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Original Article

Fabrication and mechanical properties of *h*-BN based composites containing dual glass phases

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ABSTRACT

In this work, *h*-BN based composites containing amorphous silica and ytterbium silicate glass phases were successfully fabricated by *in situ* hot pressing process. The powder mixtures of *h*-BN, Yb_2O_3, SiO_2 and sintering additive (Al_2O_3) were hot pressed at 1880 °C for 1 h under 30 MPa in Ar atmosphere. In the composites, β-$Yb_2Si_2O_7$ phase produced from the reaction between Yb_2O_3 and SiO_2 disappears completely and transforms to Yb-Si-Al-O glass when the amount of Al_2O_3 is ≥1.5 wt.%. The Vickers hardness, flexural strength, and compressive strength reached the maximum values of 2.38 ± 0.08 GPa, 337 ± 22 MPa and 950 ± 34 MPa, respectively, as 1.5 wt.% Al_2O_3 was added. The strengthening effects were attributed to the fine spherical microstructure of Yb-Si-Al-O glass particles, strong [AlO_4] coordination state, and residual compressive stress in glass phases.

1. Introduction

Hexagonal boron nitride (*h*-BN) is well-known as an important ceramic material for its salient properties, such as high refractoriness, thermal stability, high thermal shock resistance [1–7], electrical insulating properties, chemical inertness, good machinability and so on [8–12]. Such unique properties make it possible to be used in many fields. However, due to the layered structure analogous to graphite, B and N atoms are bonded by strong covalent bonds within each layer, and it is difficult to be densified by traditional sintering techniques. Besides, some shortcomings such as low hardness and strength limit its widespread application as a structural ceramic.

A series of works have reported that incorporation of a second phase is an effective way to overcome those weaknesses mentioned above and improve other properties of *h*-BN material [3,13–25]. For example, ZrO_2 was selected to improve mechanical properties and wear resistance of *h*-BN [6,26]. However, the mismatch of thermal expansion coefficients (CTE) between *h*-BN and ZrO_2 causes the possibility of failure of *h*-BN-ZrO_2 composites. Trice et al. [19] introduced $Y_2Si_2O_7$ into *h*-BN and attained the BN-$Y_2Si_2O_7$ composites. It was demonstrated that the flexural strength could increase by increasing either alignment of individual *h*-BN grains or the content of additives. Similar to that, the mechanical properties, especially high-temperature mechanical properties, were improved by introducing Y_2SiO_5 phase to the *h*-BN matrix [20,21]. Wen et al. [13] prepared a series of *h*-BN/fused silica composites by hot-pressed sintering method. When the composite contained 40 vol.% fused silica, the highest relative density reached 99%, and the optimal flexural strength was 246 MPa. Tian et al. [23] investigated the *h*-BN/30 vol.% SiO_2 composite with different contents of AlN. When 5 vol.% AlN was added to the composite, the phase compositions include BN, SiAlON and amorphous silica. Under the combined effects of SiAlON and amorphous silica, both the flexural strength and fracture toughness achieve the maximum values at the same time.

As mentioned above, *h*-BN based composites containing hard particles and/or a kind of glass phase have been widely investigated. And it can be found that glass phase especially amorphous silica usually presents better strengthening effect. In this study, *h*-BN based composites containing dual glass phases were fabricated by *in situ* hot pressing process for the first time. Amorphous silica and ytterbium silicate glass phases were simultaneously introduced into the *h*-BN matrix. Vitrification effect of Al_2O_3 on ytterbium silicate glass, microstructure and binding energy evolution were systematically investigated. The room-temperature mechanical properties were measured and the strengthening mechanisms were discussed.

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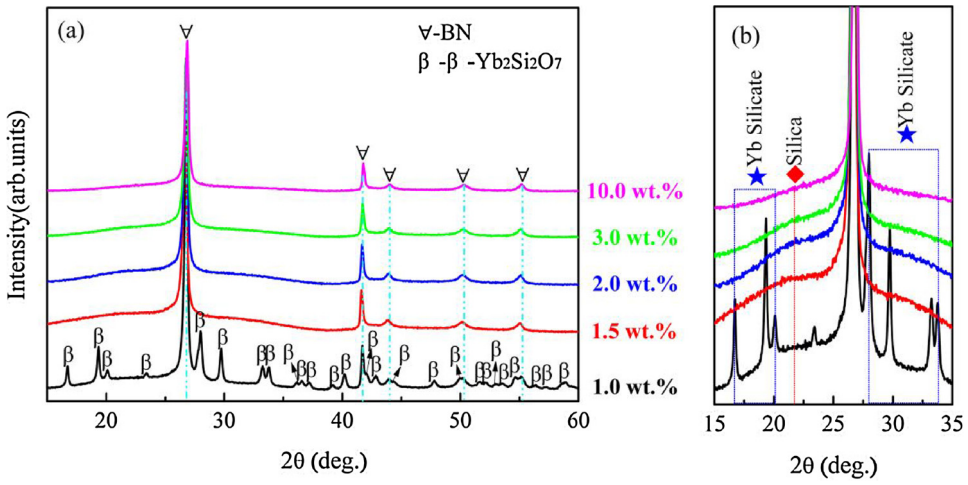

Fig. 1. XRD patterns of *h*-BN based composites with different amounts of Al_2O_3 as sintering additive.

