Microstructural evolution of h-BN matrix composite ceramics with La–Al–Si–O glass phase during hot-pressed sintering

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Abstract: BN/La–Al–Si–O composite ceramics were fabricated by hot-pressed sintering using hexagonal boron nitride (h-BN), lanthanum oxide (La₂O₃), alumina (Al₂O₃), and amorphous silica (SiO₂) as the raw materials. The effects of sintering temperature on microstructural evolution, bulk density, apparent porosity, and mechanical properties of the h-BN composite ceramics were investigated. The results indicated that La–Al–Si–O liquid phase was formed during sintering process, which provided an environment for the growth of h-BN grains. With increasing sintering temperature, the cristobalite phase precipitation and h-BN grain growth occurred at the same time, which had a significant influence on the densification and mechanical properties of h-BN composite ceramics. The best mechanical properties of BN/La–Al–Si–O composite ceramics were obtained under the sintering temperature of 1700 °C. The elastic modulus, flexural strength, and fracture toughness were 80.5 GPa, 266.4 MPa, and 3.25 MPa·m^{1/2}, respectively.

Keywords: h-BN matrix composite ceramics; La–Al–Si–O glass phase; microstructural evolution; nanocrystalline precipitation; mechanical properties

1 Introduction

Hexagonal boron nitride (h-BN) and its matrix composite ceramics are typical structural-functional ceramics that

have been widely used in many fields, such as aerospace, machinery, metallurgy, energy, and electronics [1–7]. Compared with alumina, zirconia, silicon carbide, and silicon nitride ceramic, which have the high hardness and high strength, h-BN presents the relatively low hardness and good machinable properties because of its hexagonal layered crystal structure similar to that of graphite [8–11]. Furthermore, h-BN ceramics are difficult to sintering densification, so the low melting point sintering additives and/or second phase are usually

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added to improve the properties of h-BN composite ceramics [12–16].

Some researches have been investigated on the microstructural evolution during sintering and the properties of h-BN ceramics [17–21]. Zhang *et al.* [22] obtained h-BN powders composed of amorphous and nanocrystalline BN by ball milling, and then sintered them under different temperatures and pressures. Higher sintering pressure was more favorable to the preferred orientation growth of the in-plane direction of h-BN grains along the pressure direction, and higher sintering temperature promoted the mass transfer and grain growth. They referred that the structural fluctuation of amorphous BN resulted in the turbostratic boron nitride (t-BN) phase formation during the sintering process, and stacking faults usually existed in the as-grown h-BN grains [22].

For h-BN matrix composite ceramics, some research results have revealed the microstructure changing mechanisms [23–28]. Niu *et al.* [29] investigated the effects of raw h-BN particle size on the textured microstructures of magnesium aluminosilicate (BN–MAS) composite ceramics. With the increase of raw h-BN particle size, h-BN grains tended to orientate with the direction perpendicular to the hot-pressing direction, and the densification effect of MAS phase on BN–MAS composite ceramics decreased with increasing raw h-BN particle size due to the uneven dispersion of MAS phase [26,29]. Cai *et al.* [30–32] researched the influence of sintering process on the BN–MAS composite ceramics. The sintering pressure had a great influence not only on the mechanical properties of composite ceramics, but also on the crystallization of MAS and structural order of h-BN. The nucleophilic attack of N on M ($M = Mg^{2+}$, Al^{3+} , and Si^{4+}) and the electrophilic attack of B on O were the crucial factors on the formation of amorphous MAS phase. In addition, chemical bonding was formed between h-BN and MAS, and the matrix atoms diffused into the h-BN layers, leading to a strong bonding interface [30–32].

La–Al–Si–O glass phase has been reported on promoting sintering densification and improving the room/elevated-temperature mechanical properties of h-BN matrix composite ceramics [33–37]. However, the microstructural evolution during sintering process and its effect on properties of these material systems have not been fully revealed yet, which also has important implications for guiding the composition design and process optimization of composite ceramics.

In this study, BN/La–Al–Si–O composite ceramics were sintered under different temperatures from 1500 to 1900 °C. The phase composition, nanocrystalline precipitation, and grain growth were systematically investigated. The corresponding mechanical properties were tested to reveal the influence of microstructural evolution on the performance of composite ceramics.

2 Experimental

2.1 Material fabrication

Commercial powders of h-BN (99.5%, 0.3 µm, Advanced Technology & Materials Co., Ltd., China), hexagonal La_2O_3 (99.9%, 1.0 µm, Wuxi Meifang Industry Co., Ltd., China), rhombohedral Al_2O_3 (> 98%, 1.5 µm, Showa Denko K.K., Yokohama, Japan), and amorphous SiO_2 (99.9%, 3.5 µm, Lianyungang Guangyu Quartz Co., Ltd., China) were used as the raw materials. The volume ratio of h-BN:(La_2O_3 – Al_2O_3): SiO_2 was 70:10:20, and the mole ratio of La_2O_3 : Al_2O_3 was 1:2.

