyses of BNNSs and BNNSs/FS composites were examined via a SU-70 type thermal field emission scanning electron microscope (FESEM). A JEOL JEM-2100 high-resolution transmission electron microscope was used to characterize the pristine BNNSs and to investigate the microstructure and bonding between BNNSs and FS matrix. Crystalline phases of the hot pressed bodies were determined by XRD analysis on a Rigaku Dmax-rcdiffractometer with Ni-filtered Cu Kα radiation at a scanning rate of 4° min^{-1}. Fourier transformation infrared (FTIR) spectra