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Effect of time on microstructure and hardness of βSiAlON–cubic boron nitride composites during spark plasma sintering

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Abstract

βSiAlON–cubic boron nitride (cBN) composites were consolidated by spark plasma sintering, and the effects of holding time and heating rate on the phase transformation of cBN and Vickers hardness were investigated. The cBN phase transformed into hexagonal BN (hBN) and the hardness decreased with increasing holding time. The phase transformation from cBN to hBN was retarded by increasing the heating rate, resulting in increased hardness.

Keywords: A. Spark plasma sintering; C. Hardness; D. Boron nitride; D. SiAlON

1. Introduction

Cubic boron nitride (cBN) has high hardness and thermal conductivity, and is characterized by greater thermal stability and lower reactivity with iron than diamond. Therefore, cBN has been employed in cutting tools, particularly for high-speed machining of hardened steel and cast iron [1]. Because of the low-sinterability of cBN due to its strong covalent nature and low self-diffusion coefficients of B and N [2–4], cBN bodies have been sintered at an ultra-high pressure of more than 5 GPa with various kinds of additives [5–8]. Since ultra-high pressure sintering is costly and employed only for small size products, a moderate pressure sintering of less than 100 MPa is advantageous for expansion of the application of cBN [9].

SiAlON has also been applied to cutting tools due to its high hardness and good oxidation resistance. Thus, a composite material consisting of cBN and SiAlON is a candidate material for cutting tools due to its high hardness, fracture toughness and good oxidation resistance. However, dense cBN containing ceramic composites are difficult to produce by conventional sintering techniques [10–12].

We have previously reported the preparation of cBN-based composites under a moderate pressure of 100 MPa by spark plasma sintering (SPS). Al_2O_3 and TiN retarded the densification of cBN-based composites [10,13], whereas βSiAlON accelerated the densification of the composites [14]. The phase transformation from cBN to hBN in the βSiAlON–cBN composite was suppressed as compared with a monolithic cBN body, and Al_2O_3–cBN and TiN–cBN composites. Since the phase transformation from cBN to hBN causes the degradation of density and hardness of the composites, the parameter of time during the SPS process, i.e., holding time and heating rate, is crucial for preparation of a dense β SiAlON–cBN composite.

In the present work, βSiAlON–cBN composites were sintered by changing holding times and heating rates at a pressure of 100 MPa by SPS, and their phase transformation, densification and hardness were studied.

2. Experimental procedure

βSiAlON ($Si_3Al_3O_3N_5$, Ube Industries, average particle size: 1.4 μm) and cBN (SBN-F, Showa Denko, 2.8 μm) powders were used. 80 vol% βSiAlON and 20 vol% cBN were ball-milled for 48 h and sintered at 1650 and 1700 °C up to 1800 s at heating rates of 0.3–3.7 °C/s at 100 MPa in a vacuum using an SPS apparatus (SPS-210LX, SPS Syntex). The surface

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temperature of the graphite die was measured by an optical pyrometer. The phase transformation of cBN was studied by X-ray diffractometry (XRD; Geigerflex, Rigaku). The cBN content after sintering was determined by XRD, where (1 1 1) cBN, (0 0 2) hBN and (0 2 0) βSiAlON peaks (the strongest XRD peaks of each phase) were chosen to quantify the cBN content [15]. The density was calculated by an Archimedes method. The specimen surface was observed by scanning electron microscopy (SEM; S-3100H, Hitachi) after polishing with 1 μm diamond slurry. Vickers hardness was measured with a micro-hardness tester (HM-221, Mitutoyo) at an applied load of 0.98 N.

3. Results and discussion

Fig. 1 shows the effect of holding time on the relative density of the βSiAlON–BN (20 vol% cBN) sintered at 1650 and 1700 °C. The density with sintering at 1650 °C without holding time was 98%. The density increased further and reached nearly 100% at 600 s and then slightly decreased at 1800 s. At 1700 °C, the βSiAlON–BN composite sintered without holding time was close to full density, and then decreased with increasing holding time.

Fig. 2 shows the effect of holding time on the cBN content in the βSiAlON–BN composite (20 vol% cBN) sintered at 1650 and 1700 °C. At 1650 °C, the cBN content in the BN phase was 89% for holding time until 300 s, and then decreased to 71% at 1800 s. At 1700 °C, the cBN content at holding time up to 60 s was 82%, and decreased to 29% at 1800 s.

Fig. 3(a) and (b) show the SEM micrographs of the surface of the βSiAlON–BN composite (20 vol% BN) sintered at 1650 and 1700?C for 600 s, respectively. The dark phase is cBN grains. Angular-shaped cBN grains remained in the βSiAlON matrix at 1650 °C, whereas many pores and cracks were observed at the βSiAlON/BN interface at 1700 °C. These cracks were caused by the volume change due to the phase transformation of cBN to hBN [14]. This corresponds to the decrease of the density with the holding time as depicted in Figs. 1 and 2.

