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Effect of SiC whisker addition on microstructure and mechanical properties of silicon carbide

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탄화규소 휘스커 첨가가 탄화규소의 미세구조와 기계적 특성에 미치는 영향

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Abstract β-SiC powder with or without the addition of 1-3 wt% of β-SiC whiskers (seeds) was hot-pressed at 1850℃ for 1 h using Al_2O_3 and Y_2O_3 as sintering aids. The hot-pressed materials were subsequently annealed at 1950℃ to enhance grain growth. The introduction of β-SiC whiskers into β-SiC does not affect the microstructure as well as mechanical properties significantly because the whiskers are not viable in the presence of liquid phase during hot-pressing. The strengths and fracture toughnesses of the hot-pressed and subsequently 5 h-annealed materials with 1 wt% β-SiC whiskers and without β-SiC whiskers were 465 MPa and 5.8 MPa·$m^{1/2}$, and 451 MPa and 5.5 MPa·$m^{1/2}$, respectively.

요 약 소결조제로 Al_2O_3와 Y_2O_3를 첨가한 β-SiC 분말에 seed로서 1 wt% 및 3 wt% β-SiC 휘스커를 첨가한 시편과 첨가하지 않은 시편을 1850℃에서 1시간 동안 고온가압소결하였다. 고온가압소결한 시편은 입자성장을 촉진하기 위하여 1950℃에서 5-10 시간 동안 열처리를 행하였다. Seed로서 1-3 wt% β-SiC 휘스커의 첨가는 액상소결 탄화규소의 미세구조와 기계적 특성에 의미 있는 영향을 미치지 않았다. 이는 β-SiC 휘스커가 액상이 존재하는 고온가압소결 조건에서 불안정하기 때문이라고 생각된다. β-SiC 휘스커를 1 wt% 첨가하여 1950℃에서 5시

간 동안 열처리한 시편의 강도 및 파괴인성은 각각 465 MPa 및 5.8 MPa $m^{1/2}$이었고, 동일조건에서 제조된 β-SiC 휘스커를 첨가하지 않은 시편의 강도 및 파괴인성은 각각 451 MPa 및 5.5 MPa·$m^{1/2}$이었다.

1. Introduction

Since the first successful sintering of SiC with small additions of boron and carbon in the 1970s [1], SiC has been attracted much attention as one of the promising candidates for high-temperature structural applications due to their excellent high-temperature strength, creep, and wear resistances. However, application of this ceramic as a structural material has been limited by their low fracture toughness, typically of 2.5 to 4 MPa·$m^{1/2}$, even though their superior other properties. A number of attempts have been made to improve the fracture toughness of SiC, with toughness values reported as high as 4 to 8 MPa·$m^{1/2}$ [2-6]. These efforts include toughening by heterophase dispersion [2], by incorporation of α-SiC platelets in a β-SiC matrix [3], by incorporation of β-SiC whiskers in a β-SiC matrix [4], and by introduction of both elongated α-SiC grains and weak interface in microstructure [5-9], in which grain bridging and crack deflection are promoted by the presence of weak interface. The latter efforts were achieved by liquid phase sintering with the addition of oxide additives, such as Al_2O_3 and Y_2O_3, and subsequent annealing for grain growth of elongated α-SiC grains. This provided self-reinforced material with duplex microstructure, akin to that of tough Si_3N_4 [10]. The development of elongated α-SiC grains has been achieved through the $\beta \rightarrow \alpha$ phase transformation.

Recently, several reports have been published on the effect of seeds on microstructure and mechanical properties of self-reinforced SiC [6,11,12]. These efforts include seeding by α-SiC [6,11] or large β-SiC particles [12]. SiC whiskers may have an advantage as seeds for microstructural control because of their high aspect ratio if they are viable under sintering conditions.

In the present work, β-SiC whiskers were selected as seeds for self-reinforced microstructure, and the effect of β-SiC whisker addition on microstructure and mechanical properties of self-reinforced SiC was investigated.

2. Experimental procedure

Commercially available fine β-SiC powder (Ibiden Co., Ogaki, Japan, Ultrafine grade) and β-SiC whiskers (American Matrix Inc., Knoxville, U.S.A, grade I) were used as starting materials. The powder characteristics of starting materials are shown in Table 1. A scanning elec-

Table 1
Characteristics of starting materials

No.	Raw material	Characteristic		
1	β-SiC powder*	Specific surface area(m^2/g)		16.7
		Impurity (wt%)	SiO_2	1.55
			C	0.75
		Phase		β
2	β-SiC whisker*	Diameter (μm)		1-3
		Length (μm)		30-200
		Impurity (wt%)	Ca	0.10
			Mg	0.03
			Al	0.02
			Fe	0.02
			Ni	0.02
		Phase		β

*Data were supplied from manufacturers.