The weighed powders were mixed with Al_2O_3 balls and ethanol medium for 12 h. The obtained slurry was dried, and then passed through a 100-mesh sieve. The mixed powders were put into a graphite die and cold-compacted uniaxially under 5 MPa pressure. The obtained green compacts were hot-press sintered at different temperatures for 1 h under 20 MPa with N_2 atmosphere to maintain the partial pressure of nitrogen and prevent h-BN decomposition. The heating rate was $15\text{ }^{\circ}C \cdot min^{-1}$ and the samples cooled down to room temperature in the furnace spontaneously. According to the phase diagram of La–Al–Si–O system, liquid phase can be formed at about 1700 °C. Therefore, the sintering temperatures from 1500 to 1900 °C were chosen in this study.

2.2 Material characterization

Phase compositions were identified by X-ray diffractometer (XRD, D/max- γ B Cu $K\alpha$, Rigaku Co., Japan) with a scanning speed of $4\text{ }(^{\circ}) \cdot min^{-1}$. The detailed microstructures were investigated by transmission electron microscope (TEM, Talos F200X, FEI Co., USA), and the concentration of elements was detected by scanning electron microscope (SEM) equipped with energy dispersive spectroscopy (EDS, EDAX Inc., Mahwah, NJ, USA) detector. The TEM samples were firstly cut into the small pieces with the thickness of

about 0.2 mm, and then polished to about 0.07 mm by 2000-mesh abrasive paper. Ar ion etching instrument was used to create thin regions to meet the requirement of TEM observation. The bulk densities and apparent porosities of samples were measured by Archimedes law referring to the China National Standard GB/T 25995-2010. The flexural strength was measured by three-point bending method using a universal testing machine (Instron-5569, USA), and Young's modulus was obtained through the stress–strain curve. The sample size was 3 mm × 4 mm × 36 mm with a span of 30 mm and the crosshead speed was 0.5 mm·min^{−1}. The loading direction was perpendicular to the sintering pressure direction. Fracture toughness was measured using the single edge notched beam (SENB) method. The sample size was 2 mm × 4 mm × 20 mm with a notch of 2 mm, and the crosshead speed was 0.05 mm·min^{−1}. The fracture morphologies were observed using SEM (NanoLab 600i, FEI Co., USA).

3 Results and discussion

Figure 1(a) presents the XRD of BN/La–Al–Si–O composite ceramics sintered under different temperatures. There were only obvious diffraction peaks corresponding to h-BN phase (JCPDF 34-0421), whereas the diffraction peaks of La₂O₃, Al₂O₃, SiO₂, and their possible reaction products were not found. Considering the total volume content of the adding La–Al–Si–O was about 30%, which had exceeded the minimum threshold of XRD detection. Thus, we inferred that the amorphous glass phase was formed during hot-pressed sintering process, which was difficult to be characterized by XRD.

Comparing with the peaks of h-BN in different composite ceramics, with the increase of sintering temperature, the relative peak intensity of the corresponding (002) lattice plane increased gradually. Graphitizing index (*GI*) is an indicator of crystallization degree of graphite and similar crystalline structure materials [38], and it is calculated by the following equation:

$$GI = \frac{\text{Area}(100) + \text{Area}(101)}{\text{Area}(102)} \quad (1)$$

where Area(100), Area(101), and Area(102) denote the integral intensity intensities of the corresponding (*hkl*) reflexes of h-BN. Theoretically, the *GI* value of ideal h-BN crystal is about 1.6, and a lower *GI* value is corresponded to better crystallization of h-BN grains.

Figure 1(b) shows the calculated *GI* values of h-BN grains in composite ceramics. With the increase of sintering temperature, *GI* values showed a decreasing trend, which basically conformed to the change rule of exponential function. From 1500 to 1700 °C, the *GI* value decreased rapidly from 13.7 to 3.4; and from 1700 to 1900 °C, the *GI* value decreased slowly from 3.4 to 2.4. The sintering temperature had a significant influence on the crystallization growth of h-BN in composite ceramics. The higher sintering temperature was conducive to heat and mass transfer in liquid phase environment, and the better h-BN grains grow during hot-pressed sintering.

