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Texture Development in Liquid-Phase-Sintered β-SiC by Seeding with β-SiC Whiskers

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ABSTRACT

Silicon carbide ceramics seeded with 10 – 30 wt% SiC whiskers are fabricated by hot pressing and annealing. A quantitative texture analysis including calculation of the Orientation Distribution Function (ODF) is used for obtaining the degrees of preferred orientation of the fabricated samples. The microstructure and crystallographic texture are discussed with respect to the effect of β-SiC whisker seeds on the resulting fracture toughness values. The SEM microstructures and the texture data reveal a correlation between texture and fracture toughness anisotropy.

Key words : Silicon carbide, Texture, Anisotropy, Phase transformation

1. Introduction

Interest in the fabrication of textured SiC[1-6] has recently expanded because these materials appear to have superior properties relative to randomly oriented SiC. The textures of SiC ceramics vary for different processes and they can be modified by changing the processing variables during fabrication. This implies that the analysis of texture in terms of the orientation distribution would provide a useful tool to understand and control the underlying mechanisms during processing of polycrystalline SiC ceramics. Hot forging has been reported to produce textured SiC ceramic.[3,4] Kim *et al.*[5] produced a textured SiC by annealing as-hot pressed β-SiC, where the texture formation mechanism was identified as phase-transformation induced anisotropic grain growth. It has also been observed that texture can develop via seeded anisotropic grain growth.[1,2]

This letter reports the effect of β-SiC whisker (seeds) content on texture development and fracture toughness anisotropy of β-SiC ceramics. Three LPS SiC ceramics with varying amounts of β-SiC whisker seeds were prepared by hot-pressing and subsequent annealing.

2. Experimental Procedure

Commercially available β-SiC (Ultrafine T-1 grade, Sumitomo-Osaka Cement Co., Tokyo, Japan), SiO_2 (Reagent grade, Kanto Chemical Co., Inc., Tokyo, Japan), MgO (99.99% pure, Wako Pure Chemical Industries, Ltd., Osaka, Japan), Y_2O_3 (99.9% pure, Shin-Etsu Chemical Co., Tokyo, Japan), Al_2O_3 (AKP-30, Sumitomo Chemicals, Tokyo, Japan), and AlN (Grade F, Tokuyama Soda Co., Tokyo, Japan) powders were prepared to an oxynitride composition of $Y_{0.124}Mg_{0.160}Si_{0.414}Al_{0.302}O_{1.400}N_{0.151}$ by ball milling in ethanol for 24 h using a polyethylene jar and SiC balls. The oxynitride composition had an appreciable SiC solubility at high temperatures. The SiC powder was eventually blended with a 15 wt% powder mixture of oxynitride composition and milled in hexane for 20 h using SiC media so as to avoid contamination during processing. Then, 10 – 30 wt% β-SiC whiskers (Tokamax, Tokai Carbon Co., Ltd., Tokyo, Japan) were added as seeds, and the composition was milled for 4 h. The batch compositions are given in Table 1. After milling, the slurry was dried and hot-pressed at 1900°C for 1 h under a pressure of 20 MPa in a nitrogen atmosphere. The heating rate was 20°C/min, and the cooling rate was ~40°C/min from 1900°C to 1200°C. Some samples were further annealed at 2000°C for 4 h under a pressure of 20 MPa in a nitrogen atmosphere in order to observe the evolution of texture.

The sintered density was determined using the Archimedes method. The theoretical density of the materials, 3.21 g/cm^3, was calculated according to the rule of mixtures (the theoretical density of the oxynitride glass was 3.18 g/cm^3).[7] The hot-pressed and the annealed materials were cut and polished, and then etched by a plasma of CF_4 containing 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 {004},{100}.{102},{110}, and {114} were mea-

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Table 1. Batch Composition and Relative Density of the Sintered Materials

Sample designation	Composition (wt%)			Relative density (%)
	β-SiC (T-1 grade)	SiC whisker	Oxynitride glass	
SA10	75	10	15	99.0
SA20	65	20	15	99.5
SA30	55	30	15	99.8

*Hot-pressing condition 1900°C/1 h/20 MPa/N_2; Annealing condition 2000°C/4 h/20 MPa/N_2

sured 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 $L_{max}=22$.[8] The C-coefficients were determined with a maximal rank of series expansion $L_{max}=22$ using twenty iterations. Complete pole figures calculated from the ODF have been used to represent the texture of SiC in the present paper. The fracture toughness was estimated by measuring crack lengths generated by a Vickers indenter.[9]

3. Results and Discussion

The relative densities for the hot-pressed materials are shown in Table 1. The microstructures of the hot-pressed and annealed materials with different amount of seeds are shown in Figs. 1 and 2. Silicon carbide grains are etched away by CF_4 plasma, and thus the microstructures are delineated by the grain-boundary glassy phase.