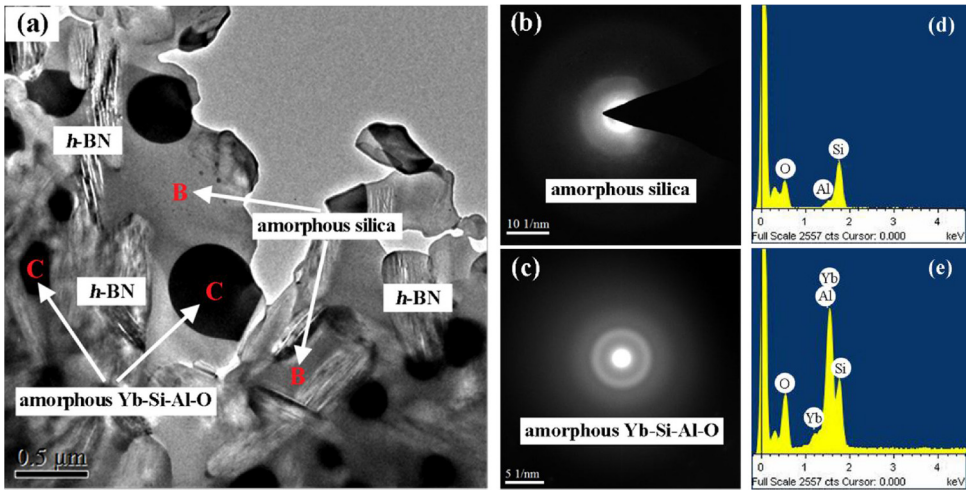

Fig. 2. (a) Bright field image of YBSB-1.5Al_2O_3; (b) selected area electron diffraction pattern of "B" in (a); (c) selected area electron diffraction pattern of "C" in (a); (d) EDS spectrum corresponding to "B"; (e) EDS spectrum corresponding to "C".

2. Experimental procedure

2.1. Materials fabrication

In this work, *h*-BN based composites containing amorphous silica and ytterbium silicate glass phases were fabricated by *in situ* hot pressing process. The initial volume ratio of BN:SiO_2:$Yb_2Si_2O_7$ was designed as 67:28:5. Commercially available powders of *h*-BN (98%, 0.7 μm), Yb_2O_3 (99.9%, 3 μm) and SiO_2 (99%, 1 μm) were selected as primary materials. Yb_2O_3 and SiO_2 can firstly react with each other to form $Yb_2Si_2O_7$ phase during the heating process ($Yb_2O_3 + 2SiO_2 \rightarrow Yb_2Si_2O_7$) [27]. And Al_2O_3 was used as sintering additive to accelerate the vitrification process of $Yb_2Si_2O_7$ phase. In order to describe conveniently, the composites in this study were abbreviated to YBSB-$x$$Al_2O_3$ (x wt.%) hereafter. Various amounts of Al_2O_3 ranging from 1.0 to 10.0 wt.% was added to obtain a series of *h*-BN based composites.

The starting powders were weighed and ball-milled in a Si_3N_4 jar with Si_3N_4 balls and ethanol as media for 6 h. Thereafter, the obtained powders were dried at 60 °C for 12 h, and then passed through an 80 mesh sieve. Raw powders were compacted uniaxially under 10 MPa in a graphite mold pre-sprayed with a layer of *h*-BN. The compacted mixtures were heated at the rate of 8 °C/min from room temperature to 1880 °C and held for 1 h under an flowing Ar atmosphere in a furnace using graphite as the heating element, while the pressure was increased to 30 MPa gradually, then the sample was cooled down to room temperature in the furnace. Finally, samples with dimensions of $\phi 50 \times 5\,mm^3$ were obtained. The as-prepared sample surfaces were machined off ; in order to remove the contaminants before characterization.

2.2. Materials characterization

Phase compositions were identified by a step-scanning X-ray diffractometer (D/max-2400; Rigaku, Tokyo, Japan) with CuK_α radiation. The polished surfaces were observed using SUPRA 35 scanning electron microscope (SEM, LEO, Oberkochen, Germany). The microstructure and electron diffraction were investigated by 120 kV Tecnai Spirit T12 transmission electron microscope (TEM, FEI, Eindhoven, Netherlands). The apparent density was measured by Archimedes method.

The X-ray photoelectron spectroscope (XPS) analysis was carried out by Surface Analysis System (ESCALAB250, VG) using monochromatic Al-Kα radiation (HV = 1486.6 eV). All spectra were calibrated by the standard energy of C 1s peak (284.6 eV). The XPSPEAK41 software program was used to fit all XPS spectra.

The Vickers hardness was measured at a load of 4.9 N with a dwell time of 15 s, the given value is the average of seven separate measurements. Three-point bending tests were performed to measure the flexural strength. The distance of the fixture for three-point bending test is 30 mm and the sample size is $3 \times 4 \times 36\,mm^3$. The crosshead speed