Fig. 4 demonstrates the effect of the holding time on the Vickers hardness of the βSiAlON–BN composite (20 vol% BN)

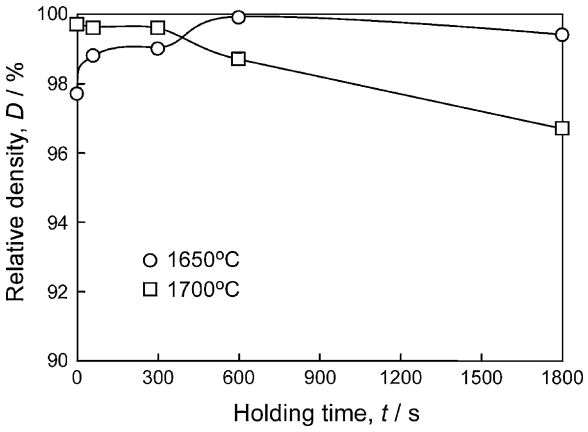

Fig. 1. Effect of holding time on the relative density of βSiAlON–BN composite (20 vol% cBN) at 1650 and 1700 °C for 0–1800 s.

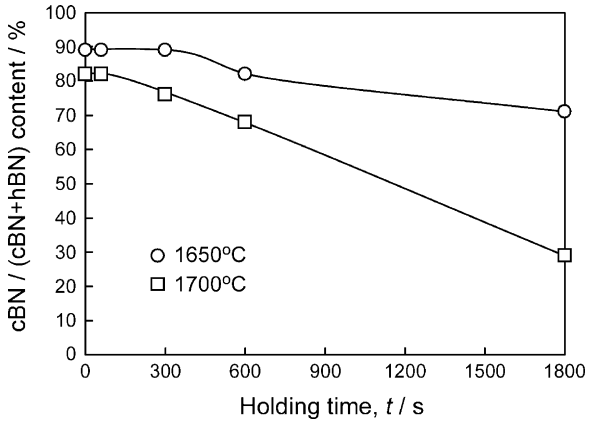

Fig. 2. Effect of holding time on the cBN content in βSiAlON–BN composite (20 vol% cBN) at 1650 and 1700 °C for 0–1800 s.

sintered at 1650 and 1700 °C. The hardness of the βSiAlON–BN composite sintered at 1650 °C was higher than that sintered at 1700 °C. Since the phase transformation of cBN at 1650 °C was

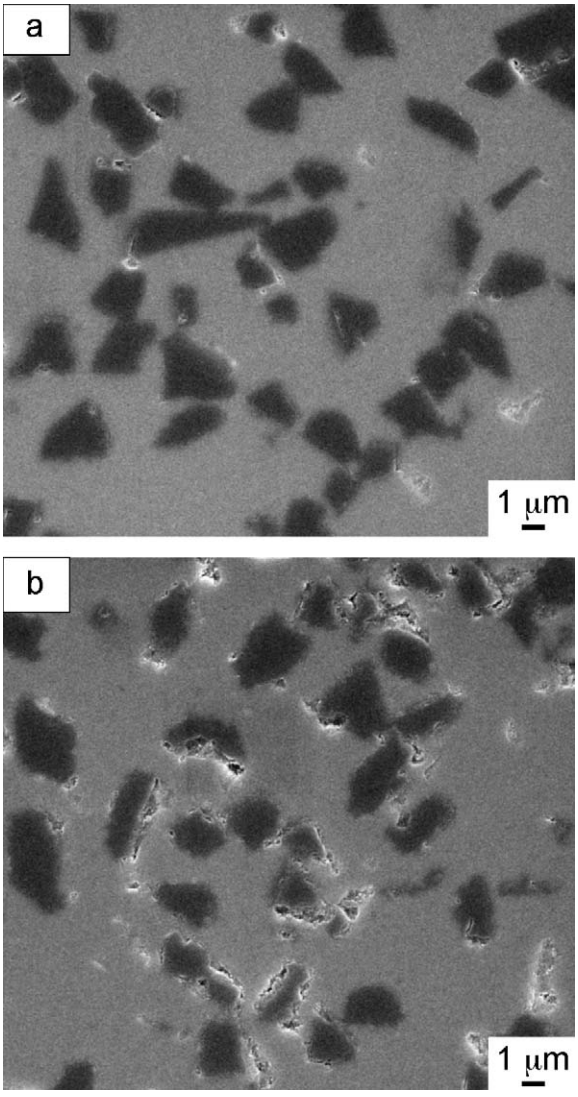

Fig. 3. SEM micrographs of the polished surface of βSiAlON–BN composite (20 vol% cBN) at 1650 °C (a) and 1700 °C (b) for 600 s.