Figs. 1 and 2 show SEM micrographs of three samples with 10, 20, and 30 wt% whiskers, respectively. Interestingly, the same material shows distinctly different images depending on the planes observed. As shown in Fig. 1, microstructures taken from the surface parallel to the hot-pressing axis mainly consist of equiaxed grains growing from the whisker seeds. In contrast, images taken from the surface perpendicular to the stress axis show duplex microstructures consisting of elongated grains and equiaxed grains (Fig. 2). Large grains growing from the whisker seeds appear as elongated grains on the surface perpendicular to the hot-pressing direction. The material with 10 wt% seeds exhibits a bimodal grain distribution whereas the material with 20 wt% seeds displays a unimodal distribution. In Fig. 1(b), because of the impingement of growing grains, the sample containing 20 wt% seeds is composed of large grains only. The number of large grains is proportional to the amount of seeds added. The average aspect ratios of the large grains decrease with 30 wt% addition of seed. In the material with 30 wt% seeds, the driving force for grain growth in the length direction decreases because distances between large grains are very short. Furthermore, the driving force for grain growth in the diameter direction increases due to contact among growing seeds grains, reflecting grains with low aspect ratio (Fig. 1(c)). Figs. 1 and 2 reveal a core-rim grain structure. Large elongated grains growing from the whisker seeds appear to be aligned well on the surface perpendicular to the hot-pressing direction (Fig. 2) and equiaxed grains on the surface are parallel to

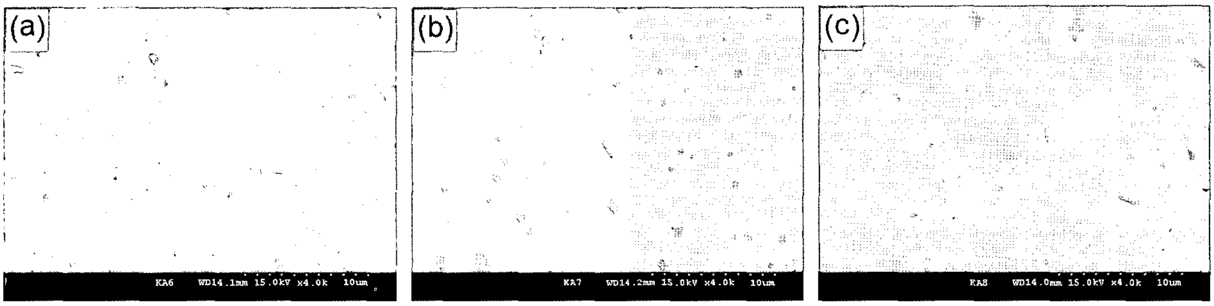

Fig. 1. Scanning electron micrographs from surface parallel to the hot-pressing axis for hot-pressed and annealed β-SiC ceramics containing : (a) 10 wt% (b) 20 wt%, and (c) 30 wt% β-SiC whiskers as seeds.

