# Effect of Large α-Silicon Carbide Seed Grains on Microstructure and Fracture Toughness of Pressureless-Sintered α-Silicon Carbide

**Young-Wook Kim, Kyeong-Sik Cho\* and June-Gunn Lee\***

*Department of Materials Science and Engineering, Seoul City University,*
*90 Jeonnong-Dong, Dongdaemoon-Ku, Seoul 130-743, Korea*
*\*Division of Ceramics, Korea Institute of Science and Technology P.O. BOX 131,*
*Cheongryang, Seoul 130-650, Korea*
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α-SiC powder with or without the addition of 0.1 wt% of large α-SiC particles (seeds) was pressureless-sintered at 1950°C for 0.5, 2, and 4 h using $Y_3Al_5O_{12}$ (yttrium aluminum garnet, YAG) as a sintering aid The materials without seeds had an equiaxed grain structure. In contrast, the materials with seeds sintered for 2 and 4 h had a duplex microstructure with large elongated grains and small equiaxed grains. Addition of large α-SiC seeds into α-SiC accelerated the grain growth of some α-SiC grains during sintering and resulted in the increased fracture toughness of the sintered materials. The fracture toughnesses of materials with or without seeds sintered for 4 h were 6.6 and 5.2 MPa $m^{1/2}$, respectively.

***Key words :*** α-SiC, Microstructure, Fracture toughness, Large seed grain

## I. Introduction

O xide doped, liquid-phase sintered SiC containing elongated grains has high fracture toughness because of a bridging[1] or a crack deflection[2] toughening mechanism. The development of elongated grains has been achieved through the β→α phase transformation.[3-5] Hence, a raw powder of β-SiC and sintering or heat-treatment temperature as high as 2000°C were usually used for the phase tranformation and microstructural control.[3-5] Recently, in situ-toughened SiC with duplex microstructure has been fabricated at a temperature as low as 1850°C without appreciable β→α phase transformation.[6,7] This suggested that a small difference in the grain size gave a sufficient driving force for abnormal grain growth of some grains and a duplex microstructure might be developed without phase transformation as in β-Si₃N₄.[8-10]

Development of a duplex microstructure has not been reported when a raw powder of α-SiC is liquid-phase sintered with oxide additives. A wide particle size distribution in starting α-SiC powder may give enough driving force for abnormal grain growth of some α-SiC grains and result in a duplex microstructre as in β-SiC.[7] The wide particle size distribution of starting-powder has been obtained through the addition of large α-SiC seeds into a raw powder of α-SiC in this study. The objectives of this study were to develop in situ-toughened α-SiC with a duplex microstructure using α-SiC powder and to investigate the effect of addition of large α-SiC seeds on the microstructure

and fracture toughness of sintered α-SiC.

## II. Experimental Procedure

Commercially available fine α-SiC powder (designated as F, Showa Denko, Tokyo, Japan, grade A-1) and relatively coarse α-SiC powder (designated as C, Lonza-Werke Gmbh, Waldshut-Tiengen, Germany, grade UF-10) were used as starting powders. Powder C was added to act as seeds for grain growth. These powders mainly consisted of α-SiC and were similar in purity to common β-type raw powders, as shown in Table 1. To prepare a powder composition without seeds, 88 wt% powder F and 12 wt% $Y_3Al_5O_{12}$ (yttrium-aluminum garnet, YAG, 99.9% pure, High Purity Chemicals, Chiyoda Sakado, Japan) were ball milled in ethanol with SiC grinding balls for 24 h To prepare a powder composition containing seeds, 87 wt% powder F and 12 wt% $Y_3Al_5O_{12}$ were ball milled in ethanol for 22 h, and then 1 wt% powder C was added, followed by additional milling for 2 h. About 10% of powder C, particles larger than 2 μm, as shown in Table 1, are considered presumably to act as seeds for grain growth. Hence, 1 wt% addition of powder C corresponds to 0.1 wt% addition of large seed grains. The milled slurry was dried, uniaxially pressed at 20 MPa, and isostatically pressed at 140 MPa. Sintering was performed in a graphite resistance furnace. The specimens were sintered at a heating rate of 600°C/h and maintained at 1950°C for 0.5, 2, and 4 h in an atmosphere of flowing argon. A powder bed with the same composition as the specimens was used to

**Table 1.** Characteristics of Starting Powders

| Powder | F | C |
|---|---|---|
| Average particle size (μm) | 0.47 | 0.74 |
| Cumulative mass percent finer | | |
| 10 (wt%) | 0.22 μm | 0.30 μm |
| 50 (wt%) | 0.47 μm | 0.74 μm |
| 90 (wt%) | 1.10 μm | 2.10 μm |
| Specific surface area (m²/g)* | 18 | 10 |
| Impurities (wt%)* | | |
| Oxygen | 0.14 | 0.84 |
| Free C | 0.46 | 0.15 |
| Phase | α | α |

Data were supplied from manufacturers.

