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Investigation on Addition of Talc on Sintering Behavior and Mechanical Properties of B_4C

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The effect of high alumina talc powder as a sintering aid on densification of boron carbide powder was investigated. The in situ reaction of talc with boron carbide generate SiC, MgB_2, and alumina, which all aid the sintering process and permit pressureless sintering at temperatures between 2050 and 2150 °C. The microstructure-property relationship has been studied in the B_4C-talc system for talc content between 0 and 30 wt.%. It became clear the addition of talc powder has a significant effect on the sintering behavior of B_4C. B_4C composites along with talc powder additions were sintered up to 98% of theoretical density. The mechanical properties were improved over those of samples without talc addition.

Keywords alumina, boron carbide, ceramic, magnesium diboride, silicon carbide, sintering, talc

1. Introduction

Among the outstanding physical and mechanical properties of boron carbide is its hardness, which stands after those of diamond and C-BN (Ref 1-26). This specific property comes along with other attractive properties such as high impact and wears resistance, low density, high melting point, and excellent resistance to chemical agents, as well as high capability for neutron absorption. However, its extreme sensitivity to brittle fracture and the difficulties involved in fabricating dense B_4C materials have limited its use in industrial applications (Ref 1-12). Because pure B_4C is very difficult to sinter with higher than approximately 80% theoretical density, a variety of second phases have been added as sintering aids (Ref 1-8). Nonoxide ceramics such as SiC (Ref 1-3), TiC (Ref 4), C (Ref 5-7), and TiB_2 (Ref 14-22) have also been found to be very effective as sintering additives for B_4C. Metallic sintering aids such as Al (Ref 8), Si (Ref 12), Ti (Ref 13), Mg, and Fe are frequently added to provide a medium for liquid-phase sintering. Metallic phases at the grain boundaries generally deteriorate the unique properties of hard ceramics. However, in these cases, either large amounts of second phase or very high sintering temperatures are required for full densification (Ref 1-18). It has been frequently observed that small amounts of oxides such as Al_2O_3 (Ref 20, 21), TiO_2 (Ref 19, 22), and Cr_2O_3 (Ref 23) are very effective in improving the sinterability of B_4C. These powders must be very fine and have high purity to improve sintering. Thus, they are generally very expensive.

In the current study, the effect of talc addition on the densification behavior of B_4C has been investigated. Mechanical properties, such as hardness and fracture toughness of B_4C have been measured and correlated with the variations in density and composition of the body.

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2. Experimental Procedure

High-purity B_4C (B:C ratio of 3.8-3.9) and high-alumina talc powders (26.62%Al_2O_3:47.78%SiO_2:25.6%MgO) were used as starting materials. The average size and the specific surface area measured [by the Brunauer-Emmett-Teller (BET) method] of the B_4C powder were 1.33 μm and 6.64 m^2/g, respectively. Up to 30 wt.% of talc was added as the sintering aid. The powders were ball milled in isopropyl alcohol for 8 h using high-purity Al_2O_3 balls. The mixture was then dried in a rotary vacuum evaporator and passed through a 60 mesh screen. The powder mixtures were cold pressed under 80 MPa into samples 30 × 3 × 60 mm in size. The green samples were then sintered using a microprocessor controlled graphite element furnace. The heating and cooling rates were 10 °C/min and furnace cooling, respectively.

For microstructural examinations, dense sintered bodies were surface ground and polished with diamond paste down to 1 μm surface finish. The polished surfaces were then electrically etched in a 0.1% KOH solution with a current density of $0.1A/cm^2$ for 10-20 s. Microstructures of the specimens were observed using scanning electron microscopy (SEM), and the phases were characterized by the x-ray diffraction (XRD) method. The density was measured by Archimedes method. The aluminum, magnesium, and silicon concentrations in the fired specimens and some green compacts were determined using wet chemistry and atomic adsorption. An approximate theoretical density was calculated for the various quaternary compositions of the B_4C-Al_2O_3-SiC-MgB_2 system. This approximation was based on the measured Al, Mg, and Si concentrations from which the volumetric percentage of phases was derived. Based on this method, the composition of sintered samples at 2050 °C for 1 h was determined (Table 1). The same method has already been used by other researchers for such systems as B_4C-ZrO_2 (Ref 12), B_4C-TiC (Ref 4), and B_4C-TiO_2 (Ref 19).