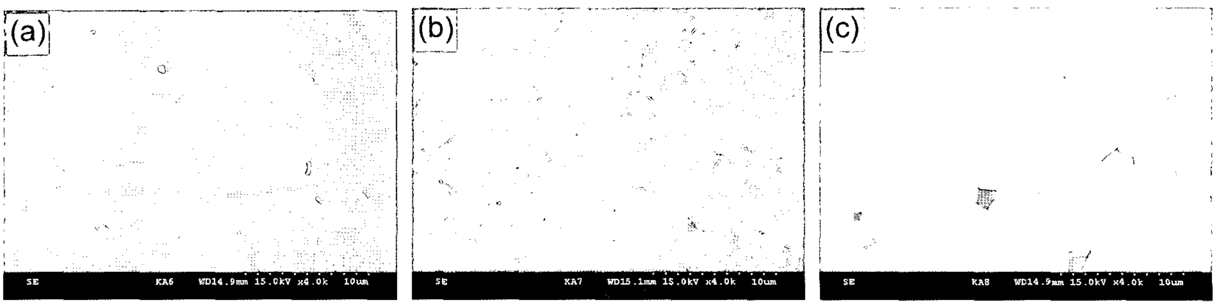

Fig. 2. Scanning electron micrographs from surface perpendicular to the hot-pressing axis for hot-pressed and annealed β-SiC ceramics containing : (a) 10 wt% (b) 20 wt%, and (c) 30 wt% β-SiC whiskers as seeds.

the hot-pressing direction (Fig. 1). Figs. 1(a)-(c) show the microstructures on the surface parallel to the hot-pressing direction, clearly displaying differences in the degree of alignment among the samples. Further addition of seeds (20 wt%) led to a better degree of alignment. It appears that larger elongated grains are grown from whiskers, and whiskers in this experiment operate as seeds for abnormal grain growth. When the amount of seeds is further increased (30 wt%), the respective interactions of the seed increase, leading to a decrease in the aspect ratio of large grains. These results suggest that the growth of grains proceeds predominantly along the c-axis of hexagonal β-SiC grains, which is related to the β→α phase transformation of SiC during annealing.

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 or the stress axis is at the center of each pole figure. Fig. 3(a)-(c) present the calculated 4H (004) pole figures of the hot-pressed and annealed materials seeded with 10 – 30 wt % SiC whiskers. The calculated 4H (004) pole figure of

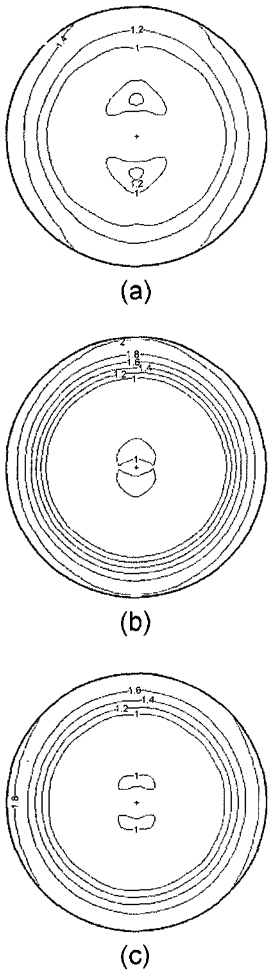

Fig. 3. Calculated (004) pole figures for the hot-pressed and annealed materials : (a) 10 wt%, (b) 20 wt%, and (c) 30 wt% β-SiC whiskers as seeds.

the hot-pressed and annealed material with 10 wt% seeds is shown in Fig. 3(a). The maximum intensity is 1.4 times random at an orientation of 90° from the center of the pole figure. The texture is almost random in the material with 10 wt% whiskers, but the pole figure still exhibits some intensity at an orientation of 90° from the hot-pressing direction. The results indicate that some preferably oriented α-SiC grains, whose basal plane is parallel to the stress axis, have grown during annealing. This is in agreement with other reports for both SiC[3,5] and Si_3N_4.[10-14]

Further addition of whiskers (20 wt%) increases the degree of the preferred orientation. The maximum intensity is 2.0 times random at an orientation of 90° from the center of the pole figure. The degree of the preferred orientation of the material with 20 wt% whisker increases, i.e. from 1.4 (Fig. 3(a)) to 2.0 (Fig. 3(b)). The presence of a large amount of SiC whiskers accelerates the growth of large grains during annealing and leads to an increase in texture degree. The strong effect of whisker amounts on the texture is attributed to the preferred grain growth of the α-SiC grains from the whisker seeds that are produced by the β→α phase transformation during annealing, as illustrated clearly in the SEM micrographs of Fig. 1. It is apparent from these results that the growth of these grains has proceeded predominantly along the direction of the whisker, i.e., along the c-axis of hexagonal α-SiC grains during annealing, as expected. However, adding more seeds up to 30 wt% leads to a decrease in the texture intensity, compared to the material with 20 wt% seeds due to the impingement of growing grains from the seeds. The calculated maximum for the material with 30 wt% whiskers is 1.8 times random for an orientation of 90 from the hot-pressing direction. When the amount of SiC whiskers is increased, the intensity decreases slightly. Comparing the degrees of texture for the material with 10 – 30 wt% whiskers, it is clear that there is a relatively strong preferred orientation for the material with 20 wt% seeds.

