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Fabrication and mechanical properties of silicon nitride laminate composites

Dong-Woo Shin* and Hai Guo

Ceramic Matrix Composites (CMCs) Lab., Division of Advanced Materials Science and Engineering, Engineering Research Institute, Gyeongsang National Univesity, Jinju, Gyeongnam 660-701, Korea

A compact material with a laminated structure similar to the structure of nacre has been successfully made using a procedure described in this paper. This laminated structural design provides high-fracture toughness to Si_3N_4/BN-Al_2O_3 ceramic composites. The dense Si_3N_4 laminate (99.6% of theoretical density) had a flexural strength of 500 Mpa and a fracture toughness of 15 MPa·m$^{1/2}$. The introduction of reinforcement (15 wt% SiC whiskers) within the Si_3N_4/BN-Al_2O_3 laminated ceramic composites further increases the strength and fracture toughness values to 650 MPa and 20 MPa·m$^{1/2}$, respectively.

Key words: Silicon nitride, laminate composites, fracture toughness, SiC whiskers.

Introduction

The high strength to weight ratio, and the high stiffness, and oxidation resistance make Si_3N_4 an important structural material. A serious drawback, however is its brittle failure behavior. Tremendous efforts have been made to improve the fracture toughness of Si_3N_4 ceramics either by the addition of an reinforcement phases such as whiskers and particulates, or by designing a fine crystalline structure [1]. Natural biomaterials, such as nacre, bamboo and wood because of their composite structures exhibit excellent mechanical properties [2-4]. This provides a possible way to improve the fracture toughness of ceramics through bio-mimetic designing.

Nacre having a laminate structure is composed of aragonite sheets with protein layers between them (Fig. 1). An important characteristic of this structure is the coordination of different toughening mechanisms on different scales. The first fine structure is a laminated structure, and a secondary fine structure is the aragonite platelet reinforcing the inorganic sheets. These fine structures give nacre high fracture toughness.

Clegg et al. invented a novel and powerful method to fabricate a ceramic composite with a laminated structure [5]. They stacked together SiC sheets and graphite and improved the fracture toughness from 3.6 to 15 MPa·m$^{1/2}$. Additional work has also been done on this system [6-8]. All illustrated the potential of this reinforcing method in improving the fracture toughness.

Anisotropy provided an easier method to design the microstructure of sheets in laminated ceramics. Kiyoshi introduced the concept of anisotropy in laminated ceramics and developed a novel process to control the microstructure of self-reinforced Si_3N_4 [9-10]. The key point of this process was that the size and distribution of elongated large grains were controlled by seeds of rodlike β-Si_3N_4 single crystals having a preferred orientation through tape casting. Si_3N_4 ceramics fabricated by this method showed an improvement in fracture toughness to the order of 11 MPa·m$^{1/2}$.

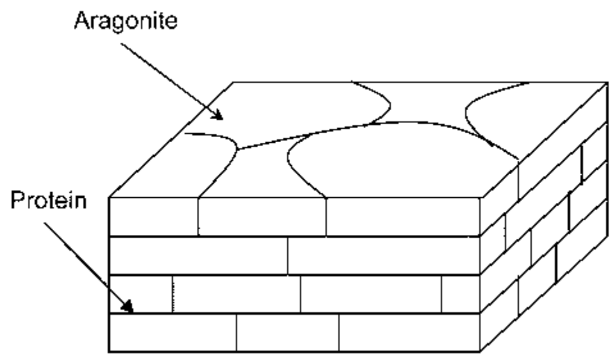

Fig. 1. Schematic drawing of the nacre structure of a mollusk shell.

Si_3N_4 laminate composites have been processed in a similar manner to the structure of nacre in the present paper. β-Si_3N_4 single crystals and SiC whiskers (SiC_w) were added to sheets to act as the secondary toughening phase. BN was selected as the separation layer, in which Al_2O_3 was added to adjust the interfacial strength.

Experimental Procedures

The processing flow chart of laminated Si_3N_4 ceramics is shown in Fig. 2. β-Si_3N_4 seeds used in this study were prepared by heating a powder mixture of 90 wt% α-Si_3N_4, 7 wt% Y_2O_3, and 3 wt% Al_2O_3 at 1850 °C for

*Corresponding author:
Tel : +82-55-751-5332
Fax: +82-55-755-6755
E-mail: dwshin@gsnu.ac.kr

