



Thermal, electrical, and mechanical properties of pressureless sintered silicon carbide ceramics with yttria-scandia-aluminum nitride



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ABSTRACT

The effects of the polytype of SiC starting powders on the thermal, electrical, and mechanical properties of pressureless sintered SiC ceramics with a new additive system (6.5 vol% Y_2O_3 - Sc_2O_3 -AlN) were investigated. Powder mixtures prepared from α - or β -SiC powders were sintered at 1950 °C for 6 h in a nitrogen atmosphere without an applied pressure. We found that both specimens could be sintered to >96% of the theoretical density without an applied pressure. The SiC ceramic fabricated from β -SiC powders showed lower electrical resistivity, higher thermal conductivity, and better mechanical properties than that from α -SiC powders. The flexural strength, fracture toughness, hardness, electrical resistivity and thermal conductivity values of the SiC ceramics fabricated from β -SiC powders were 520 MPa, 5.1 MPa m^{1/2}, 25.0 GPa, $6.7 \times 10^{-1} \Omega \text{ cm}$ and $110 \text{ Wm}^{-1} \text{ K}^{-1}$ at room temperature, respectively. The new additive system achieved the highest thermal conductivity in pressureless liquid-phase sintered SiC ceramics.

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1. Introduction

Silicon carbide (SiC) is an important engineering ceramic because of its good thermal conductivity, oxidation resistance, wear resistance, corrosion resistance, and high-temperature mechanical properties [1–8]. SiC is being used in many applications, especially in heaters, heater plates, susceptors, dummy wafers, focus rings for semiconductors and in light-emitting diode (LED) processing. These applications take advantage of its excellent thermal conductivity as well as other properties.

Heat is carried by phonons in SiC ceramics because of the deficiency of free electrons. Although the thermal conductivity values of single crystalline SiC ceramics are very high (347 for 4H-SiC and $490 \text{ Wm}^{-1} \text{ K}^{-1}$ for 6H-SiC) [9,10], those of polycrystalline SiC ceramics are quite low because of the scattering of phonons by point defects, phase boundaries, grain boundaries, and secondary phases [8,11,12]. Several strategies for selecting additives for improving thermal conductivity of polycrystalline liquid-phase sintered SiC (LPS-SiC) ceramics were suggested including the following [8,11–15]:

- (1) Use of sintering additives that can pick up oxygen from the SiC lattice because the lattice oxygen can cause additional Si vacancies according to the following defect equation, $\text{SiO}_2 \rightarrow \text{Si}_i + 2\text{O}_c + \text{V}_{\text{Si}}$. These vacancies result in phonon scattering.
- (2) Use of sintering additives without Al and Al compounds because the Al can create additional Si vacancies according to the following defect equation, $\text{Al}_2\text{O}_3 \rightarrow 2\text{Al}_{\text{Si}} + 3\text{O}_c + \text{V}_{\text{Si}}$. These vacancies can also cause phonon scattering.
- (3) Minimizing oxide additive content because the thermal conductivity of oxide phases is much lower than that of the SiC lattice, resulting in lower thermal conductivity.
- (4) Use of additives that can suppress the $\beta \rightarrow \alpha$ phase transformation of SiC because the phase transformation can lead to increased phonon scattering at the 3C/4H and 3C/6H interfaces in SiC grains.

Examples of additive compositions which can satisfy the above requirements include Y_2O_3 - La_2O_3 , Y_2O_3 -TiN, and Y_2O_3 - Sc_2O_3 systems. The maximum thermal conductivity values achieved in each system were $211 \text{ Wm}^{-1} \text{ K}^{-1}$ in SiC ceramics sintered with 5 vol% Y_2O_3 - La_2O_3 [11], $211 \text{ Wm}^{-1} \text{ K}^{-1}$ in SiC ceramics sintered with 3 vol% Y_2O_3 -TiN [14] and $234 \text{ Wm}^{-1} \text{ K}^{-1}$ in SiC ceramics sintered with 1 vol% Y_2O_3 - Sc_2O_3 [8]. However, all of the above LPS-SiC ceramics were processed with an applied pressure, i.e., via a hot-pressing route. The thermal conductivity values of LPS-SiC

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ceramics processed without an applied pressure were in the range of 55–90 Wm⁻¹ K⁻¹. Specifically, the maximum thermal conductivity values achieved in each system without an applied pressure were 70 Wm⁻¹ K⁻¹ for SiC ceramics sintered with 10 vol% Al₂O₃-Y₂O₃ [16], 85 Wm⁻¹ K⁻¹ for SiC ceramics sintered with 3 vol% Y₃Al₅O₁₂-AlN [17], 85 Wm⁻¹ K⁻¹ for SiC ceramics sintered with 7 wt% Al₂O₃-Er₂O₃ [18], and 90 Wm⁻¹ K⁻¹ in SiC ceramics sintered with 7 wt% Al₂O₃-CeO₂ [18].

