Effect of Boron Carbide Additive on SiC Ceramic Sintered by Solid-Phase Method under Ar Atmosphere

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Abstract: With boron carbide (B₄C) as sintering additives, silicon carbide (SiC) ceramics were prepared in 2100°C by solid-phase sintering under argon atmosphere. The change of microstructure, phase composition and density for all sintered samples were studied. The results showed that with the increase of B₄C addition, the density of SiC ceramic was significantly increasing between 0-4 wt% of B₄C addition and slightly increasing between 4-8 wt%. XRD analysis showed that all the ceramics without B₄C were SiC-6H crystal, while those with B₄C contained SiC-4H crystal. B₄C turned SiC-6H to SiC-4H, and the more B₄C added, the more change it made. SiC ceramic particles changed from equiaxial shape without B₄C to flake shape with B₄C added, and the particle sizes increased significantly with the amount of B₄C additive.

1. Introduction

SiC ceramic is characterized by high strength, high hardness, reliable chemical stability, good thermal-impact resistance, and creep resistance. It has been widely used in defense, nuclear energy and space technology, automotive industry, and marine engineering. Therefore, it has become one of the most promising structural ceramics [1]. And due to the high ratio of SiC Covalent bonds and the small self-diffusion coefficient, Silicon carbide is difficult to sinter [2]. Therefore, it is often necessary to add sintering aids to promote the sintering of SiC ceramics. The sintering aid commonly used in solid phase sintering is the Al-B-C-B₄C system. SiC ceramic prepared by the solid-phase method sintering not only has a low content of sintering aids, but also does not have a low melting point at the crystal boundary, which can make the SiC ceramic materials have the advantages of good high temperature stability [3, 4], but the densification of pressureless sintering is less than that of hot pressed sintering [5,6]. Therefore, it is necessary to study the effect of B₄C as a kind of effective sintering additive on the densification of solid-phase sintered SiC ceramics.

In this study, SiC ceramic tablets were prepared at 2100°C using coarse SiC powder and fine SiC powder as the original powder, B₄C as sintering additive. The effects of variation of B₄C on the densification of SiC ceramic were studied.
2. Experimental

2.1. Raw material

Coarse SiC powder, (D50 = 8.7 μm), fine SiC powder (D50 = 1.5 μm), B4C powder (D50 = 4.3 μm), polyethylene pyrrolidone (PVP), etc.

| Table 1. Formulation of SiC ceramic with variation of B4C additive. |
|-------------------------|--------|--------|--------|--------|--------|
|                         | a      | b      | c      | d      | e      |
| Coarse SiC powder (wt%) | 59.4   | 58.2   | 57     | 55.8   | 54.6   |
| Fine SiC powder (wt%)   | 39.6   | 38.8   | 38     | 37.2   | 36.4   |
| B4C (wt%)               | 0      | 2      | 4      | 6      | 8      |
| PVP (wt%)               | 1      | 1      | 1      | 1      | 1      |

2.2. Preparation of SiC ceramic

Coarse SiC powder (D50 = 8.7 μm), fine SiC powder (D50 = 1.5 μm), B4C powder, PVP powder with the relative weight ratios were added into the distilled water, respectively. After 4hour’s ball milling, the slurries were dried to powder at a constant temperature (about 105˚C). After sifted out from 0.38mm sieve, they were tableted with pressure of 120 Mpa. After the moulding, the blank bodies was gradually heated up to 2100˚C in a SiC sintering furnace and furnace-cooled after heat preserved for 1h to obtain sintered SiC ceramics.

2.3. Characterization

The density of Silicon carbide ceramics was measured by Archimedes drainage method. The crystal phase changes before and after the addition of B4C to Silicon carbide ceramics were analyzed using an X-ray diffractometer (Japanese Shimadzu XRD-6100, X-ray source was CuK, scanning range of 10˚ to 80˚, scanning speed of 8 ˚/min); The morphology of Silicon carbide ceramics was observed under scanning electron microscope (SU3500, Hitachi Corporation, Japan).