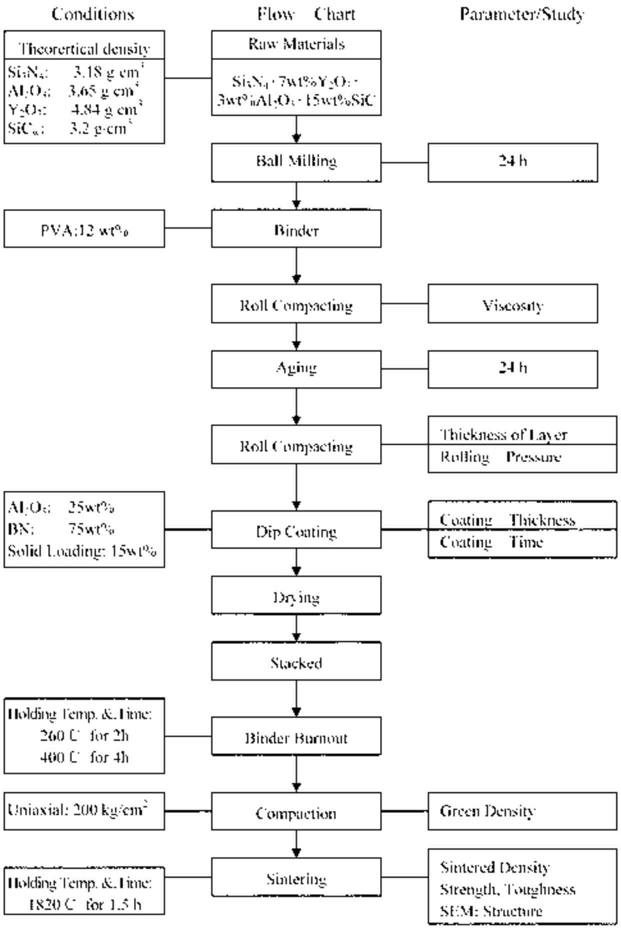

Fig. 2. Flow chart describing the fabrication procedure of laminated Si_3N_4 ceramics.

1.5 h under a nitrogen pressure of 0.5 MPa [5]. The synthesized seeds consisted of rodlike β-Si_3N_4 single crystals with a mean diameter of 0.5 mm and a mean length of 4.0 mm. Sheets were formed using 12 wt% poly (vinyl) alcohol (PVA) as the organic binder, 3 wt% glycerol as the plasticising agent, and a ball-milled mixture of α-Si_3N_4 (Founder Corporation, Beijing, China), 7 wt% Y_2O_3 (purity > 99.9%, Hokke Chemicals, Tokyo, Japan), 3 wt% Al_2O_3 (purity > 99.9%, Founder Corporation, Beijing, China), and 15 wt% SiC whiskers (TWS-400, Hokke Chemicals, Tokyo, Japan) or 3 wt% β-Si_3N_4 seeds. The sheets were formed first through coarse compact rollers and then through fine compact rollers. The green sheets were 0.2 mm thick. The sheets were cut and coated with an interfacial slurry consisting of 75 wt% BN and 25 wt% Al_2O_3 slurry. The green sheets were cut (32 mm × 38 mm) and stacked in a particular order in a graphite die. Heat treatments at 260 °C for 2 h and at 400 °C for 4 h were used to remove the organic binder. Subsequently, the green body was hot pressed at 1800 °C for 1.5 h under a nitrogen atmosphere to give a laminated Si_3N_4. The microstructure of the laminated ceramics was examined using a SEM (CSM950, OPTON, Germany). Specimens were sliced into test bars (3 mm × 4 mm × 32 mm for bending strength and 4 mm × 6 mm × 32 mm for fracture toughness measurements). Flexural strength was measured with a span of 30 mm and a crosshead speed of 0.5 mm/minute at room temperature. The fracture toughness was determined by the single-edge-notch-beam method at room temperature with a crosshead speed of 0.05 mm/minute.

Results and Discussion

Microstructure

The microstructure of laminated Si_3N_4 ceramics is illustrated in Fig. 3. The scanning electron macrograph, Fig. 3(a) shows the laminates, and micrograph, Fig. 3(b) shows a cross section of a laminate. The average thickness of Si_3N_4 and BN sheets is 70 and 25 mm, respectively. Fig. 3(b) reveals the two dimensional distribution of elongated grains and SiC_w in Si_3N_4 sheets. Figure 3(b) shows the structure features resemblance with the nacre structure, such as sheet/interface arrangement and reinforcement within the sheets.

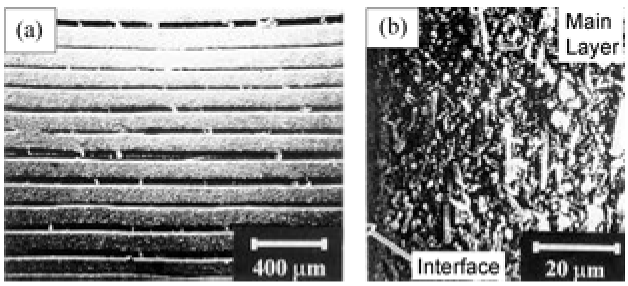

Fig. 3. SEM micrographs of macro-and micro-structures of a laminated Si_3N_4 ceramic. (a) macrostructure, (b) detailed structure of Si_3N_4 sheets.

