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# Effect of sintering conditions on microstructure orientation in α-SiC prepared by slip casting in a strong magnetic field

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**Abstract**

In general, the mechanical and physical properties of a crystal depend on the direction of the crystal axis. The controlled development of the crystallographic texture in ceramics is very useful for improvement of their properties. The preparation of the textured SiC polycrystal was achieved by slip casting in a strong magnetic field. The effects of the sintering conditions and sintering additives on the degree of orientation in the SiC were investigated. The pressing during the liquid phase sintering prevented the development of a texture in the SiC prepared by slip casting in a strong magnetic field.



*Keywords:* Slip casting; SiC; Hot pressing; Sintering; Magnetic field

## 1. Introduction

Silicon carbide (SiC) as a high-temperature material has many advantages, such as high strength and elastic modulus, low density and good resistance to wear. These properties make SiC a good candidate for use in a wide variety of applications. Many studies have reported the control of its microstructure in order to improve the sinterability, mechanical properties, etc., using many kinds of additives.[1–5] Several studies have reported that the sinterability and the mechanical properties of the SiC were improved by colloidal processing,[6–10] because the colloidal processing is generally able to control the dispersion of powders in a suspension and to produce a fine and dense microstructure.

Tailoring of the crystallographic texture in ceramics is very useful for the development of their properties, because the mechanical and the physical properties of a crystal depend on the direction of the crystal axis.[11] Many studies have reported the production of textured ceramics, such as by hot forging[12] and the templated grain growth (TGG) method.[13,14] Preparing the textured SiC by hot forging and the TGG method has also been reported.[15–17]

On the other hand, a strong magnetic field can be obtained based on the development of a superconducting magnet which has been used to control the crystallographic orientation even in diamagnetic ceramics.[18–24] A strong magnetic field was applied to particles in stable suspensions during the consolidation process. The particles with asymmetric unit cells, such as a tetragonal and hexagonal structure, were rotated to an angle minimizing the system energy by a magnetic torque. The magnetic torque, $T$, attributed to the interaction between the anisotropic susceptibility due to the asymmetric unit cell and the applied magnetic field is estimated using Eq. (1).[25,26]

$$T = -\frac{\Delta\chi V B^2}{2\mu_0}\sin 2\theta \qquad (1)$$

where $\Delta\chi$ ($=|\chi_\parallel - \chi_\perp|$) is the anisotropy of the susceptibilities, which is measured in the direction parallel ($\chi_\parallel$) and perpendicular ($\chi_\perp$) to the $c$-axis in the tetragonal and the hexagonal crystal systems, $V$ is the volume of each particle, $\mu_0$ is the permeability in a vacuum, $B$ is the applied magnetic field and $\theta$ is the angle between the easy magnetization axis in a crystal and the imposed magnetic field direction. This is the driving force for magnetic alignment. However, when small particles were used for preparing a fine microstructure, it is difficult to effectively apply a magnetic field in order to rotate small diamagnetic particles, because small particles tend to spontaneously agglomerate due to their strong attractive interactions (van der Waals forces). It is necessary to colloidally disperse the particles in a liquid in order to effectively utilize the magnetic field to rotate the par-

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ticles due to reduction in the attractive interaction between the particles.

In a previous paper, we reported that control of the crystallographic texture in a polycrystalline SiC could be achieved using colloidal processing and a strong magnetic field.[27] In this study, we report the effect of the sintering conditions and sintering additives on the degree of orientation and the textured microstructure.

## 2. Experimental procedure

A commercially available silicon carbide powder (OY-20, Yakushima Denko Co., Ltd., Japan) was used as the starting powder. Alumina powder (TM-DAR, Taimei Chemicals Co., Ltd., Japan) and yttria powder (RU-P, Shin-Etsu Chemical Co., Ltd., Japan) with the average particle sizes of 0.15 μm and 1.0 μm, respectively, were used as the sintering additives. Aqueous suspensions of pH 10 were prepared that contained 30 vol% solids; the solids consisted of SiC that included 5 mass% $Al_2O_3$ and 5 mass% $Y_2O_3$ and without sintering additives. The pH of the suspensions was adjusted using tetramethylammonium hydroxide (25 mass% in methanol). The suspensions were ultrasonicated for 10 min and stirred for more than 4 h. The suspensions were then consolidated by slip casting after evacuation in a vacuum desiccator to remove as many air bubbles as possible. A strong magnetic field of 12 T was applied to the suspension during the slip casting at room temperature. The direction of the magnetic field was parallel to the casting direction. For comparison, some samples were prepared by slip casting without applying a magnetic field. The green compacts were isostatically densified so as not to disturb the particle orientation by cold isostatic pressing (CIP) at 392 MPa for 10 min and then isothermally sintered by hot pressing (HP) at 2273 K for 2 h and a pressure of 40 MPa in an Ar atmosphere and spark plasma sintering (SPS) at the desired temperatures for 2 h at a pressure of 80 MPa in a nitrogen atmosphere.