In Figs. 2(a)–2(c), TEM characterization was used to investigate the detail microstructures of BN/La–Al–Si–O composite ceramics hot-press sintered under 1500, 1700, and 1900 °C, and the corresponding element distributions of B, N, O, Al, Si, and La were shown in Fig. 2(d). The h-BN grains showed typical lamellar morphologies and were uniformly dispersed in all the

Fig. 1 (a) XRD patterns and (b) crystallization *GI* of BN/La–Al–Si–O composite ceramics.

Fig. 2 Microstructures of BN/La–Al–Si–O composite ceramics sintered under different temperatures: (a) 1500, (b) 1700, and (c) 1900 °C; (d) B, N, O, Al, Si, and La element distribution corresponding to the sample in (c) that sintered at 1900 °C.

composite ceramics. La–Al–Si–O glass phase filled in the space between h-BN grains and had a good combination with h-BN grains, and there were few obvious interfacial cracks. It could be obviously observed that the grain size of h-BN became bigger with increasing sintering temperature, because the liquid phase had better heat and mass transfer effect at higher temperatures to promote the growth of h-BN grains.

Some pores were observed in the sample sintered at 1500 °C. This was due to the relatively low fluidity of the liquid phase at this sintering temperature, which could not fully fill the gaps between the h-BN grains. However, in the sample sintered at 1900 °C, a small number of pores were also found. This was because the grown h-BN grains overlapped each other to form closed pores, which could not be filled by liquid phase. By contrast, no obvious pores were found in the sample sintered at 1700 °C, indicating this sintering temperature was favorable for obtaining composite ceramics with high relative densities.

Interface microstructures between h-BN grains and La–Al–Si–O phase of composite ceramics sintered under different temperatures are presented in Figs. 3(a)–3(c), and the corresponding elemental line scanning profiles from h-BN zone to La–Al–Si–O zone are shown in Figs. 3(d)–3(f). No defects such as crack could be

observed at the phase boundary, indicating a good wettability between La–Al–Si–O glass phase and h-BN grains. The changes of elemental contents were continuous. La, Al, Si, and O content of glass phase increased, whereas B and N content of h-BN phase decreased gradually along the arrow direction. When the sintering temperature increased from 1500 to 1900 °C, the width of the measuring diffusion zone of three samples (h-BN grains and La–Al–Si–O glass phase of composite ceramics sintered under 1500, 1700, and 1900 °C) at the two phase interface increased from about 38 to more than 55 nm, indicating higher sintering temperatures were more beneficial to the element diffusion in the phase interface region during hot-pressed sintering process. In Fig. 3(g), high-resolution transmission electron microscopy (HRTEM) results exhibited the detailed interface zone formed by atom diffusion between La–Al–Si–O glass phase and the h-BN phase, which showed a gradual transition from order to disorder arrangement. On the whole, continuous, defect-free, and mutually diffusive grain boundary was beneficial to provide the good interface bonding and better performance of composite ceramics.

As shown in Figs. 4(a)–4(c), the precipitation nanocrystalline was also found in La–Al–Si–O glass phase, and with the increase of sintering temperature, the size of these precipitated grains showed a gradual increasing trend. High sintering temperature was more likely to form precipitation phase with bigger size. Through selecting electron diffraction analysis in Fig. 4(d), the precipitated phase was identified as cristobalite, which meant the precipitated cristobalite phase and La–Al–Si–O glass phase were coexisted in the composite ceramics. This phenomenon was also reported in the similar multiple oxide systems, such as $\text{CaO-Al}_2\text{O}_3\text{-SiO}_2$, $\text{La}_2\text{O}_3\text{-Al}_2\text{O}_3\text{-SiO}_2$, and $\text{Y}_2\text{O}_3\text{-Al}_2\text{O}_3\text{-SiO}_2$. Inhomogeneous nucleation in glass phase is the main reason for the formation of cristobalite nanocrystals [39,40].

Figure 5 shows the bulk densities and apparent porosities of BN/La–Al–Si–O composite ceramics sintered under different temperatures. With the increase of sintering temperature, bulk density of h-BN composite ceramics first increased and then decreased, whereas apparent porosity exhibited the opposite tendency. The composite ceramic sintered at 1700 °C had the highest bulk density and the lowest apparent porosity. With the increase of sintering temperature, the liquid phase had better fluidity and wettability, and could well fill into the voids formed by the overlap of h-BN

Fig. 3 Interface microstructures between h-BN grains and La-Al-Si-O glass phase of composite ceramics sintered under different temperatures: (a) 1500, (b) 1700, and (c) 1900 °C; (d–f) elemental line scan profiles corresponding to lines A, B, and C in (a–c), respectively; (g) HRTEM corresponding to the interface of La-Al-Si-O phase and h-BN grain in (c).