This means that for the material with 20 wt% whiskers,

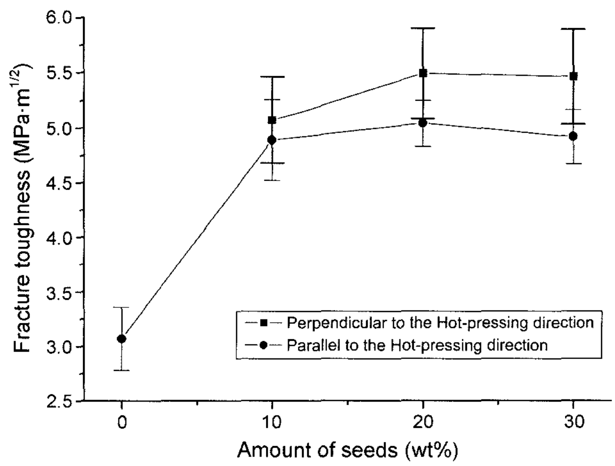

Fig. 4. Fracture toughness of the liquid-phase sintered β-SiC ceramics as a function of seeds.

most of the grains tend to align their basal planes parallel to the stress axis, or in other words, the c axes of α-SiC grains are normal to the stress axis. It can be deduced from the SEM micrograph that the material with 20 – 30 wt% whiskers has some preferred orientations, and α-SiC is textured, as evidenced by the pole figure measurements. Under the applied pressure during densification the majority of whiskers were aligned perpendicular to the stress axis. The newly formed α-phases growing anisotropically from the whisker seeds are aligned well with the c axis normal to the hot pressing direction. This finding agrees well with observations of the SEM micrographs of the material with 10 – 30 wt% whiskers. The microstructures and the texture data reveal that the majority of whiskers were aligned perpendicular to the stress axis during hot-pressing and the grains with whiskers grew significantly after annealing and increased the texture intensity.

Fig. 4 shows the fracture toughness as a function of seed content. The as hot-pressed material without seeds has a fracture toughness of 3.4 MPa · $m^{1/2}$. In contrast, the material with 10 wt% seeds, which is hot-pressed and annealed, has a fracture toughness of 5.0 MPa · $m^{1/2}$ on the surface perpendicular to the stress axis. The growth of coarse α-SiC grains produced a remarkable improvement in toughness. Adding more seeds up to 20 wt% leads to an increase in toughness. The facture toughness value of the samples with 20 wt% whiskers was 5.5 MPa · $m^{1/2}$ on the surface perpendicular to the hot-pressing direction. However, the addition of more seeds up to 30 wt % results in a decrease in toughness, compared to the material with 20 wt% seeds, because of the impingement of growing grains. Interestingly, the fracture toughness varies depending on the planes measured. The fracture toughness values measured from the surface perpendicular to the hot-pressing direction are higher than those measured from the surface parallel to the stress axis. This finding is in agreement with both the quantitative texture analysis results and microstructural evidence reflecting the anisotropic nature of the material with 10 – 30 wt% whiskers. The material with 10 – 30 wt% whiskers has a preferred SiC grain orientation of the c-axis perpendicular to the hot-pressing direction.

4. Conclusions

Texture can evolve in LPS SiC ceramics with whisker seeds during hot pressing and annealing. The maximum intensity of a sample with 20 wt% whiskers is 2.0 times random at an orientation of 90° from the center of the pole figure. The majority of whiskers are aligned perpendicular to the stress axis during hot-pressing and the grains with whiskers grow significantly during annealing and increase the texture intensity. During annealing, newly formed α-phases growing anisotropically from the whisker seeds are aligned with the c-axis normal to the hot-pressing direction. Fracture toughness anisotropy correlates well with texture intensities.

Acknowledgments

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