Generally, solid-state sintered SiC (SSS-SiC) ceramics with boron (B) or B-containing additives showed higher thermal conductivity than LPS-SiC ceramics processed without an applied pressure. The thermal conductivity values were 100 Wm⁻¹ K⁻¹ for SSS-SiC ceramics with 2 vol% BN [19], 140 Wm⁻¹ K⁻¹ for SiC ceramics with 2.1 wt% B₄C-C [20], 124 Wm⁻¹ K⁻¹ for SiC ceramics with 2.6 wt% AlN-B₄C-C [21], and 192 Wm⁻¹ K⁻¹ for SiC ceramics with 0.8 wt% B-C additives [22]. However, the SSS-SiC ceramics have a few drawbacks compared to LPS-SiC, such as poor mechanical properties and the possibility of boron contamination during semiconductor and LED processing. Thus, LPS-SiC ceramics are preferred for applications in semiconductor and LED processing.

Industrial demands for applications of LPS-SiC parts in semiconductor and LED processing, e.g., susceptors, trays, and wafer boats, require the development of SiC ceramics with a higher thermal conductivity and lower electrical resistivity than the current LPS-SiC ceramics processed without an applied pressure, i.e., via a pressureless sintering route. This is because industry requires more rapid heating and cooling schedules during production.

Most previous researches on pressureless LPS-SiC ceramics have focused on the Al₂O₃-Y₂O₃ additive system [23–25], although a few other additive compositions have also been investigated for pressureless LPS-SiC. Those additive compositions include Y₃Al₅O₁₂-AlN [17] and Al₂O₃-RE₂O₃ (Re = Ce, Er, Lu) [18]. The addition of a third oxide, in addition to Al₂O₃-Y₂O₃, was also investigated for lowering the sintering temperature and/or for improving the fracture toughness of LPS-SiC ceramics. Those additive compositions include Al₂O₃-Y₂O₃-MgO [26], Al₂O₃-Y₂O₃-TiO₂ [27], and Al₂O₃-Y₂O₃-CaO [28] systems. The flexural strength and fracture toughness of the SiC ceramics sintered with 10 wt% Al₂O₃-Y₂O₃-MgO were 377–440 MPa and 4.8–5.2 MPa m^{1/2}, respectively [26]. Those values of the SiC ceramics sintered with 10 wt% Al₂O₃-Y₂O₃-TiO₂ were 516 MPa and 4.6 MPa m^{1/2}, respectively [27]. The fracture toughness and hardness of the SiC ceramics sintered with 10 wt% Al₂O₃-Y₂O₃-CaO were 5.7 MPa m^{1/2} and 23.2 GPa, respectively [28]. However, no thermal conductivity data were reported for the pressureless sintered SiC ceramics with the above ternary additive systems.

Based on the above review, we suggest several strategies that may improve the thermal conductivity of pressureless LPS-SiC ceramics: (1) sintering in a nitrogen atmosphere is preferred for suppressing the β→α phase transformation of SiC; (2) the addition of Al-containing additives should be avoided or minimized for minimizing V_{Si} concentration in SiC lattice; and (3) if an Al-containing additive is added for successful densification, AlN is preferred over Al₂O₃ for minimizing oxygen contamination in SiC lattice.

