Texture Development and Phase Transformation in Liquid-Phase-Sintered SiC Ceramics.

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Abstract. By using Y_2O_3 and Al_2O_3 as sintering aids, the effect of additive amount on the texture was investigated during the liquid-phase sintering of fine SiC powders, and the resulting microstructure and phase transformation of the hot-pressed and subsequently annealed materials were investigated. In this investigation quantitative texture measurements, including the pole figures and X-ray diffraction patterns, are used in conjunction with scanning electron microscopy to demonstrate the degree of preferred orientation and texture development mechanisms in these materials. The results show that annealing can produce textures and the degree of preferred orientation of hot-pressed and annealed SiC enhances with increasing amount of additives. The strong effect of additive amounts on texture was due to the anisotropic grain growth by the β→α phase transformation during annealing. The presence of a large amount of sintering aids accelerated the grain growth of elongated large grains during annealing and increased the texture intensity.

Introduction

The relationship between preferred orientation and mechanical properties of structural ceramics, which have been hot-worked, is one of the important issues to be investigated for engineering applications [1-4]. Several reports have been published in regard to in situ-toughened SiC [5-8]. It is well documented that high fracture toughness can be achieved through the development of elongated α-SiC grains, i.e., microstructural control for toughening was based on the β→α phase transformation. Highly textured SiC samples were fabricated by adding SiC platelets as seed and sintering the samples at relatively high temperature (≥ 2250^{o}C) [9]. Recently, Xie et al. [10] have investigated the texture development in hot-forged liquid-phase-sintered SiC ceramics with varying α/ β ratio in the starting powder. In this study, various in situ-toughened SiC ceramics that possess different additive amounts were prepared by hot-pressing and subsequent annealing. The preferred orientation for the hot-pressed and the annealed SiC was investigated using quantitative texture analysis and the texture formation and its mechanism were explained based on the microstructural observations.

Experimental Procedure

Commercially available β-SiC (Ultrafine, Ibiden Co., Ltd., Nagoya, Japan), Al_2O_3 (AKP-30,

Sumitomo Chemicals, Tokyo, Japan), and Y_2O_3 (grade Fine, H. C. Starck, Berlin, Germany) were used as the starting powders. The powder mixtures of SiC containing 5 wt% Al_2O_3-Y_2O_3 (designated as BS5), 10 wt% Al_2O_3-Y_2O_3 (BS10), and 20 wt% Al_2O_3-Y_2O_3 (BS20) as sintering additives were milled in ethanol with SiC grinding balls for 24 h. The ratio of Al_2O_3:Y_2O_3 was fixed as 6:4 by weight. Batch compositions are shown in Table 1. The milled slurry was dried, subsequently sieved through a 60 mesh screen, and hot-pressed at 1800℃ for 1 h under a pressure of 20 MPa in an argon atmosphere. The hot-pressed materials were heated further at 1900℃ for 4 and 8 h under an atmospheric pressure of Ar to enhance grain growth. Densities were measured using the Archimedes method. The theoretical densities of the specimens were calculated according to the rule of mixtures. X-ray diffractometry (XRD) was used to determine the crystalline phases. The hot-pressed and annealed materials were cut, polished, and then etched by using a plasma of carbon tetrafluoride (CF_4) that contained 7.8% oxygen gas. The microstructures were observed by scanning electron microscopy (SEM).

The texture of the materials was determined via pole-figure measurements, using the Schultz reflection method and Cu radiation on a pole-figure goniometer (Model XRD 3000, Seifert, Ahrensburg, Germany). The pole figures of {111}, {220} and {311} were measured for the hot-pressed material, and the pole figures of {100}, (004), {102}, {110} and {114} were measured for the hot-pressed and annealed material. From such incomplete pole figures, the ODF was calculated after correction and symmetrization using the series expansion method with expansion to $L_{max} = 22$. The C-coefficients were determined with a maximal rank of series expansion $L_{max} = 22$ using twenty iterations. Complete pole figures that were calculated from the ODF have been used to represent the texture of SiC in the present paper.