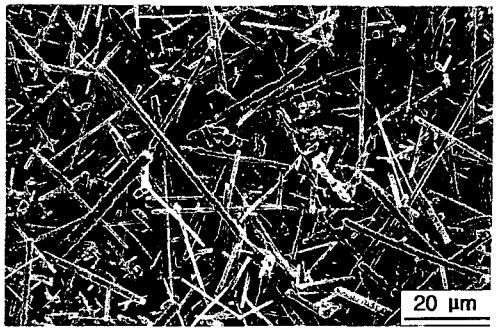

Fig. 1. SEM micrograph of β-SiC whiskers.

tron micrograph (SEM) of β-SiC whiskers is shown in Fig. 1.

To prepare a powder composition without seeds, 86 wt% β-SiC, 6 wt% Al_2O_3 (Sumitomo Chem. Co., 99.9 % pure, AKP-30), and 8 wt% Y_2O_3 (H.C. Starck, 99.9 %, grade Fine) were ball milled in ethanol with SiC grinding balls for 24 h. To prepare powder compositions containing seeds, 83-85 wt% β-SiC, 6 wt% Al_2O_3, and 8 wt% Y_2O_3 were ball milled in ethanol for 22 h, and then 1-3 wt% β-SiC whiskers (seeds) were added, followed by additional milling for 2 h. The milled slurry was dried and hot-pressed at 1850℃ for 1 h under a pressure of 20 MPa in an argon atmosphere. The hot-pressed materials were heated further at 1950℃ for 5 and 10 h under an atmospheric pressure of argon to enhance grain growth. A powder bed with the same composition as the specimens was used to suppress the weight loss of the specimens during annealing [13]. The sample designations are given in Table 2.

Relative density was determined by the Archimedes method. X-ray diffraction using Cukα radiation was performed on all the ground specimens. The hot-pressed and an-

Table 2
Sample designation

Annealing time at 1950℃ (h)	Sample designation		
	Material without β-SiC whisker	Material with 1 wt% β-SiC whisker	Material with 3 wt% β-SiC whisker
Hot-pressed	A0	B0	C0
5	A1	B1	C1
10	A2	B2	C2

nealed materials were cut and polished, and then etched by a plasma of CF_4 containing 7.8 % O_2. The microstructures were observed by SEM. The fracture toughness was estimated by measuring crack lengths generated by a Vickers indenter with a load of 196 N [14]. Bend tests were performed on 8 specimens at each condition using a four-point method with outer and inner spans of 20 and 8 mm, respectively, and a crosshead speed of 0.5 mm/min.

3. Results and discussion

The characteristics of SiC materials obtained with β-SiC whiskers and without β-SiC whiskers are summarized in Table 3. The relative densities of >97.5 % were achieved by hot-pressing with a holding time of 1 h at 1850℃. The sintered densities were found to decrease with increasing the annealing time at 1950℃. It may be due to the reactions between SiC and oxide additives (Al_2O_3 and Y_2O_3) at annealing temperature (1950℃) [15]. These reactions probably result in the formation of volatile components such as AlO, Al_2O, and CO [13], which make the substantial weight loss and the reduction of liquid content, leading to the decrease in the sintered density.

Figure 2 shows the microstructural change of SiC materials with annealing time. The microstructures of hot-pressed materials with or without β-SiC whiskers composed of equiaxed grains and the polytype of the materials was found to be composed primarily of 3C (β-SiC) and traces of α-SiC and yttrium aluminum garnet ($Y_3Al_5O_{12}$, YAG) by XRD. When the annealing time was increased, the shape of the grains changed from equiaxed to elongated and the grain size and aspect ratio increased, as shown in Fig. 2 (b), (c), (e), and (f), which indicate, referring to the phase analysis in Table 3 the marked growth of α-SiC. It is well documented that the $\beta \rightarrow \alpha$ phase transformation of SiC takes place at high temperatures (≥1950℃), especially in the presence of proper liquids. The 5-and 10-h annealed materials with 1 wt% β-SiC whiskers (Fig. 2(e) and (f)) have similar microstructures with the equivalent materi-