**Table 2.** Characteristics of Sintered α-SiC

| Speci-mens | Powder composition (wt%) | Sintering time at 1950°C (h) | Relative density (%) |
|---|---|---|---|
| A1 | 88 F-SiC+12 $Y_3Al_5O_{12}$ | 0.5 | 98.1 |
| A2 | 88 F-SiC+12 $Y_3Al_5O_{12}$ | 2 | 97.6 |
| A3 | 88 F-SiC+12 $Y_3Al_5O_{12}$ | 4 | 96.3 |
| B1 | 87 F-SiC+1 C-SiC+12 $Y_3Al_5O_{12}$ | 0.5 | 98.5 |
| B2 | 87 F-SiC+1 C-SiC+12 $Y_3Al_5O_{12}$ | 2 | 97.8 |
| B3 | 87 F-SiC+1 C-SiC+12 $Y_3Al_5O_{12}$ | 4 | 96.5 |

**Table 2.** Continued

| Speci-mens | Grain morphology | Average grain size (μm) Equiaxed | Average grain size (μm) Elongated* | Phase |
|---|---|---|---|---|
| A1 | Equiaxed | 0.69 | - | α |
| A2 | Equiaxed | 1.16 | - | α |
| A3 | Equiaxed | 1.57 | - | α |
| B1 | Equiaxed | 0.67 | - | α |
| B2 | Equiaxed and elongated | 0 74 | l=2.81 t=1.02 | α |
| B3 | Equiaxed and elongated | 1.33 | l=6.01 t=1.75 | α |

*l and t are length and thickness of elongated grains, respectively.

suppress the weight loss of the specimens.

The relative density was determined by the Archimedes method. X-ray diffraction using Cukα radiation was performed on all the ground specimens. The sintered specimens were cut and polished, and then plasma etched by $CF_4$ containing 7.8% $O_2$. The microstructures were observed by scanning electron microscopy (SEM). The grain size of equiaxed grains was directly determined from the average of the shortest and the longest grain diagonal. Grains having an aspect ratio (the longest grain diagonal/the shortest grain diagonal) of > 2 are defined as elongated grains in the present work. A total of 800 to 1100 grains per specimen was used for the statistical analysis. The length and thickness of elongated grains were determined from the longest grain diagonal and the shortest grain diagonal, respectively, in the two dimensional image. The fracture toughness was estimated by measuring crack lengths generated by a Vickers indenter with a load of 196 N.[11]

## III. Results and Discussion

The characteristics of α-SiC obtained are summarized in Table 2. The relative densities of > 98% were achieved by pressureless-sintering with a holding time of 0.5 h at 1950°C. The sintered densities were found to decrease with increasing sintering time. These results suggest that prolonged exposure to high temperatures may cause reactions between SiC and YAG and probably result in the formation of volatile components such as AlO, $Al_2O$, and CO,[12] which form isolated pores inside the specimen and make substantial weight loss, leading to the decrease in the sintered density.

Fig. 1 shows the microstructure of the α-SiC with or without seeds sintered at 1950°C for 0.5, 2, and 4 h. In the case of the α-SiC without seeds sintered for 0.5 h, the microstructure was composed of equiaxed grains and the polytype of the sintered body was 6H (α-SiC). When the holding time was increased, the grains grew without changing the shapes. In the case of the α-SiC with seeds sintered for 0.5 h, the microstructure was composed of equiaxed grains. When the holding time was increased, the shapes of some

of the grains changed from equiaxed to elongated and the grain size and aspect ratio increased without appreciable 6H→15R phase tranformation. Polytypes of all the sintered bodies were mainly 6H (> 90%) with trace of 15R. These results suggest that the addition of large seeds accelerated the grain growth of some α-SiC grains during sintering and resulted in the duplex microstructure. Large α-SiC seeds are suspected to act as nuclei for the grain growth of elongated large grains because the grain growth of SiC grains are controlled by Ostwald ripening,[13] i.e., solution of some α-SiC grains which are smaller than the critical size, mass transport by diffusion through the liquid, and reprecipitation on the other α-SiC grains which are larger than the critical size. The true shape of elongated grains is considered presumably as plate-shaped.