Table 1. A comparison of the mechanical properties of different composites

Sample No.	Secondary reinforcement	Interface composition	Preparation method	Bending strength, (MPa)	Fracture toughness, ($MPa \cdot m^{1/2}$)
1	SiC whisker (15 wt%)	BN/Al_2O_3 (3:1)	Compact rolling	650±75	20±1
2	β-Si_3N_4 (3 wt%)	BN/Al_2O_3 (3:1)	Compact rolling	498±23	15±1
3	β-Si_3N_4 (3 wt%)	_	Cold press	750±72	11±1
4	SiC whisker (15 wt%)	_	Cold press	820±70	8±1

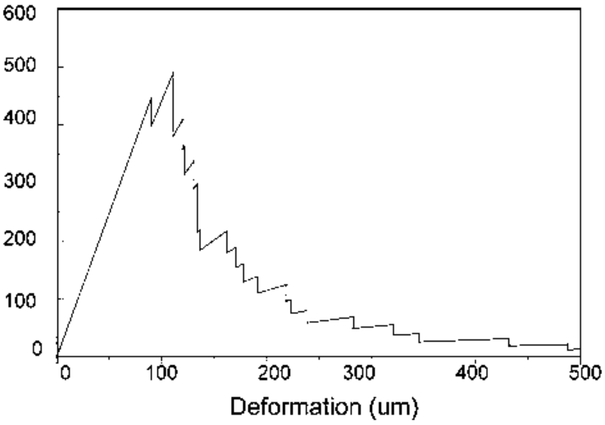

Fig. 4. Typical load-deformation curve of a laminated Si_3N_4 ceramic.

Mechanical Properties

The mechanical properties of laminated Si_3N_4 ceramics are summarized in Table 1. The introduction of a weak interfaces BN/Al_2O_3 improves the fracture toughness from 11 to 15 $MPa{\cdot}m^{1/2}$ in the case of β-Si_3N_4 reinforced samples. Whereas an increase from 8 to 20 $MPa{\cdot}m^{1/2}$ was observed in the case of SiC_w reinforced samples. The introduction of weak interfaces always degrades mechanical properties, however, whisker reinforcements restricts this effect, as observed in the present case also. The experimental data reveals that the use of additional reinforcement material, i.e. SiC_w not only improves the toughness to a greater extent but also preserves the bending strength. The reinforcement with SiC_w gives laminated ceramics a structure similar to nacre by toughening between and within laminates. This reinforcement is the factor that improves the toughness in laminated ceramics.

A typical load deflection curve for sample 1 is shown in Fig. 4. This indicates brittle fracture in a non-castrophic manner. This behavior is the result of two toughening mechanisms: weak interface and reinforcement.

Crack Propagating Behavior

Figure 5 shows a typical crack propagation in a laminated Si_3N_4 ceramic. As shown in Fig 5(a), a major crack propagates through the specimen under tension. The major crack progresses through a Si_3N_4 sheets to the next interface or sometimes it extends along the BN interface between the sheets.

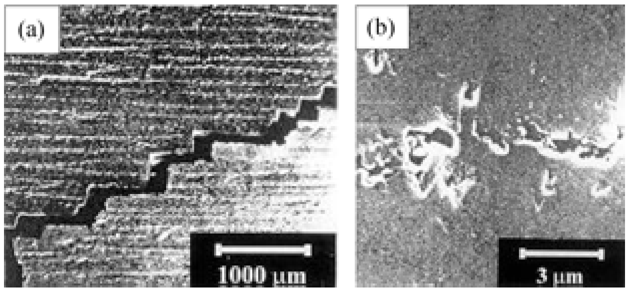

Fig. 5. SEM micrographs of crack propagation in a laminated ceramic. (a) major crack, (b) pull-out and debonding of whiskers inside sheets.

In SiC_w-reinforced samples, a pullout and debonding of SiC_w and elongated grains occur, as seen in Fig. 5(b). Therefore, the propagation and extension of the crack inside the sheets also take place so as to enhance the toughness and strength of the material. This phenomenon is similar to bulk Si_3N_4/SiCw mateial [1].

Conclusions

The present study concludes that a biomimetic of the nacre structure into laminated Si_3N_4 ceramics leads to an increase in fracture toughness and makes the material reliable for structural use.

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