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LETTER

Effect of inert filler addition on microstructure and strength of porous SiC ceramics

Su-Ho Chae · Young-Wook Kim

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Porous ceramics are of technological interest because of their unique characteristics such as low density, low thermal conductivity, high thermal shock resistance, high permeability, and high specific strength [1–5]. Specifically, porous silicon carbide (SiC) ceramics are used as molten metal filters, diesel particulate filters, and preforms for metal matrix composites on account of their excellent permeability, thermal shock resistance, specific strength, and corrosion resistance at high temperatures [6–9]. Different processing routes for porous SiC ceramics have been developed for specific applications to satisfy the associated requirements of porosity, pore size, and degree of interconnectivity. These manufacturing techniques include replica techniques [10, 11], sacrificial template techniques [12, 13], and reaction techniques [8, 14]. Recently, a new processing method for fabricating porous SiC ceramics was developed based on the following strategy [15, 16]: (1) pyrolysis of carbon-filled polysiloxane at 1000 °C, which leads to the conversion of polysiloxane to silicon oxycarbide (SiOC); (2) carbothermal reduction of SiOC and C mixture at 1450 °C, which converts the mixture to SiC ceramic; and (3) liquid-phase sintering of SiC using Al_2O_3–Y_2O_3 as a sintering additive at 1750–1950 °C. This letter reports the effect of inert filler addition on the microstructural development and flexural strength of porous SiC ceramics processed by the above method using polymer microbeads as a pore former and submicron SiC powder as an inert filler.

The raw materials used in this experiment included: polysiloxane (GE Toshiba Silicones Co., Ltd, Tokyo, Japan), carbon black (Korea Carbon Black Co., Ltd., Inchon, Korea), SiC (FCP15C, Norton AS, Lillesand, Norway), poly (methyl methacrylate-*co*-ethylene glycol dimethacrylate) microbeads ($\sim$8 μm, Sigma–Aldrich Inc, St. Louis, MO, designated as PMMA), Al_2O_3 (AKP30, Sumitomo Chemical Co., Tokyo, Japan), Y_2O_3 (H.C. Starck GmbH & KG, Goslar, Germany), and MgO (High Purity Chemicals, Osaka, Japan). To prepare a powder composition without a filler (designated as PSC), 68.79% polysiloxane, 10.78% carbon, 3.10% Al_2O_3, 0.88% Y_2O_3, and 0.44% MgO were mixed with 16.01% PMMA microbeads. A powder composition with 20 wt% fillers (designated as PCSF) was prepared by mixing 59.79% polysiloxane, 9.37% carbon, 3.36% Al_2O_3, 0.96% Y_2O_3, and 0.48% MgO with 8.65% SiC and 17.39% PMMA microbeads. The batches were milled in ethanol for 24 h using SiC grinding balls. The milled powder was then dried and pressed uniaxially into rectangular bars at 28 MPa. The compacts formed were cross-linked by heating to 200 °C in air. The cross-linked samples were pyrolyzed at 1000 °C for 1 h in argon at a heating rate of 1 °C/min. The heat treatment allows the conversion of polysiloxane in the specimens to silicon oxycarbide [17]. The pyrolyzed specimens were further heat treated in argon at 1450 °C for 0.5 h at a heating rate of 10 °C/min and subsequently sintered at 1750–1950 °C for the liquid-phase sintering of SiC using Al_2O_3, Y_2O_3, and MgO. For the flexural strength measurements, bar-shaped samples were cut and polished to a size of $4 \times 5 \times 30$ mm^3. Bend tests were performed on five specimens for each condition at a crosshead speed of 0.5 mm/min using a four-point method with inner and outer spans of 10 and 20 mm, respectively.