To obtain LPS-SiC ceramics with higher thermal conductivity than the reported values (55–90 Wm⁻¹ K⁻¹), a SiC ceramic with 5 vol% equimolar Y₂O₃-Sc₂O₃ additives was fabricated by pressureless-sintering. However, the relative density of the pressureless-sintered SiC ceramic with Y₂O₃-Sc₂O₃ was only 88%. In this study, to densify the SiC ceramic containing Y₂O₃-Sc₂O₃ additives without an applied pressure, a small amount of AlN (1.5 vol%) was introduced as an additional sintering additive in addition to the 5 vol% Y₂O₃-Sc₂O₃. Previous works on LPS-SiC suggested that the crystallographic modification of the initial SiC powder has a strong influence on the microstructure and properties of the resulting ceramics [23,29]. The objective of this research

is to investigate the effects of polytype of SiC starting powders on the sintered density, crystalline phase, and microstructure of the pressureless sintered SiC ceramics with a new additive system (Y₂O₃-Sc₂O₃-AlN). The thermal, electrical, and mechanical properties of the ceramics were measured, and the results were correlated with the sintered density, crystalline phase, and microstructure of the ceramics.

2. Experimental procedure

Commercially available α-SiC (~0.5 μm, UF-15, H.C. Starck, Berlin, Germany), β-SiC (~0.5 μm, BF-17, H.C. Starck, Berlin, Germany), Y₂O₃ (0.4 μm, 99.99% pure, Kojundo Chemical Lab Co., Ltd., Sakado-shi, Japan), Sc₂O₃ (~9 μm, 99.99% pure, Kojundo Chemical Lab Co., Ltd., Sakado-shi, Japan), and AlN (~1.5 μm, Grade F, Tokuyama Soda Co., Ltd., Sakado-shi, Japan) were used as the starting materials. Two batches of powder mixtures were mixed in ethanol for 24 h using SiC balls and a polypropylene jar (Table 1). The ethanol used contains 1 wt% poly(ethylene glycol) as a binder. The additive content was fixed at 6.5 vol% and consisted of 1.5 vol% AlN and 5 vol% equimolar Y₂O₃-Sc₂O₃. The mixtures were dried, sieved (60 mesh), and uniaxially pressed under an applied pressure of 25 MPa. The green bodies (40 × 40 × 5 mm) were subsequently cold isostatically pressed at 280 MPa. Burning out prior to pressureless sintering was done at 450 °C for 30 min with a heating rate of 2 °C/min in a nitrogen atmosphere. The pressureless sintering of the samples (three samples at each condition) was carried out in a graphite furnace at 1850–2000 °C for 6 h in a nitrogen atmosphere. All the tested specimens in this investigation were cut from the as-sintered specimens.

The relative densities of the pressureless-sintered specimens were determined using the Archimedes method. Theoretical densities of the specimens were calculated according to the rule of mixtures, as shown in Table 1. X-ray diffraction (XRD; D8 Discover, Bruker AXS GmbH, Karlsruhe, Germany) using Cu Kα radiation was performed on the ground powders. The XRD data were analyzed using the Rietveld refinement method for quantitative phase analysis of SiC polytypes. The sintered specimens were polished and etched with CF₄ plasma containing 10% oxygen. The etched microstructure and fracture surface were observed by scanning electron microscopy (SEM, S4300, Hitachi Ltd., Hitachi, Japan). Mean grain sizes were measured by the line intercept method, where the multiplication constant was 1.56 based on a tetrakaidecahedral grain shape [30]. Seven hundred to nine hundred grains were measured for each specimen to obtain a mean grain size.

The thermal diffusivity was measured using the laser flash method. Differential scanning calorimetry (DSC, Model Q200; TA instrument Inc., New Castle, DE, USA) and thermal diffusivity measurement equipment (Model LFA 447; NET-ZSCH GmbH, Selb, Germany) were used for measuring the heat capacity (C_p) and thermal diffusivity (α), respectively. Samples (2.84 mm × 2.84 mm × 1 mm for measuring the heat capacity and 10 mm × 10 mm × 4 mm for measuring the thermal diffusivity) were cut from the pressureless sintered specimens and were polished. The C_p and α were measured five times each, and the average values were used to calculate the thermal conductivity at 25 °C. The thermal conductivity (κ) was calculated according to the following equation [31],

$$\kappa = \alpha \rho C_p \quad (1)$$

where ρ is the density of the sample. The average phonon mean free path (λ) was calculated according to the equation [15],

$$\lambda = 3\kappa / C_p \nu \quad (2)$$

where ν is the average velocity of sound in the SiC ceramic. The velocity of sound in SiC is 11820 m/s at room temperature [32]. The

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