Table 1. Characteristics of both the hot-pressed and the annealed SiC

Sample designation	Composion (wt%)			Relative density(%)	Annealing time at 1900°C(h)	Crystalline phase	
	SiC	Al_2O_3	Y_2O_3			Major	Trace
BS 5	95	3	2	98.9	Hot-pressed	β-SiC	YAG
					4	α-SiC	YAG
					8	α-SiC	YAG
BS 10	90	6	4	98.2	Hot-pressed	β-SiC	YAG
					4	α-SiC	YAG
					8	α-SiC	YAG
BS 20	80	12	8	97.2	Hot-pressed	β-SiC	YAG
					4	α-SiC	YAG
					8	α-SiC	YAG

Results and discussion

The characteristics of the hot-pressed and annealed SiC are summarized in Table 1. Relative densities of ≥97% were achieved by hot-pressing and subsequent annealing. Phase analysis on the hot-pressed material via XRD showed that the major phase was β-SiC. The microstructures of the hot-pressed materials are shown in Fig. 1. Silicon carbide grains are etched away by the CF_4 plasma, so that the microstructures were delineated by the grain-boundary glassy phase. The microstructures of the hot-pressed materials consist of fine SiC grains and a small number of relatively large grains. These large grains grow preferentially during hot pressing, because the grain growth occurs via solution-reprecipitation [11]. The microstructures of the hot-pressed and annealed materials with different amount of additives are shown in Fig. 2. Phase analysis of the hot-pressed and annealed materials by XRD showed the presence of α-SiC as a major phase and YAG as a trace, indicating the occurrence of β→α transformation of SiC during annealing (see Table 1). As shown in Fig. 2, the microstructures of the hot-pressed and annealed materials consisted of elongated α-SiC grains, which were the typical microstructure of self-reinforced SiC. The true shape of elongated grains is considered as a plate-shape [11].

Fig. 3 shows the calculated pole figures for all samples undergoing hot-pressing and annealing. All pole figures given here are normal to the page, i.e.; the hot-pressing direction on the stress axis is at the center of each pole figure. The calculated {111} pole figures for the 3C phase in the as-hot- pressed specimens are shown in Fig. 3 (a-c). These pole figures show repeating contours with