S.-H. Chae · Y.-W. Kim (✉)
Department of Materials Science and Engineering,
The University of Seoul, 90 Jeonnong-dong, Dongdaemun-gu,
Seoul 130-743, Korea
e-mail: ywkim@uos.ac.kr

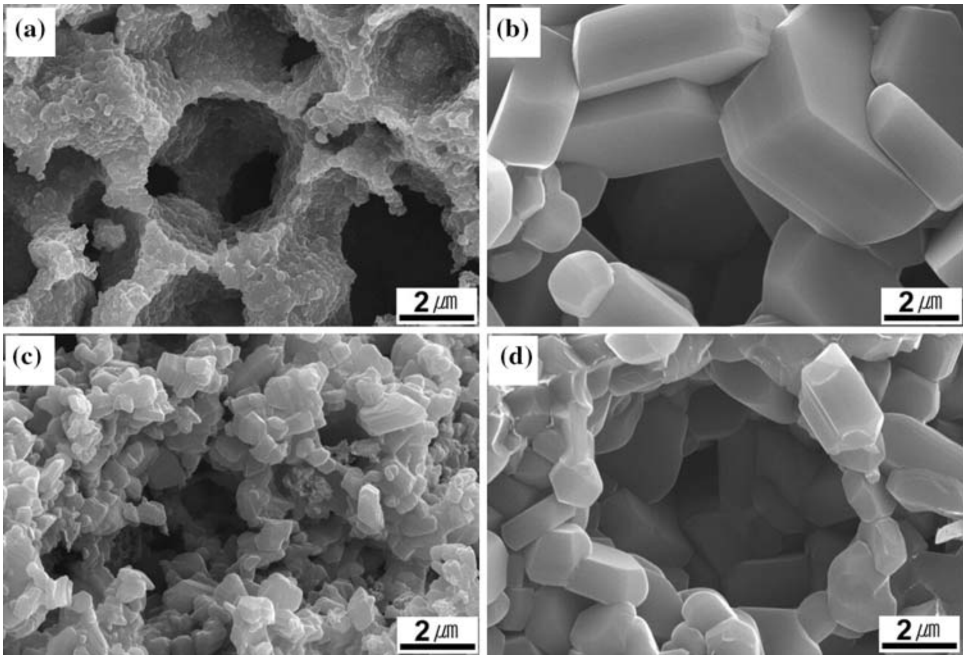

Fig. 1 Effect of the sintering temperature on the microstructure of the porous SiC ceramics. **a** PSC sintered at 1750 °C. **b** PSC sintered at 1950 °C. **c** PSCF sintered at 1750 °C. **d** PSCF sintered at 1950 °C

Figure 1 shows typical fracture surfaces of the porous SiC ceramics. The figures show well-distributed open cells with porous struts in the cellular structure. When sintered at 1750 °C for 1 h (Fig. 1a, c), the grain size (0.1–0.5 μm) of the PSC specimen was much finer than that (0.4–1.8 μm) of the PSCF specimen. The SiC grains in the PSC specimen were synthesized by a carbothermal reduction of polysiloxane-derived SiOC, which resulted in the formation of submicron grains. In contrast, the SiC grains observed in PSCF specimen were grown from the filler grains. The average grain size of the SiC filler was ~0.5 μm. Hence, the filler grain could be grown at the expense of the submicron SiC grains synthesized from the polysiloxane-derived SiOC through an Ostwald ripening mechanism.

When sintered at 1950 °C for 1 h, the morphology of the SiC grains changed from equiaxed to faceted grains in both specimens due to the $\beta \rightarrow \alpha$ phase transformation of SiC. However, the grain size (2–10 μm) of the PSC specimen was larger (1–4 μm) than that of the PSCF specimen. Rapid coarsening of SiC grains was observed in the 1950 °C-sintered PSC specimen, which was attributed to the higher driving force of finer SiC grains for grain growth [18]. In contrast, the finer microstructure obtained in the PSCF specimen was attributed to the impingement of growing grains, which were added as fillers. Therefore, the addition of SiC filler resulted in a larger grain size than the specimen without fillers when sintered at 1750 °C, whereas a smaller grain size was obtained when sintered at 1950 °C.

Figure 2 shows the porosities of the porous SiC ceramics as a function of the sintering temperature. The porosities of the PSC specimens ranged from 49 to 63% and those of the PSCF specimens ranged from 49 to 64%, depending on the sintering temperature. There were no significant differences in the porosity of both specimens. The porosity decreased with increasing temperature and showed a minimum at 1900 °C for both specimens. The porosity increased again when sintered at 1950 °C. The decrease in porosity with increasing sintering temperature from 1750 to 1900 °C was attributed to the further densification of porous SiC ceramics during sintering. The weight loss of the PSC and PSCF specimens increased from 72.6 to 75.1% and from 65.9 to 74.1%, respectively, as the sintering temperature was increased from 1900 to