For mechanical testing, samples were cut to dimensions of

3 × 4 × 45 mm and ground with a 800 grit diamond grinding wheel. The tensile side of the specimens was polished with diamond paste down to 1 μm finish. To measure the hardness, a Vickers indentor was used with a load of 1.96 N. The flexural strength was measured by the four-point flexural test method using a universal testing machine with a crosshead speed of 0.5 mm/min. The inner and outer spans of the jig were 20 and 40 mm, respectively. The fracture toughness of the specimens was determined by the indentation strength method. After indenting the polished surface at 98 N with a Vickers indentor for 15 s, the fracture strength was measured with the four-point flexural configuration.

3. Results and Discussion

Extensive reaction occurred between the B_4C and the talc powder when they were sintered at 2050 °C for 1 h. Formation of MgB_2, SiC, and Al_2O_3 during sintering is clearly shown in XRD patterns in Fig. 1. Before sintering, only B_4C, alumina, and talc peaks are present (Fig. 1). After sintering, however, MgB_2, SiC, and alumina peaks were detected, and all the talc peaks had disappeared, indicating there was a reaction between the B_4C and talc to form these phases.

Microstructure and sintering behavior of boron carbide remarkably improve by the addition of talc powder. Figures 2 and 3 show SEM images indicating the influence of talc on densification of boron carbide.

The figures show that boron carbide-free talc sintered at 2050 and 2150 °C is not fully densified. The addition of 30 wt.% talc to the starting powder results in higher density. This is due to the fact that at sintering temperature, the SiO_2 and MgO content of talc converts to SiC and MgB_2, but Al_2O_3 remains at grain boundaries. Formation of these phases is detected by XRD analysis.

Table 1 Phase composition after sintering at 2050 °C

	Composition, wt.%		
Phase	**10% talc**	**20% talc**	**30% talc**
Al_2O_3	2.81	5.6	9.64
SiC	3.4	7.2	11.55
MgB_2	3.1	6.56	10.5

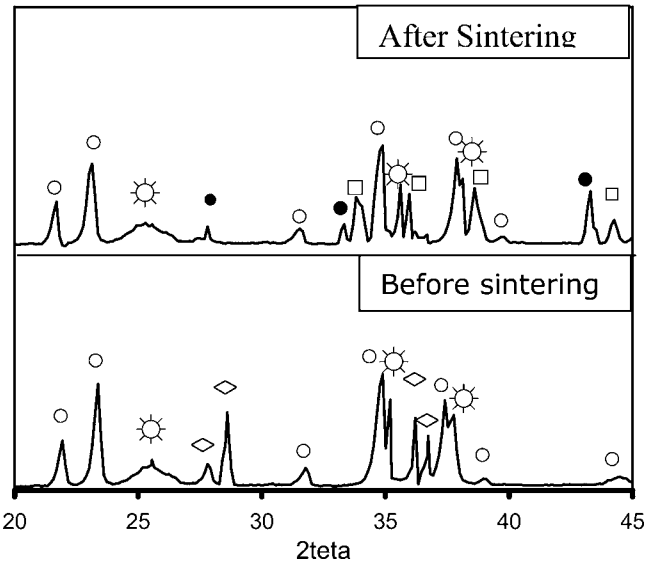
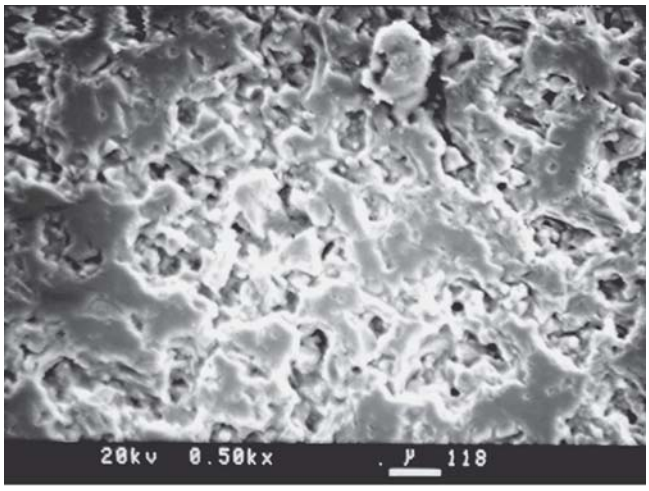