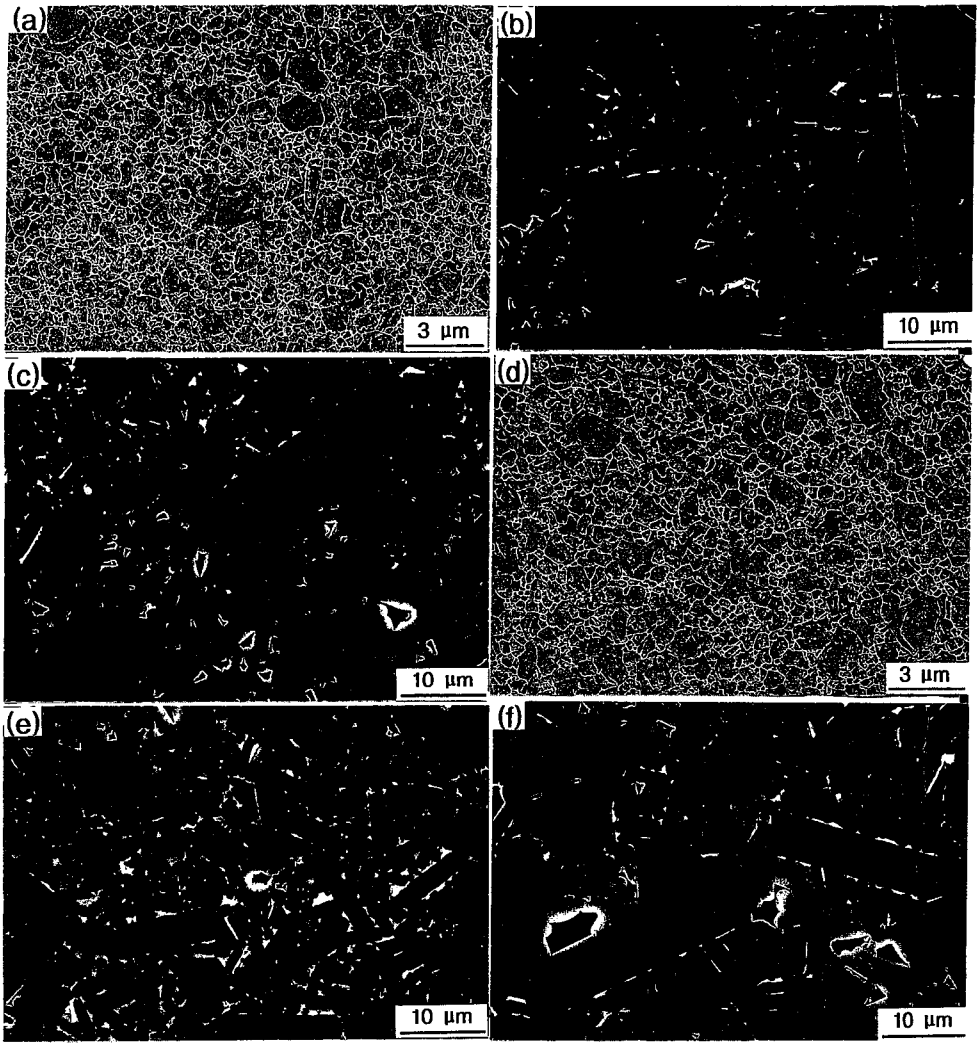

Fig. 2. SEM micrographs of SiC materials:(a) A0, (b) A1, (c) A2, (d) B0, (e) B1, and (f) B2 (refer to Table 2).

als without β-SiC whiskers (Fig. 2(b) and (c)). The microstructures of 5-and 10-h annealed materials with 3 wt% β-SiC whiskers were not contained in Fig. 2 because they had almost the same microstructures with the equivalent materials with 1 wt% β-SiC whiskers. These results suggest that the microstructure of self-reinforced SiC was not affected by the addition of β-SiC whiskers. It indicates that the β-SiC whiskers are not viable in the presence of liquid phase during hot-pressing, as evidenced by Fig. 2(d), where we cannot find the presence of SiC whiskers. That phenomenon may have resulted from both the instability of β-SiC under

Table 3
Characteristics of hot-pressed and annealed SiC

Material	Annealing time at 1950℃ (h)	Relative density (%)	Crystalline phase		Flexural strength (MPa)	Fracture toughness (MPa·m$^{1/2}$)
			Major	Trace		
A0	Hot-pressed	98.1	β-SiC	YAG*, α-SiC	575±69	3.5±0.4
A1	5	97.7	β-SiC	α-SiC, YAG	451±46	5.5±0.3
A2	10	97.4	α-SiC	β-SiC, YAG	401±57	4.6±0.4
B0	Hot-pressed	98.0	β-SiC	YAG, α-SiC	598±66	3.5±0.3
B1	5	97.6	α-SiC	β-SiC, YAG	465±56	5.8±0.4
B2	10	96.6	α-SiC	β-SiC, YAG	384±48	4.6±0.3
C0	Hot-pressed	97.6	β-SiC	YAG, α-SiC	604±59	3.5±0.3
C1	5	97.4	α-SiC	β-SiC, YAG	585±46	5.7±0.2
C2	10	96.7	α-SiC	β-SiC, YAG	390±52	4.5±0.4