Fig. 4 Nanocrystalline precipitation microstructures in La-Al-Si-O glass phase of composite ceramics sintered under different temperatures: (a) 1500, (b) 1700, and (c) 1900 °C; (d) diffraction patterns of amorphous phase and cristobalite nanocrystalline.

grains, which contributed to the improvement of densification. However, with the further increase of sintering temperature, h-BN grains had obvious growth,

which led to the larger pores in the mutual framework by the large h-BN grains, resulting in the decrease of bulk density. Furthermore, some liquid phase was

Fig. 5 Bulk densities and apparent porosities of BN/La-Al-Si-O composite ceramics.

extruded during hot-pressed sintering at 1900 °C because of the good fluidity of La-Al-Si-O glass phase at high temperatures. Therefore, the glass phase content in the composition of this sample is less than that of other samples, leading to lower density and higher porosity.

Figure 6 shows mechanical properties of BN/La-Al-Si-O composite ceramics sintered under different temperatures, including flexural strength, elastic modulus, and fracture toughness. With the increase of sintering temperature, the mechanical properties presented a small increase followed by a rapid decrease, which

Fig. 6 Mechanical properties of BN/La-Al-Si-O composite ceramics.

was consistent with the tendency of bulk density. The best mechanical properties of BN/La-Al-Si-O composite ceramics were obtained under the sintering temperature of 1700 °C. The elastic modulus, flexural strength, and fracture toughness were 80.5 ± 0.7 GPa, 266.4 ± 10.1 MPa, and 3.25 ± 0.05 MPa·m^{1/2}, respectively.

Fracture morphologies of BN/La-Al-Si-O composite ceramics sintered under different temperatures are shown in Figs. 7(a)–7(e). Digital Micrograph software was used to measure the grain size of h-BN at the representative areas, and the average grain size was calculated from about 30 measured h-BN grains. The grain size increased significantly with increasing sintering temperature, and the statistically average

Fig. 7 Fracture morphologies of BN/La-Al-Si-O composite ceramics sintered under different temperatures: (a) 1500, (b) 1600, (c) 1700, (d) 1800, and (e) 1900 °C; (f) statistics of average grain size.

values are listed in Fig. 7(f). As the sintering temperature changed from 1500 to 1900 °C, the average size of h-BN grains increased from 0.35 to 2.5 µm.

From the above results, we comprehensively analyzed the influence of sintering temperature on the mechanical properties of BN/La–Al–Si–O composite ceramics, which mainly included the following two points: (1) High sintering temperature facilitated heat transfer and atom diffusion in liquid phase, which were beneficial to liquid phase pore filling to increase the relative density and improve the mechanical properties; (2) the grain sizes of h-BN increased rapidly with the increase of sintering temperature, and when h-BN grains grew to larger size, the porosity of composite ceramics became higher, resulting in an adverse effect on the densifying process and mechanical properties.

The microstructural evolution process of BN/La–Al–Si–O composite ceramics during hot-pressed sintering can be illustrated in Fig. 8. Firstly, the four raw powders were uniformly mixed and heated gradually in the graphite mold (Fig. 8(a)). When the sintering temperature increased, the La–Al–Si–O liquid phase was formed and h-BN grains were uniformly distributed in the liquid phase environment (Fig. 8(b)). With the further increase of sintering temperature, the heat and mass transfer ability of the liquid phase was enhanced, and the h-BN grains began to grow significantly

(Fig. 8(c)). At the case of sintering temperature increasing or holding time extending, the grain size of h-BN further grew. At the same time, the cristobalite phase nanocrystalline was also precipitated in the liquid phase (Fig. 8(d)). These microstructural evolution mechanisms of BN/La–Al–Si–O composite ceramics are consistent with Figs. 2–4.

4 Conclusions

The BN/La–Al–Si–O composite ceramics were hot-press sintered under different temperatures to reveal the microstructural evolution mechanisms. Ternary La–Al–Si–O liquid phase was formed during the sintering process, which had a good wettability with h-BN grains and could effectively fill the pores to improve the densification of composite ceramics. Higher sintering temperature contributed to the growth and crystallization of h-BN grains through better heat transfer and atomic diffusion in liquid phase environment. Furthermore, cristobalite nanocrystals were precipitated from the liquid phase and also grow gradually with the increase of sintering temperature. The BN/La–Al–Si–O composite ceramics sintered under 1700 °C exhibited the best mechanical properties, which were attributed to the mutual influence of liquid phase environment, h-BN grain size, and precipitated phase.

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Fig. 8 Microstructural evolution mechanism diagrams of BN/La–Al–Si–O composite ceramics: (a) mixed raw powders, (b) La–Al–Si–O liquid phase formed, (c) h-BN grain growth, and (d) cristobalite nanocrystalline precipitation.

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