The sintered samples were polished using diamond suspensions and plasma etched with $CF_4$. The microstructures of the sintered samples were observed by scanning electron microscopy (SEM). The crystallographic orientation was analyzed by X-ray diffraction (XRD) and an electron back scattering diffraction pattern (EBSD). The degree of orientation was calculated by the Lotgering factor in conjunction with the XRD intensities from the surface perpendicular to the magnetic field.

## 3. Results and discussion

It is found in Fig. 1 that the shape of particles was isotropic and the sizes were bimodal distribution. Fig. 2 shows the zeta potential of α-SiC as a function of pH. The isoelectric point was a pH less than 3 and the absolute value of the zeta potential was more than 40 mV at a pH of 10 which was sufficient for dispersion in a suspension.

Fig. 3 shows the particle size distribution before and after dispersion by control of pH. When the suspension was made without controlling pH, some particles were agglomerated. After controlling the pH of 10 according to Fig. 2, large particles with the size of about 0.6 μm were remained, but small particles were dispersed and it became clear that the size distribution was bimodal.

Fig. 4 illustrates the 0 0 0 1 pole figure on the surface perpendicular to the magnetic field in the additive-free SiC prepared using a magnetic field followed by HP at 2273 K. The pole fig-

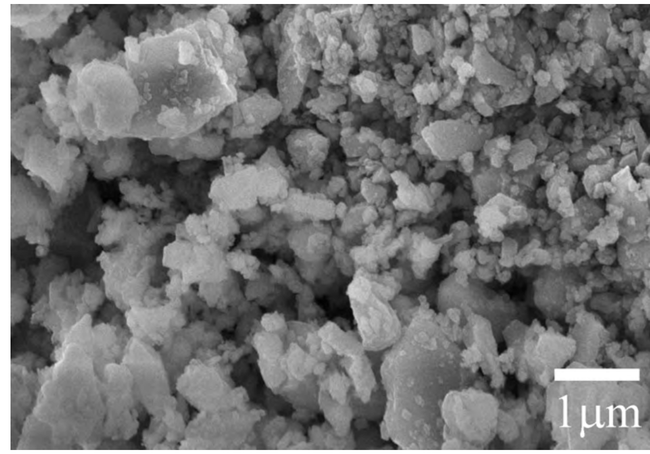


Fig. 1. SEM micrograph of the as-received SiC powder.

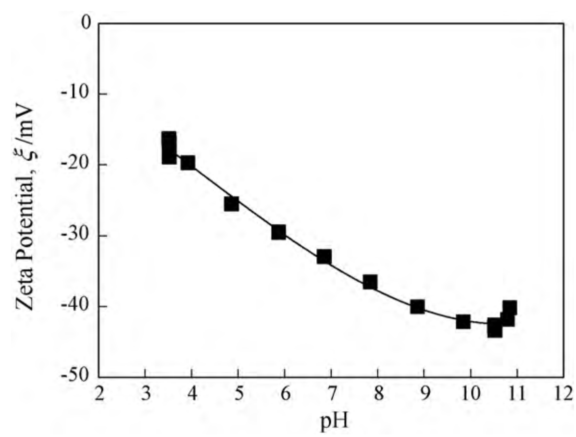


Fig. 2. Zeta potential of the SiC starting powder as a function of pH.