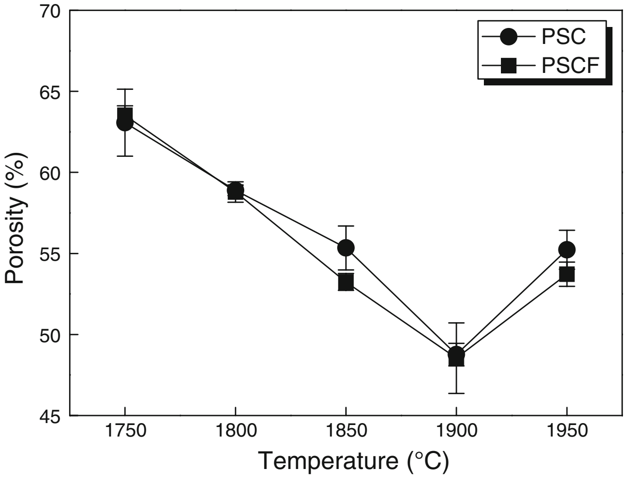

Fig. 2 Porosity as a function of the sintering temperature for porous SiC ceramics with and without fillers

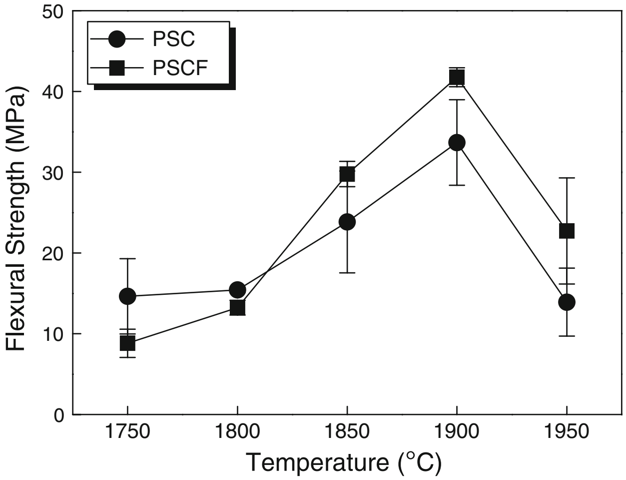

Fig. 3 Flexural strength as a function of the sintering temperature for porous SiC ceramics with and without fillers

1950 °C. Therefore, the increase in porosity with increasing annealing temperature from 1900 to 1950 °C was mainly due to the increased weight loss during annealing. Volatilization of the liquid phase and chemical reactions between SiC and the oxide additives are possible reasons for the weight loss [19–21]. The PSC and PCSF specimens showed 40.6 and 35.6% linear shrinkage during sintering at 1900 °C, respectively. Thus, the addition of SiC fillers resulted in less weight loss and less shrinkage, which would be beneficial for dimensional control.

Figure 3 shows the flexural strengths as a function of the sintering temperature for the porous SiC ceramics. The flexural strength of both specimens increased with increasing temperature up to 1900 °C and showed a maximum at 1900 °C, followed by a decrease in strength when sintered at 1950 °C. This tendency was in contrast to the change in porosity with sintering temperature (Fig. 2). Therefore, the flexural strength of the porous SiC ceramics is controlled mainly by the porosity. However, the PSCF specimen showed higher strength (~42 MPa) than PSC (~34 MPa) when sintered at 1900 °C. The porosities of the PSC and PSCF specimens were 48.8 and 48.6%, respectively. Thus, the difference in strength is due to microstructural differences in the specimens. The PSC specimen consisted of larger faceted grains than the PSCF specimen when sintered at 1900 and 1950 °C. The rapid coarsening of SiC grains resulted in decreased strength.

In conclusion, the addition of SiC fillers leads to a finer microstructure when sintered at 1900 and 1950 °C, resulting in higher strength at an equivalent porosity than the specimens without fillers. The finer microstructure obtained in the specimen with fillers was attributed to the impingement of growing grains, which were added as fillers. The addition of SiC fillers resulted in less weight loss and less shrinkage. Therefore, the addition of inert fillers is beneficial in improving the strength and dimensional control during the processing of porous SiC ceramics by a carbothermal reduction and subsequent sintering process.

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