The microstructural development of Fig. 1 (f) is similar to the in situ formation of platelike or elongated grains during sintering of oxide-doped β-SiC.[3,5,7] Such kind of a duplex microstructure has been developed only with β-SiC starting powders or β-SiC starting powder containing α-SiC seeds. However, our results suggest that in situ toughened SiC with a duplex microstructure can be fabricated using α-SiC starting powders. It also indicates that a small difference in grain size gives a sufficient driving force for abnormal grain growth of some grains and a duplex microstructure can be developed without phase transformation as in $β-Si_3N_4$.[8-10]

Fig. 2 illustrates the fracture toughness of sintered samples versus holding time. It shows that fracture toughness increases with holding time. Fracture toughness for the sintered α-SiC without seeds increased slightly from 4.4 to
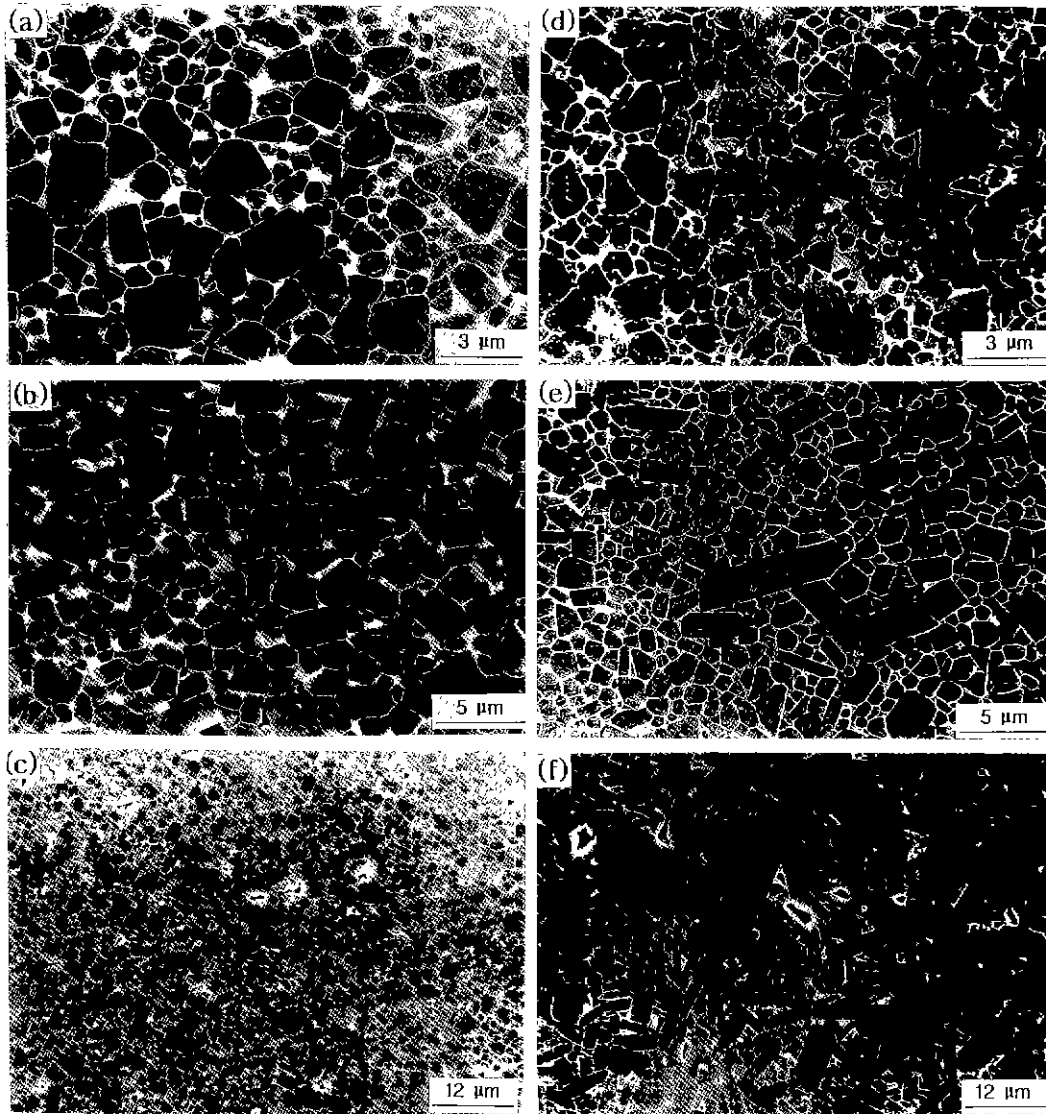
**Fig. 1.** SEM micrographs of α-SiC without seeds sintered with addition of 12 wt% Y₃Al₅O₁₂ at 1950°C for (a) 0.5, (b) 2, and (c) 4 h and α-SiC with seeds sintered with addition of 12 wt% Y₃Al₅O₁₂ at 1950°C for (d) 0 5, (e) 2, and (f) 4 h.
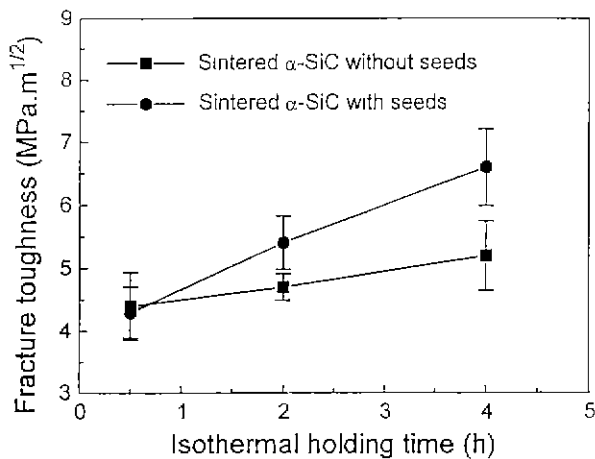