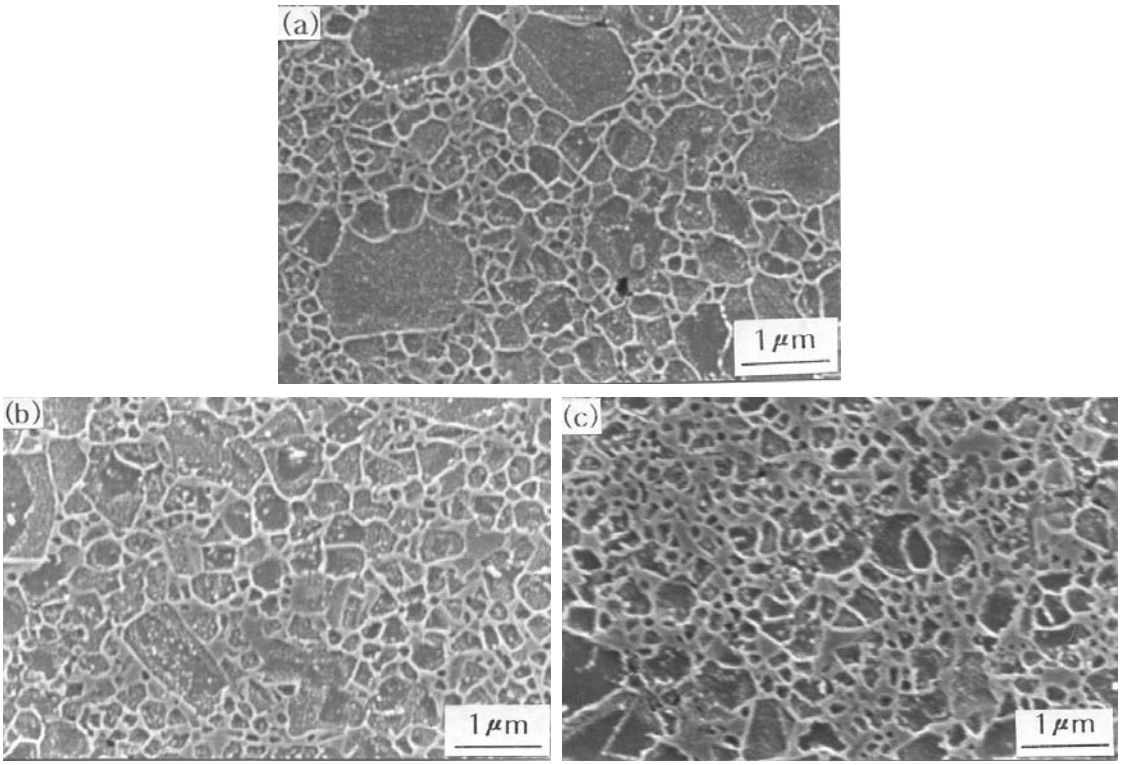

Fig. 1. SEM micrographs of hot-pressed materials : (a) BS5 (b) BS10 and BS20 (refer to Table 1).

Intensities of 1.0 and 1.2, which indicate that the calculated pole figure for the as-hot-pressed material is a diffusion one except at the center of the pole figures. The maximum intensities for the as-hot-pressed materials are 1, 1.2 and 1.4 times random at the center of pole figures for materials BS5, BS10 and BS20, respectively. The calculated (004) pole figures for the 4H phase in the hot-pressed and annealed specimens are shown in Fig. 3 (d-f). The basal pole figures for the annealed materials differ greatly from those for the hot-pressed materials. In the case of BS5(8h), the maximum intensity is 1.4 times random at an orientation of 90° from the center of the pole figure. The effect of the amount of additives on the texture is also shown in Fig. 3 (d-f). The degree of the preferred orientation of the materials after 8 h-annealing increased, i.e. from 1.4 for BS5 to 2.0 for BS20. In contrast, the diameter of SiC grains decreased with increasing amounts of additives as shown in Fig. 2. The strong effect of additive amounts on texture is attributed to the anisotropic grain growth of α- SiC grains during annealing