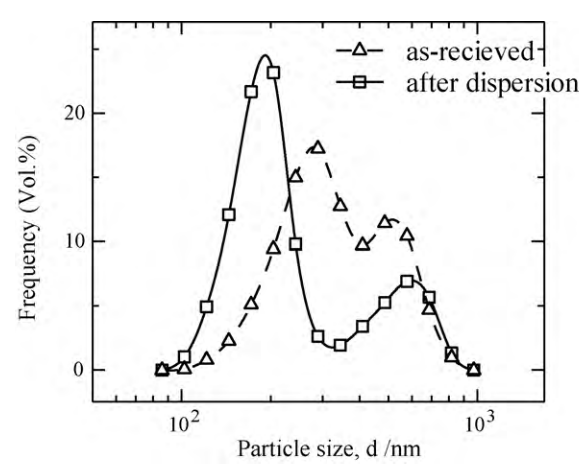


Fig. 3. Particle size distribution of SiC powder before and after dispersion by control of pH.

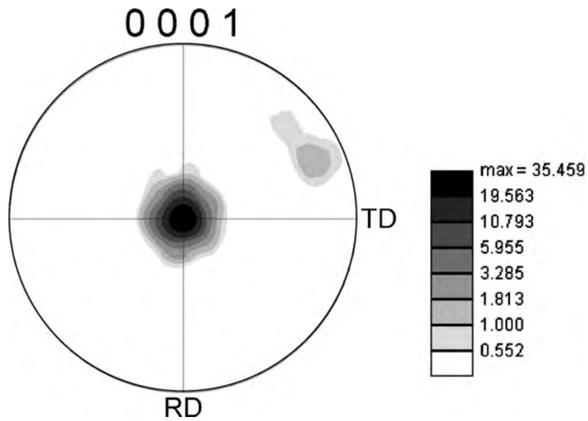


Fig. 4. 0 0 0 1 pole figure in the no-additive SiC prepared using a magnetic field followed by HP at 2273 K for 2 h in an Ar atmosphere.

ure clearly shows that the texture is radially symmetric and the (0 0 0 1) pole is very narrow. The maximum of the m.r.d. was at the center; consequently, the basal plane oriented on the plane perpendicular to the magnetic field and the *c*-axis was aligned parallel to the magnetic field.

In order to evaluate the degree of orientation, the distribution of the tilt angle between the *c*-axis and the vertical direction parallel to the magnetic field was calculated from the previous EBSD data. The result is shown in Fig. 5. The plots are our experimental data and the solid lines are the fitting curves using the March–Dollase function as follows:

$$f_{\mathrm{MD}}(r,\theta)=\left(r^2\cos^2\theta+\frac{\sin^2\theta}{r}\right)^{-3/2} \quad (2)$$

where *r* is the orientation parameter and $\theta$ is the tilt angle between the *c*-axis and the magnetic field. For the additive-free SiC prepared using a magnetic field followed by HP at 2273 K, the orientation parameter, *r*, was 0.30, indicating that approximate 72% of the grains were aligned with a tilt angle less than 10°. For the SiC with 5 mass% $Al_2O_3$ addition, *r* was 0.40, indicating that about 54% of the grains were aligned within 10° from the texture axis parallel to the magnetic field. The additives decreased the degree of orientation, because the additives inhibited the

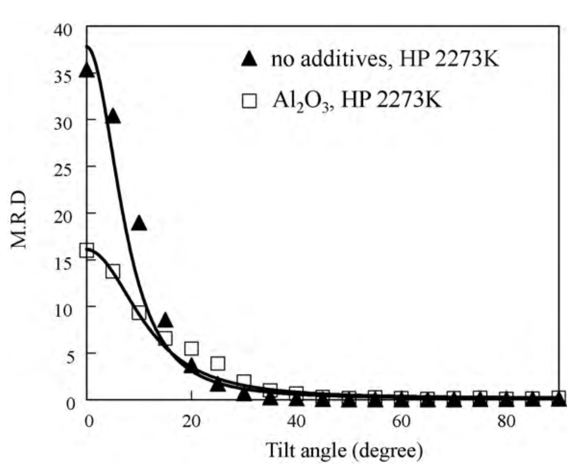


Fig. 5. Distribution of the tilt angle between the *c*-axis and the vertical direction in the additive-free SiC and the SiC containing 5 mass% $Al_2O_3$.

