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Microstructures and mechanical properties of B_4C–SiC intergranular/intragranular nanocomposite ceramics fabricated from B_4C, Si, and graphite powders

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Abstract

B_4C–SiC intergranular/intragranular nanocomposites with high hardness and high toughness were fabricated through mechanochemical processing with B_4C, Si, and graphite powders and subsequent hot pressing without any sintering aid. The milled powders are composed of stacking-disordered SiC and nanocrystalline B_4C. Most nano/micron-sized SiC particles are homogeneously dispersed in B_4C matrix, and some nano-sized SiC and B_4C particles are embedded into B_4C grains to form an intergranular/intragranular structure. The disordered characteristic of the milled powders is the essential factor for the formation of the intragranular structure, sudden densification within the narrow temperature range (1700–1900 °C), and the preparation of dense samples under a relatively low temperature (1900 °C). The relative density, Vickers hardness, and fracture toughness of the samples sintered at 1950 °C are 98.6%, 34.3 GPa, and 6.0 MPa $m^{1/2}$, respectively. The intergranular/intragranular structure plays an important role in improving fracture toughness and hardness of the composites.
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Keywords: B_4C–SiC nanocomposite; Intergranular/intragranular structure; Mechanochemical processing; Disorder–order transformation; Toughening mechanism

1. Introduction

Boron carbide (B_4C) ceramics are important high-tech engineering materials mainly due to their outstanding physical and mechanical properties, especially their extremely high hardness and low density.[1–3] However, low fracture toughness and ultrahigh sintering temperature severely impede their further development. Numerous attempts have been made to overcome these disadvantages, such as the adoption of nano/submicron-sized powders as raw materials or the introduction of a second phase into the B_4C matrix.[4–6] But B_4C powders obtained through industrial manufacturing technology are usually micron-sized.[7] These coarser powders cannot satisfy the requirements of sintering. Although nano/submicron-sized B_4C powders can be obtained through some advanced techniques, such as vapor-phase reactions, synthesis from elements, ion beam synthesis, etc.,[1] due to high cost and low yield, these techniques are limited to laboratory study. As for the second phase, different materials such as TiB_2, CrB_2, ZrB_2, SiC, TiC, Al_2O_3, etc. have been used to improve fracture toughness and lower the sintering temperature of B_4C, but the improvement effects are limited.[4,8–12] For example, the addition of Al_2O_3 into the B_4C matrix can improve fracture toughness but lower hardness of B_4C due to the low hardness of Al_2O_3. Moreover, the addition of TiB_2, CrB_2, or ZrB_2 into the B_4C matrix seriously destroys the lightweight property of B_4C. Among these materials, SiC is a hard and light material with relatively high toughness. B_4C–SiC systems have the potential to offer the combination of high hardness, relatively high fracture toughness, and lightweight property. Therefore, SiC is one of the best candidate materials.

However, SiC powders obtained through industrial manufacturing are also micron-sized and SiC also possesses poor sinterability. It is therefore difficult to fabricate dense B_4C–SiC composites or more dense nanocomposites through the traditional processing route of mixing commercial SiC and B_4C powders and subsequent solid-state sintering. Although B_4C–SiC composites have been obtained through the infiltration

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process of a green B_4C compact with molten silicon, the reliability and high temperature properties of products are extremely inferior due to the inhomogeneous structure and abundant unreacted Si contained in products.[13–15] The products prepared by the infiltration process therefore lose partial superior properties of B_4C–SiC composites. B_4C–SiC nanocomposite ceramics obtained by solid-state sintering are seldom reported. In view of the specific properties and potential applications of B_4C–SiC composites, it is meaningful to study whether the dense B_4C–SiC nanocomposite ceramics can be fabricated through solid-phase sintering under the relatively low temperature.

Recently, high-energy ball milling has received considerable scientific attention in the ceramic field because it can offer a simple, practical, industrial, economically variable but powerful way to prepare highly active nanocomposite powders. It has been confirmed that the powders obtained by milling possess disordered structure and high sintering activity.[16–19] Some composite ceramics with improved properties, such as $SiB_{0.5}C_{1.5}N_{0.5}$, SiC–BN, TiC–Ti_3SiC_2, TiB_2–TiN, and SiO_2–SiC, have been synthesized successfully by milling and subsequent hot pressing or spark plasma sintering (SPS).[16,18,20,21] However, such studies concerning B_4C–SiC composites are seldom reported.