* $Y_3Al_5O_{12}$ (yttrium aluminum garnet).

hot-pressing conditions (1850℃, 20 MPa) and the presence of much stacking faults in the whiskers (Fig. 1). The XRD results indicated that the small amount of $\beta \rightarrow \alpha$ phase transformation took place during hot-pressing (Table 3). The SiC whiskers with much stacking faults may have broken into several grains through the penetration of the liquid phase at the stacking fault boundaries during hot-pressing. Therefore, β-SiC whiskers does not act as nuclei for the grain growth of elongated large grains. Alpha-SiC whisker may be an effective seed for the microstructural control of self-reinforced SiC because of its stability at high temperatures (>1900 ℃). Unfortunately, α-SiC whiskers are not commercially available at present.

The effect of SiC whisker addition on the mechanical properties of hot-pressed and annealed materials is also shown in Table 3. The fracture toughness increased with annealing and showed the maximum at 5 h for all materials. The fracture toughness (5.7 MPa·m$^{1/2}$) of 5-h annealed material with 3 wt% β-SiC whiskers was almost the same with that (5.5 MPa·m$^{1/2}$) of equivalent material without β-SiC whiskers, as expected. The improved fracture toughness of annealed materials was attributed to the enhanced bridging and crack deflection by the elongated α-SiC grains, as shown in Fig. 3(b). Further annealing up to 10 h, however, decreased the fracture toughness slightly (~20 %), although the length and width of α-SiC grains were increased. This could be attributed to the increasing tendency of transgranular fracture with prolonged annealing, as shown in Fig. 3(c). The flex-

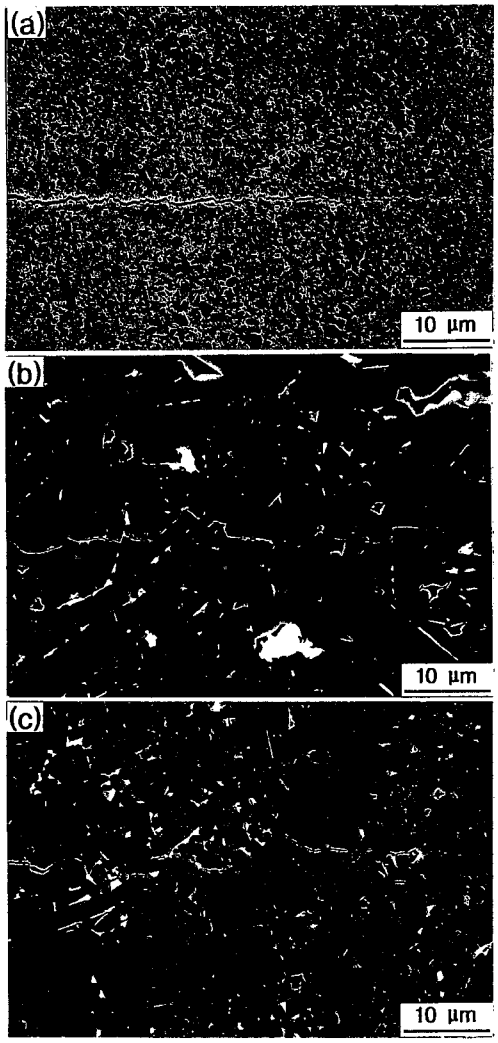

Fig. 3. SEM view of crack deflection by equiaxed grains in (a) B0 and crack bridging and deflection by elongated grains in (b) B1 and (c) B2 (refer to Table 2).

ural strength decreased with increasing annealing time for all materials. The strength (465 MPa) of 5-h annealed material with 1 wt% β-SiC whiskers was almost the same with that (451 MPa) of equivalent material without β-SiC whiskers, as expected. The decreased strength of annealed materials was attributed to both the increased tendency for relatively larger grains to fracture transgranularly and the pore growth with prolonged annealing [16].

4. Conclusions

The introduction of β-SiC whiskers into β-SiC does not affect the microstructure as well as mechanical properties significantly because the whiskers are not viable in the presence of liquid phase during hot-pressing. Alpha-SiC whisker may be an effective seed for the microstructural control of self-reinforced SiC because of its stability at high temperatures (>1900℃). The strengths and fracture toughnesses of hot-pressed and subsequently 5 h-annealed materials with 1 wt% β-SiC whiskers and without β-SiC whiskers were 465 MPa and 5.8 MPa·$m^{1/2}$, and 451 MPa and 5.5 MPa·$m^{1/2}$, respectively.

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