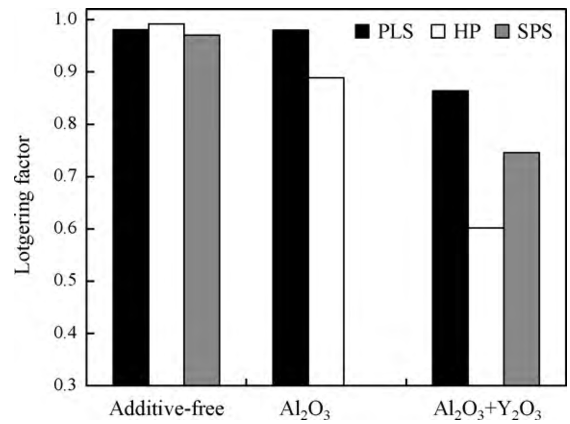


Fig. 6. Effect of the sintering conditions on the degree of orientation in the textured SiC prepared using a strong magnetic field. For the additive-free SiC, sintering temperatures in PLS and HP were 2273 K and the sintering temperature in SPS was 2223 K. For the SiC with $Al_2O_3$ addition, the sintering temperatures in PLS and HP were 2273 K. For the SiC with $Al_2O_3$ and $Y_2O_3$ additions, the sintering temperatures were 2173 K in all sintering methods.

dispersion of the SiC particles in a suspension. The decreasing degree of orientation due to the additives has also been reported in the textured AlN prepared by slip casting in a strong magnetic field.[28]

Fig. 6 shows the effect of the additives and the pressure during sintering on the degree of orientation. The degree of orientation was estimated by the Lotgering factor calculated from the XRD intensities. When the additive-free SiC and the SiC with $Al_2O_3$ addition were sintered at 2273 K at ambient pressure, both sintered samples had the same Lotgering factor. For the additive-free SiC, the degrees of orientation were almost the same regardless of the sintering process, such as the pressureless sintering, SPS and hot pressing. When using the orientation parameter calculated from the March–Dollase function, these parameters had the same value of 0.30 in both methods of pressureless sintering and hot pressing at 2273 K. However, when the sintering additives were used, the degree of orientation decreased during the SPS and hot pressing in which the pressure was applied to the green bodies during sintering. Since the liquid phase was generated at the sintering temperature in the SiC with additives addition,[29] the liquid phase to which the pressure has been applied seemed to prevent the crystallographic orientation. In the previous study,[16,17] it said that the deformation was significant factor for development of texture in SiC with liquid phase during hot forging, and grain rotation also enhanced the degree of texture during hot pressing. However, in this study, texture was already developed by a magnetic field before sintering, hence deformation and grain rotation during hot pressing and SPS suppressed the development of texture in SiC consolidated in a magnetic field. Grain growth of these textured ceramics prepared in a magnetic field enhanced the texture,[28] but for these SiC grain growth was small and the effect of grain growth on the texture was limited.

The SEM micrographs of the cross-sectional surface of the SiC with 5 mass% $Al_2O_3$ and 5 mass% $Y_2O_3$ additions are shown in Fig. 7. Fig. 7(a) and (c) shows the surfaces of the textured SiC parallel to the magnetic field sintered by the hot