Fig. 1 X-ray patterns of B_4C with 10 vol% talc before and after sintering at 2050 °C for 1 h (○) B_4C, (◊) Talc, (●) MgB_2, (□)SiC, and (☼) Al_2O_3

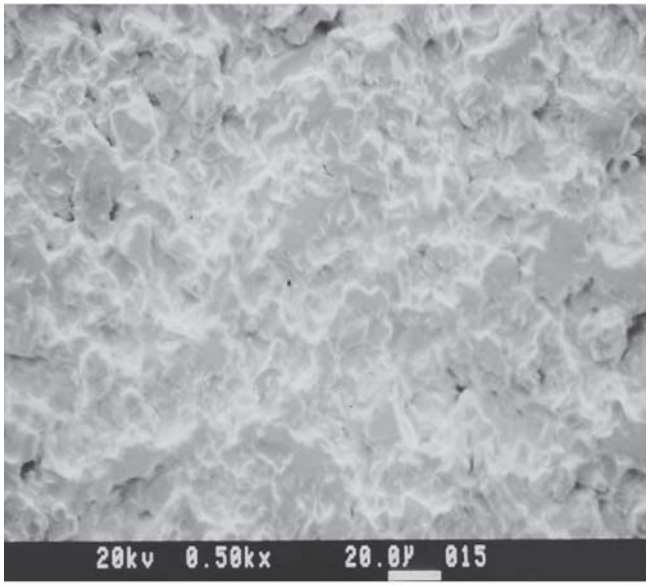
At temperatures lower than 2000 °C, anstatite liquid phase forms from the decomposition of talc at 900 °C. At sintering temperature, this liquid phase reacts with B_4C to form SiC, MgB_2, and Al_2O_3. Therefore, reactive liquid phase sintering occurs in this system. The effect of the presence of new phases on sintering behavior of B_4C has been studied by a number of researchers (Ref 1, 10, 13, 21, 24, 25). The primary effect is a considerable increase in relative density, which is caused by enhancement of mass transport within these phases or reaction products at the grain boundaries. This improves such mechanical properties as hardness and fracture toughness (Ref 21).

Limited studies have been done by Kim et al. (Ref 21) about the effect of Al_2O_3 on the sintering behavior of B_4C.

It has been reported previously that a chemical reaction occurs between B_4C and Al_2O_3 to form $AlB_{12}C_2$ at 2150 °C.



(a)



(b)

Fig. 2 SEM micrographs of samples sintered at 2050 °C for 1 h: (a) B_4C-free talc and (b) B_4C-30% talc