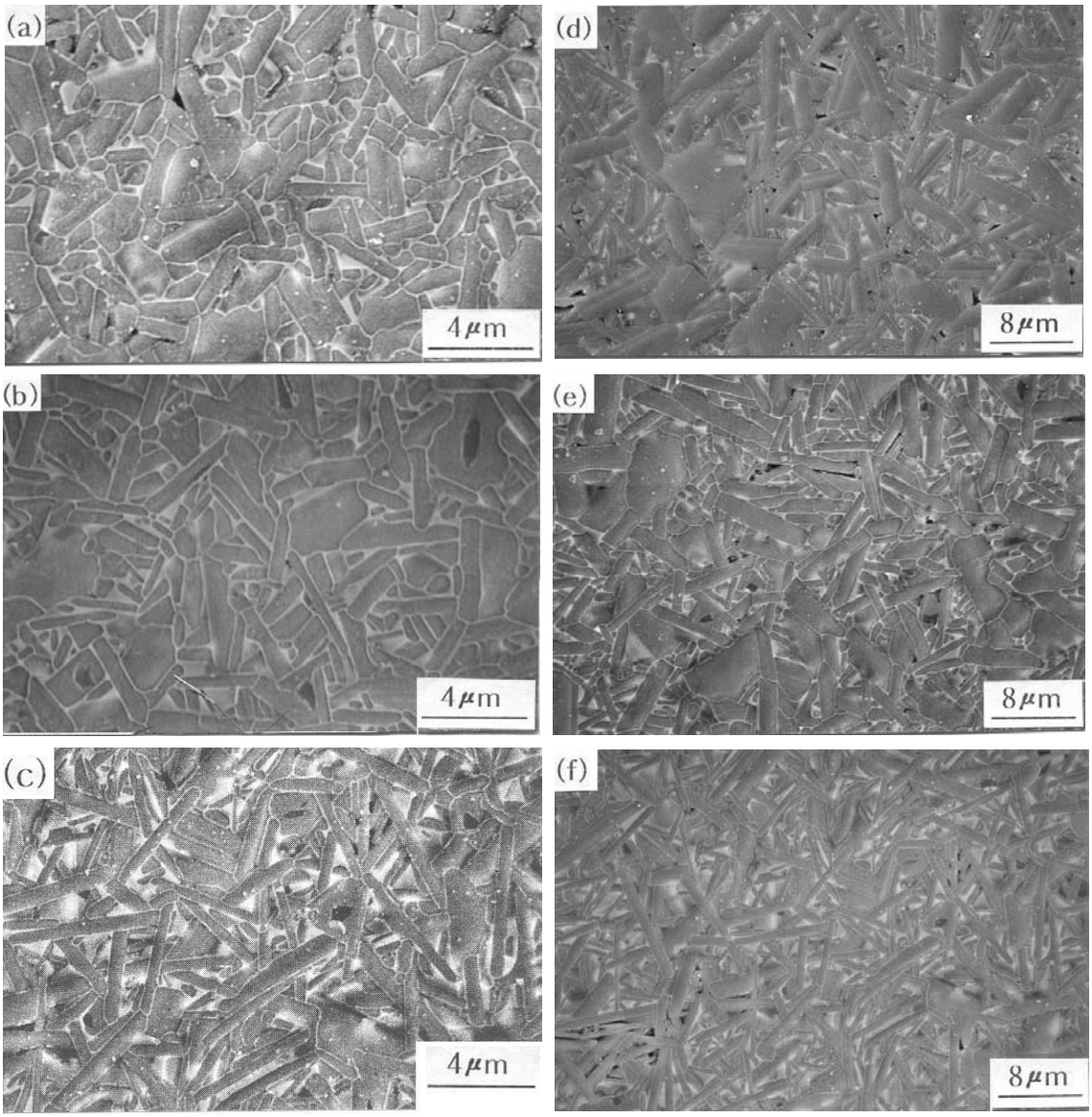

Fig. 2. SEM micrographs of hot-pressed and annealed materials : (a) BS5(4h), (b) BS10(4h), (c) BS20(4h), (d)

BS5 (8h), (e) BS10(8h), and (f) BS20(8h) (refer to Table 1)