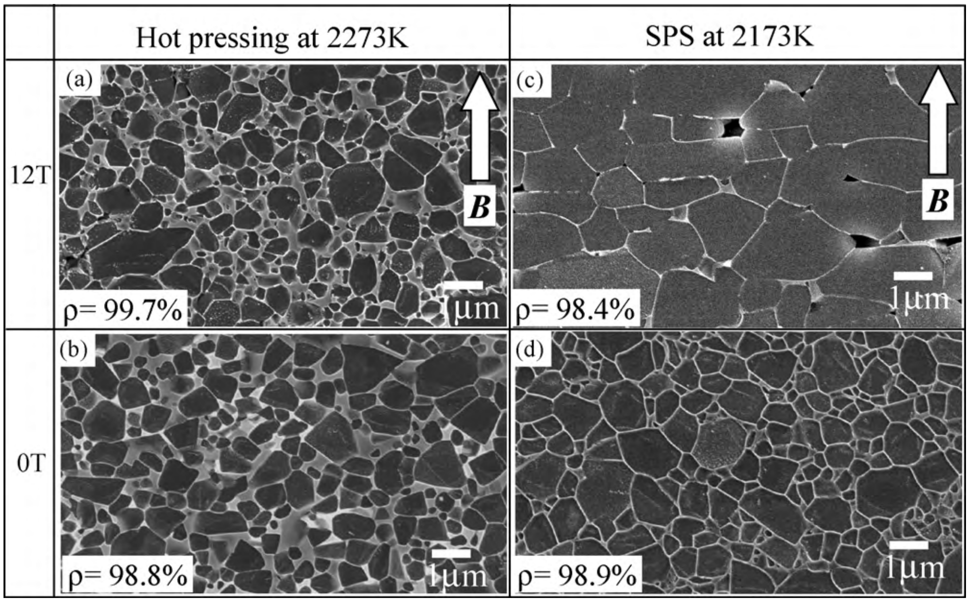


Fig. 7. SEM observation of cross-sectional surfaces of SiC containing 5 mass% $Al_2O_3$ and 5 mass% $Y_2O_3$ prepared with and without a magnetic field, followed by hot pressing and SPS at 2173 K: (a) the surface parallel to the magnetic field of SiC sintered by hot pressing in Ar, (b) the surface of the SiC without applying a magnetic field sintered by hot pressing in Ar, (c) the surface parallel to the magnetic field of SiC sintered by SPS in $N_2$ and (d) the surface of the SiC without applying a magnetic field sintered by SPS in $N_2$.

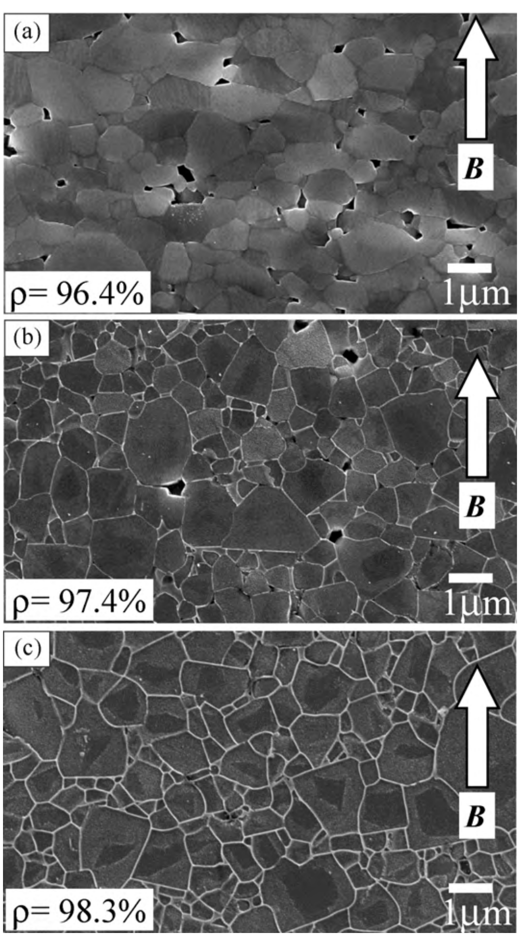


Fig. 8. SEM observation of cross-sectional surfaces parallel to the magnetic field in SiC prepared by applying a magnetic field, followed by SPS at 2173 K for 2 h in a $N_2$ atmosphere: (a) the additive-free SiC, (b) the SiC with 4 mass% $Al_2O_3$ addition and (c) the SiC with 3 mass% $Y_2O_3$ addition.

pressing and SPS, respectively. Fig. 7(b) and (d) shows the surfaces of the random SiC sintered by the hot pressing and SPS, respectively. In Fig. 7(a), (b) and (d), the equiaxed grains were observed regardless of the sintering process and the orientation. However, in the textured SiC containing 5 mass% $Al_2O_3$ and 5 mass% $Y_2O_3$, grains were elongated perpendicular to the magnetic field and large grain growth occurred when using SPS. Fig. 8 illustrates the microstructure of the cross-sectional surface of the additive-free SiC and the SiC with $Y_2O_3$ addition and with $Al_2O_3$ addition prepared in a strong magnetic field. Even using SPS for sintering, equiaxed grains were observed in these samples. Therefore, the large elongated grain growth occurred in the SiC containing both $Al_2O_3$ and $Y_2O_3$ sintered by SPS in Fig. 7(c).

## 4. Summary

Control of the crystallographic orientation in polycrystalline SiC has been achieved using a magnetic field, and the *c*-axis of the SiC crystal was parallel to the magnetic field. The pressing during the liquid phase sintering prevented the development of a texture in the SiC prepared by slip casting in a strong magnetic field. The grains became elongated and large grain growth occurred in the SiC with $Al_2O_3$ and $Y_2O_3$ additions when using SPS for densification.

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