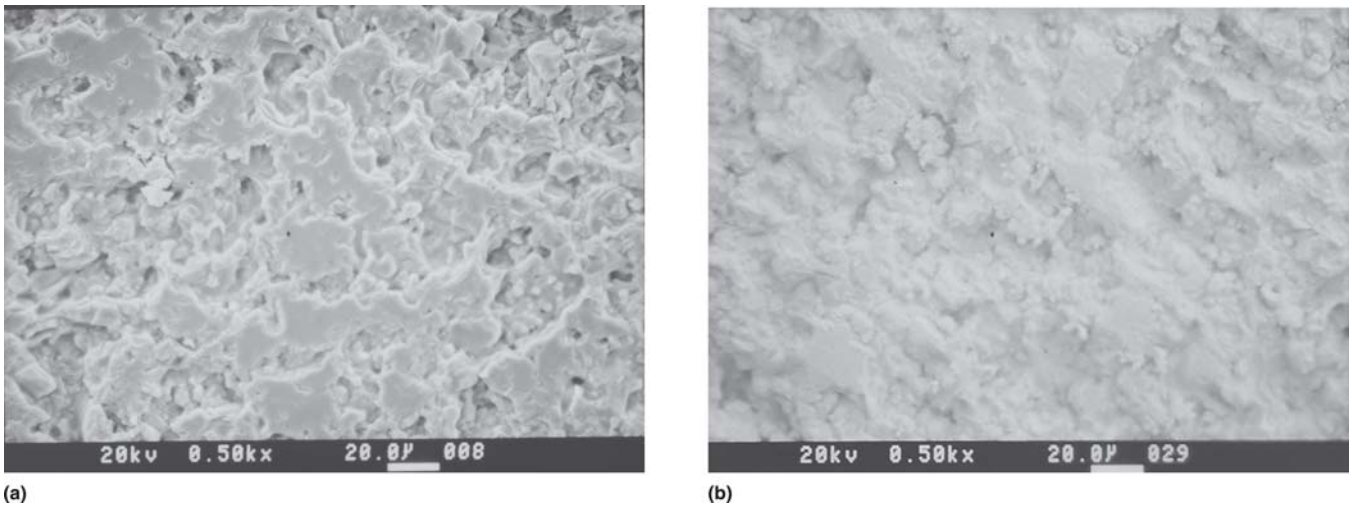


Fig. 3 SEM micrographs of samples sintered at 2150 °C for 1 h: (a) B_4C-free talc and (b) B_4C-30% talc

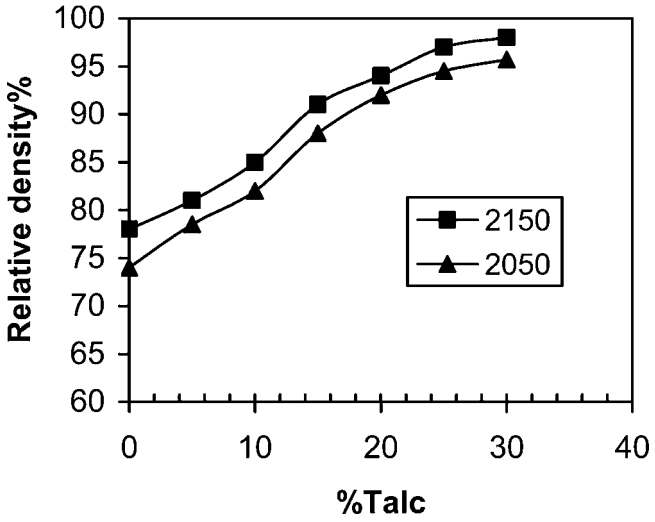


Fig. 4 Effect of talc addition on relative density of the samples sintered at (■) 2150 °C and (▲) 2050 °C

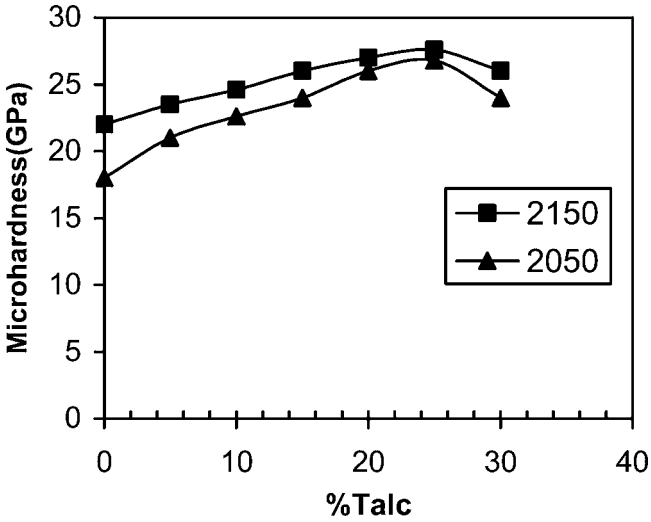


Fig. 5 Effect of talc addition on Vickers microhardness of the samples sintered at (■) 2150 °C and (▲) 2050 °C

$AlB_{12}C_2$ is known to enhance the densification of B_4C by lowering the diffusion barrier (Ref 21, 22). In this study, formation of this phase was not detected by XRD analysis.