In our previous paper,[19] we have prepared B_4C–SiC composite ceramics by mechanical milling with coarser B_4C and SiC powders as starting materials and subsequent hot pressing. However, both the B_4C and SiC particles in the milled powders were of submicron size because of their high hardness and low energy of ball milling. It is impossible to obtain B_4C–SiC nanocomposites with the submicron-sized powders. As a result, both SiC and B_4C dispersed in B_4C–SiC composites were >5 μm. The goal to obtain B_4C–SiC nanocomposites has not been realized. In this paper, in order to fabricate B_4C–SiC nanocomposite powders, which are the foundation and condition to obtain nanocomposites, the energy of ball milling is increased. More importantly, a novel synthesis method of B_4C–SiC nanocomposite powders, through mechanochemical processing with B_4C, Si, and graphite powders as starting materials, is adopted to prepare the homogeneously dispersed B_4C–SiC nanocomposite powders. The component of SiC in the B_4C–SiC nanocomposite powders is synthesized with softer Si and graphite through mechanochemical processing. Subsequently, with this kind of B_4C–SiC nanocomposite powders, the dense B_4C–SiC intergranular/intragranular nanocomposites with high hardness and high fracture toughness are fabricated successfully by hot pressing at relatively low temperatures without any sintering aid.

The compositions and microstructure of both milled powders and composite ceramics are analyzed by X-ray diffraction (XRD), scanning electron microscopy (SEM), and high-resolution transmission electron microscopy (HRTEM) with energy-dispersive X-ray spectroscopy (EDS). The effects of sintering temperature on relative density and mechanical properties are discussed. The relationships of microstructure and mechanical properties are also investigated. Meanwhile, toughening mechanisms of the composites are also discussed in detail.

2. Experimental

2.1. Synthesis of B_4C–SiC nanocomposite powders

The starting powders used in this study were 97% pure B_4C powder with an average particle size of 3 μm (Jingangzuan Boron Carbide Co., Ltd., China), 99.99% pure Si powder with a sieve size −200 mesh (Sinopharm Chemical Reagent Co., Ltd., China), and 99% pure amorphous graphite with an average particle size of 1 μm (Sinopharm Chemical Reagent Co., Ltd., China). The powder mixtures were composed of B_4C, Si, and amorphous graphite with a mass ratio of 40:7:3, corresponding to milled powders of B_4C −20 wt.% SiC according to the reaction:

$$Si + C - SiC$$

They were then mixed in a QM-2SP20-CL Planetary Ball Milling Machine (Nanjing Nan Da Instrument Plant, China). Milling was completed with stainless steel balls (Φ 12 mm) and vials (ID = 90 mm; height = 80 mm). Total milling time was 96 h and every 6-h milling was followed by 1-h cooling. With 25 g of mixed reactants and 1 mL of ethanol as process control agents, the ball–powder mass ratio was fixed to be 30:1. The rotation speed was 350 rpm. Stainless steel vials were evacuated to 0.2 Pa and flushed with Ar gas. The possible Fe contamination in the milled powders was removed through leaching in 2 M hydrochloric acid for 6 h. The solution was filtered after leaching and the purified products were washed three times with distilled water to remove residual hydrochloric acid. Then, the powders were dried in a vacuum at 60 °C for 48 h. The transfer operation of the powders was completed in a glove box with argon.

In order to verify the components of powders milled for 96 h, heat treatment was performed in a homemade high-temperature tubular resistance furnace. The resistance furnace was heated from room temperature to 900 °C in a flowing argon atmosphere. The heating rate was 3 °C/min with 60-min dwell.

2.2. Preparation of B_4C–SiC nanocomposite ceramics

A hot-pressing sintering apparatus (916G-G Press, Thermal Technology LLC, USA) was utilized to consolidate the B_4C–SiC composites. B_4C–SiC composite powders milled for 96 h were placed in a cylindrical graphite die (ID = 48 mm). A piece of 0.2-mm-thick graphite foil was used to wrap the sample. A uniaxial pressure of 30 MPa was applied through top graphite plungers. The samples were heated to 1400 °C at a heating rate of 20 °C/min in vacuum and then were heated to the selected temperature at a heating rate of 10 °C/min in argon gas with 60-min dwell. Then, the samples were cooled to room temperature by turning off the power. The sintered samples were in the form of 4-mm-thick disks with a diameter of about 48 mm. A diamond paste was used to polish the samples to reach the 0.3-μm surface finish for the measurement of Vickers hardness and fracture toughness. The polished surfaces were electrochemically etched in 0.1% KOH solution with current density of 0.1 A/cm^2 for 90 s for microstructural analysis.

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