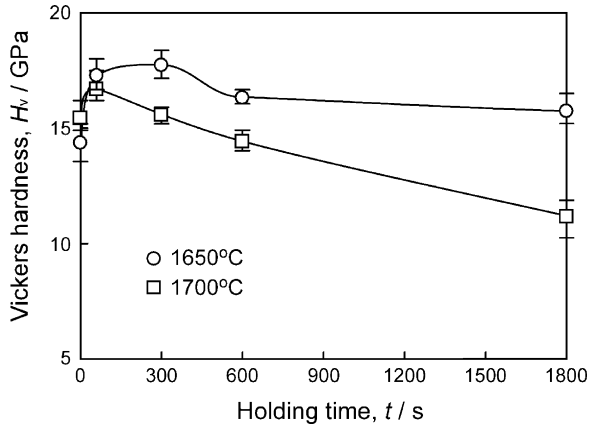

Fig. 4. Effect of holding time on the Vickers hardness of βSiAlON–BN composite (20 vol% cBN) at 1650 and 1700 °C.

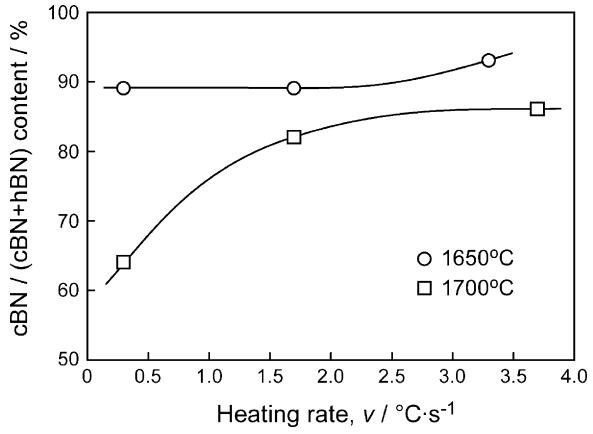

Fig. 5. Effect of heating rate on the cBN content in βSiAlON–BN composite (20 vol% cBN) at 1650 °C for 60 s and 1700 °C without holding time.

insignificant, higher hardness could be yielded by less transformation. At 1650 °C, the hardness without holding time was 14.4 GPa, while it was 17.7 GPa at 300 s. This could have resulted from the increase in the density, where the cBN content of 89% did not change during the holding time from 0 to 60 s, as shown in Fig. 2. The decrease of hardness over 300 s could have resulted from the phase transformation of cBN.

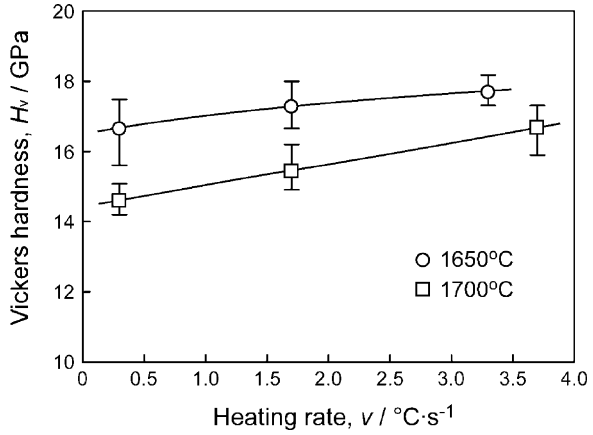

Fig. 6. Effect of heating rate on the Vickers hardness of βSiAlON–BN composite (20 vol% cBN) at 1650 °C for 60 s and 1700 °C without holding time.

Fig. 5 shows the effect of the heating rate on the cBN content in the βSiAlON–BN composite (20 vol% BN) sintered at 1650 °C for 60 s and at 1700 °C without holding time. The cBN content in the βSiAlON–BN composite increased with increasing heating rate, particularly at 1700 °C.

Fig. 6 shows the effect of the heating rate on the Vickers hardness of the βSiAlON–BN composite (20 vol% BN) sintered at 1650 °C for 60 s and at 1700 °C without holding time. The hardness at 1650 °C increased from 16.6 to 17.7 GPa with increasing heating rate from 0.3 to 3.3 °C/s. This could also have been caused by the lesser phase transformation of cBN to hBN.

4. Conclusions

The effects of holding time and heating rate during SPS on the phase transformation and Vickers hardness of βSiAlON–cBN composites were studied. The cBN content in the BN phase was 89% at 1650 °C below 300 s and decreased to 71% at 1800 s. The hardness of the βSiAlON–cBN composite sintered at 1650 °C for 300 s exhibited the maximum value of 17.7 GPa. The hardness decreased to 15.7 GPa at 1800 s. The cBN content increased from 89 to 93% and the hardness increased from 16.6 to 17.7 GPa with increasing heating rate from 0.3 to 3.3 °C/s at 1650 °C.

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