The effect of talc addition on relative density of the samples sintered at 2050 and 2150 °C as a function of talc percentage is shown in Fig. 4. As can be seen from the figure, the density increases by increasing talc in starting powder. The addition of 30 wt.% talc increased the relative density from 78 to 98%. The increasing density will improve the mechanical properties as well. In Fig. 5, the effect of talc on microhardness of the samples with 0-30 wt.% talc content has been shown. From the figures, it can be seen that the microhardness increases with increased talc content. The maximum hardness is obtained for the sample with 25 wt.% talc in its composition. This is due to the presence of fewer pores in the samples. The reduction of hardness in samples having more than 25 wt.% talc is due to the presence of fewer hard phases, compared with B_4C, as reaction products in the samples. Therefore, the increase of talc content will result in lowering the hardness. On the other hand, the thermal expansion mismatch between these phases is another cause for reduction of hardness (Ref 4, 21).

Moreover, because of higher density, the hardness of the samples sintered at 2150 °C is higher than that of the samples sintered at 2050 °C.

The lower hardness of the samples is caused by the presence of fewer hard phases in the matrix. Figure 6 illustrates the effect of talc addition on toughness of the samples. From the figure, it is evident that by increasing the talc up to 30 wt.%, the toughness of the samples increases, which is due to the higher density and presence of fewer hard phases. As can be seen, the toughness of the samples sintered at 2150 °C is higher than that of samples sintered at 2050 °C. The effect of density on the improvement of mechanical properties is related to the amount of porosity and its role on such properties.

Therefore, decreasing the porosity will enhance the mechanical properties, except hardness. The lower hardness of the samples with a greater amount of talc is related to the greater amount of Al_2O_3 and MgB_2 in such samples. The coefficient of thermal expansion of alumina is about 4.8×10^{-6}/°C, and that of B_4C, SiC, and MgB_2 is 1.7×10^{-6}/°C, 2.4×10^{-6}/°C, and 14×10^{-6}/°C, respectively. According to reports of Sigl (Ref 4),

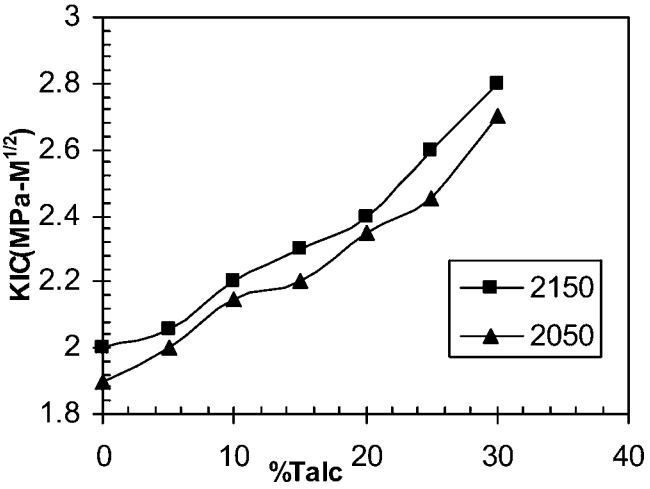


Fig. 6 Effect of talc addition on fracture toughness of the samples sintered at (■) 2150 °C and (▲) 2050 °C

the expansion mismatch between constituent will cause microcracks resulting in lowering the hardness. The same phenomenon has been reported by Kim (Ref 21) for B_4C samples having 5 wt.% alumina.

The improved toughness can be related to the reduction of porosity, which is the main source of crack initiation. Moreover, through the addition of alumina, some residual stresses exist in the samples that enhance toughness by crack deflection and microcracking mechanism (Ref 4, 20, 21). It seems that in the current study, the same phenomena control the mechanical properties.

4. Conclusions

- The addition of talc increases the sample density.
- The mechanical properties, except hardness, are improved by the addition of talc up to 30 wt.% to starting B_4C powder. Hardness first increases for samples with up to 25 wt.% talc and then decreases for samples having a higher amount of talc.
- The $AlB_{12}C_2$ composition that has been reported by other researchers in B_4C samples containing Al_2O_3 was not detected in the current study.
- It is hypothesized that the toughness improvement is caused by the presence of less-hard phases in the matrix, high density, and the presence of residual stresses around Al_2O_3, MgB_2, and SiC grains.

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