![Figure 1. Density of SiC ceramics with different ratios of B4C.](image)

3. Results and discussion

3.1. Effects of B4C on density of SiC ceramic

As can be seen from Figure 1, the density of the samples increases with the increase of the B4C quantity in the B4C addition range of 0-8 wt%. When B4C was not added (a), the density was much...
lower than that of the samples with B\textsubscript{4}C added (b, c, d, e). When the amount of B\textsubscript{4}C addition increased from 2 wt\% (b) to 4 wt\% (c), the increment of density was 0.079, which was greater than 4 wt\% (c)-6 wt\% (d) and 6 wt\% (d)-8 wt\% (e) (0.013 and 0.007). It could be seen that when B\textsubscript{4}C was added more than 4 wt\% (c), the increase effect of B4C on SiC density was gradually weakened, and after more than 6 wt\% (d), the effect was almost no longer changed, so it was of little significance to add more B\textsubscript{4}C to increase density. According to Figure 1, it could be speculated that the amount of B\textsubscript{4}C was optimal at about 6 wt\% (d).

3.2. Effects of B\textsubscript{4}C on crystal phase of SiC ceramic

As shown in Figure 2, it was known from the comparison with the standard card that the main crystal phase of SiC ceramics without B\textsubscript{4}C was SiC-6H, and the main crystal phase of SiC ceramics with B\textsubscript{4}C was SiC-4H. With the increase of B\textsubscript{4}C, the intensity of diffraction peaks of SiC-4H were gradually increasing, and those of SiC-6H were gradually weakening. It showed that B\textsubscript{4}C mainly promoted the generation of SiC-4H during the sintering.

![Figure 2. XRD spectra of SiC ceramics with different ratios of B\textsubscript{4}C.](image)

3.3. Effects of B\textsubscript{4}C on microstructure of SiC ceramic

Figure 3 contains images of SiC ceramic sections with different additions of B\textsubscript{4}C magnified by 1,000 times. It is not difficult to be seen from the figures that the particles of SiC ceramics that did not add B\textsubscript{4}C [Figure 3(a)] were close to spherical. The particles were only bounded to the adjacent particles, and the connected parts were few and the necks were relatively thin. The space between them were much in amount and small in size, the boundaries were clear, and the particle size was small; There were several particles bounded into shorter strips, forming particles of slightly larger size, in SiC ceramics with 2 wt\% B\textsubscript{4}C added [Figure 3(b)]. After the particles were bounded, the boundary become not obvious, and the connected parts were more than those in Figure a; The size of the particles in SiC ceramics with 4 wt\% B\textsubscript{4}C added [Figure 3(c)] was larger than that of Figure b, and the bonding parts were very dense. The general trend of strip bonding had basically emerged. All particles were almost interwoven into a whole, but the pores had also become larger; Most of the particles in SiC ceramics with 6 wt\% B\textsubscript{4}C added [Figure 3(d)] were sintered into regular plates, they were staggered together, and some of the particles were bonded into larger spherical particles, sandwiched between the plates or
attached to the plates, and the pores were larger than those in Figure d; There was a thicker plate structure in SiC ceramics with 8 wt% B₄C added [Figure 3(e)] than that in Figure d. The plates were closely embedded together. Only a few spherical particles adhere to the plates, and the overall structure is not much different from Figure d.

![Figure 3](image_url)

**Figure 3.** morphology of cross sections of SiC with different amount of B₄C: (a)0 wt% B₄C; (b)2 wt%; (c)4 wt%; (d)6 wt%; (e)8 wt%.

4. Summary

B₄C has a good effect on the increasing of density of SiC ceramics. With the increase of B₄C addition at the range of 0-8 wt%, the density of SiC ceramics increased. When B₄C was more than 6 wt%, the effect of increasing the dose of B₄C on the density of SiC ceramics changed little, and it was not significant to continue to increase the amount of B₄C. Therefore, the optimal amount of B₄C should be near 6 wt%. B₄C can change the SiC-6H crystal (spherical particles) to SiC-4H (plated structure), and with the increase of B₄C added, this change will be more obvious, which means, the relative content of SiC-4H will gradually increase with B₄C. The results of SEM and XRD were generally consistent. And when the amount of B₄C was 6 wt% or 8 wt%, the cross-section morphology of the samples changed little, which corresponded to the density analysis.

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