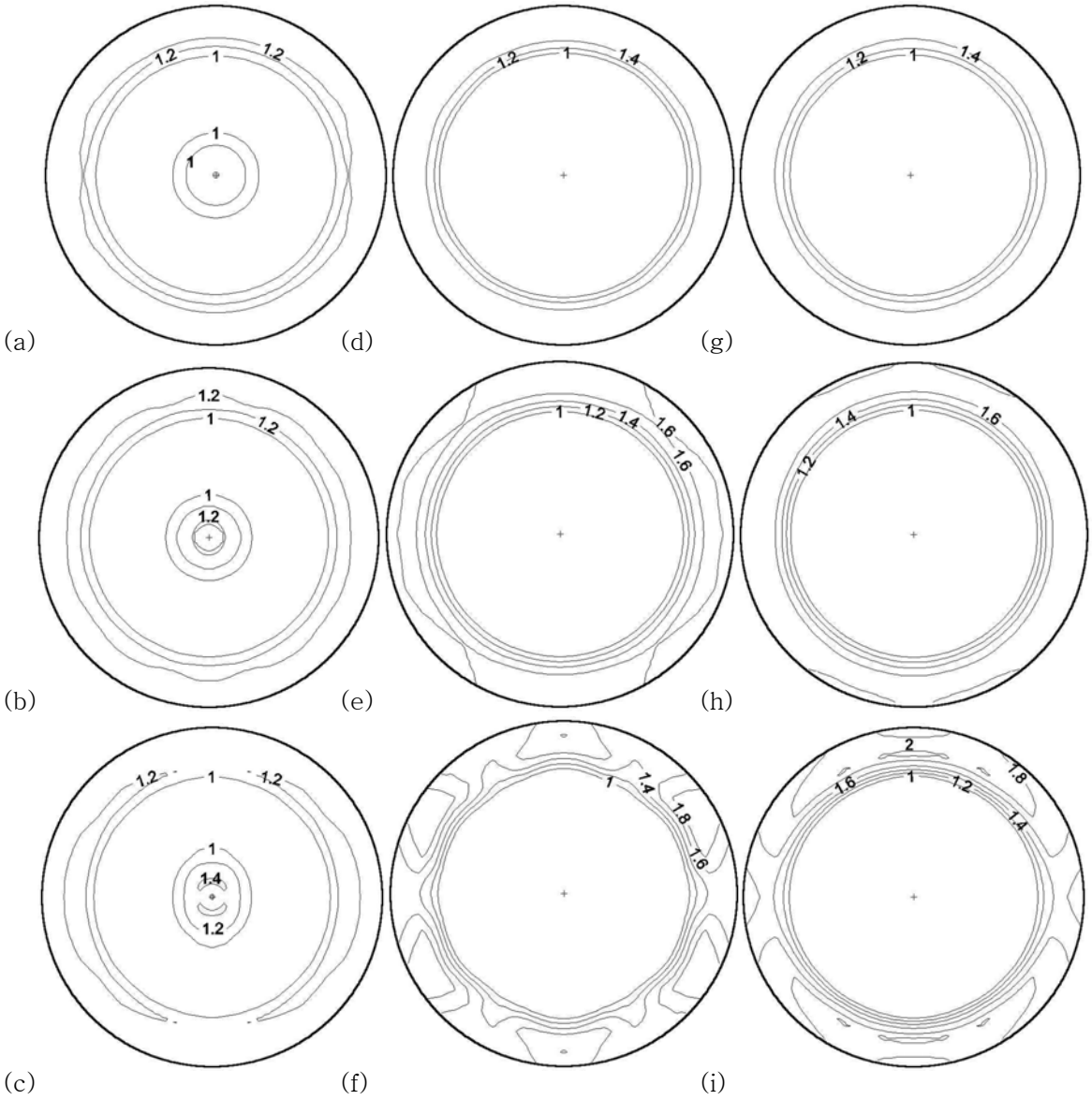

Fig. 3. Calculated {111} pole figures of hot-pressed SiC for (a) BS5, (b) BS10, (c) BS20. Calculated basal (004) pole figures of hot-pressed and annealed SiC for (d) BS5(4h), (e) BS10(4h), (f) BS20(4h), (g) BS5(8h), (h) BS10(8h), and (i) BS20(8h) (refer to Table 1).

The presence of a large amount of sintering aids accelerated the grain growth of elongated large grains during annealing and led to a tendency of the texture degree increasing. The effect of annealing time on the texture of self-reinforced SiC is also shown in Fig. 3. The texture intensity of BS20 increased, i.e. from 1.8 for BS20(4h) to 2.0 for BS20(8h), with increasing the annealing time.

Conclusion

A detailed analysis of the phase transformation from β- to α-SiC and the corresponding texture development of liquid-phase-sintered SiC with Al_2O_3 and Y_2O_3 as sintering additives are reported. In

the absence of a phase transformation a fine-grained equiaxed microstructure develops, and the β-SiC grains in the as-hot-pressed specimens are randomly oriented. In contrast, texture development occurs in various in situ-toughened SiC that possess different amounts of additives. During the subsequent heat treatment after hot-pressing, the β→α phase transformation of SiC produces a microstructure of "in situ-composites" as a result of the growth of elongated large α-SiC grains. The texture development in the hot-pressed and subsequently annealed material was due to the preferred grain growth of the plane-shaped grains whose (004) planes of α-SiC grains were preferentially oriented parallel to the hot-pressing direction. The texture intensity of samples containing in-situ grown SiC platelets with high aspect ratio is about twice that of samples with equiaxed grains.

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