**Fig. 2.** Relation between fracture toughness of sintered α-SiC and holding time at 1950°C.

5.2 $MP \cdot m^{1/2}$ with holding time. This increase in fracture toughness was considered to be due to a grain-coarsening effect. The increase in the fracture toughness of noncubic ceramics, such as $Al_2O_3$, with the increase in grain size has been reported by several investigators.[14-17] Grain bridging or microcracking in the region around the crack tip has been suggested as the toughening mechanism of the grain size effect.[14-18] In the case of sintered α-SiC with seeds, fracture toughness increased significantly from 4.3 to 6.6 $MPa \cdot m^{1/2}$ with holding time. The increased fracture toughness was related to the microstructure, which was changed from small-equiaxed to large-elongated grains with increasing holding time. Several toughening mechanisms have been reported for the liquid phase sintered SiC containing elongated grains, including crack bridging[1] or crack deflection.[2] Bridging by elongated grains behind the crack tip and deflection by small equaxed grains were observed in the sintered α-
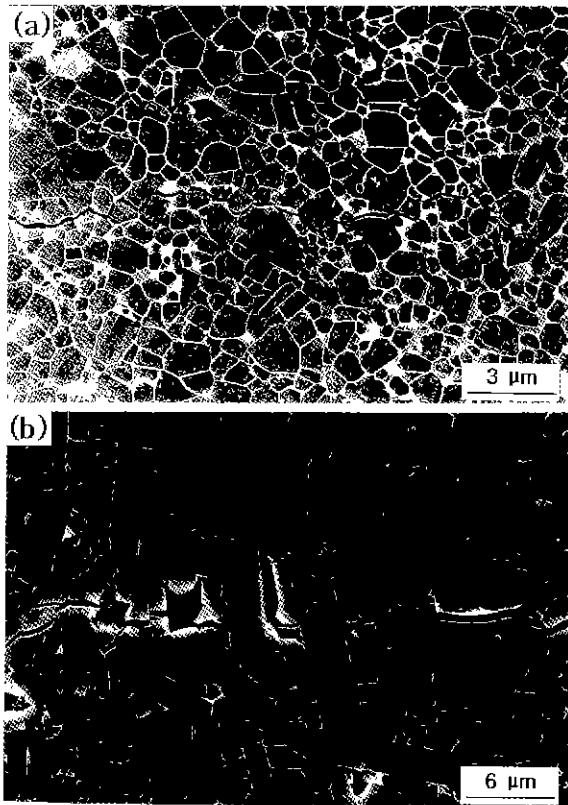
**Fig. 3.** SEM view of (a) crack deflection by equiaxed grains and (b) crack bridging by elongated grains in sintered α-SiC.

SiC (Fig. 3). Therefore, the improved fracture toughness of sintered α-SiC with seeds, comparing with the sintered α-SiC without seeds, was considered to be attributed to crack bridging by elongated grains.

## IV. Summary

In the present work, it was recognized that a duplex microstructure through in situ formation of elongated grains could be obtained with α-SiC starting powders containing large α-SiC seeds. The result of this study implies that a small difference in grain size gives a sufficient driving force for abnormal grain growth of some grains and in situ formation of duplex microstructure during sintering of SiC is possible without phase transformation as in β-$Si_3N_4$. The fracture toughnesses of materials with or without seeds sintered for 4 h were 6.6 and 5.2 MPa·$m^{1/2}$, respectively.

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