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High-temperature strength of a thermally conductive silicon carbide ceramic sintered with yttria and scandia

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ABSTRACT

The high-temperature strength of a thermally conductive SiC ceramic sintered with 1 vol% equimolar Y_2O_3–Sc_2O_3 additives (thermal conductivity = 234 W (m K)$^{-1}$) was investigated at temperatures up to 1800 °C. Observation of the ceramic using high resolution transmission electron microscopy (HRTEM) exhibited both clean and crystallized SiC/SiC boundaries, as well as clean SiC/junction phase boundaries with a fully crystallized junction phase. No microstructural or polytype changes after flexural testing at 1800 °C were observed using scanning electron microscopy and phase analysis with the Rietveld method. The ceramic maintained 93% of its room temperature (RT) strength up to 1600 °C, and showed rapid degradation at 1700 °C and 1800 °C. Degradation at temperatures above 1600 °C was due to softening of the grain boundary phase, as evidenced by the nonlinear behavior of load-displacement curves. Flexural strengths of the highly thermally conductive SiC ceramic at RT and 1800 °C were 536 MPa and 345 MPa, respectively.

1. Introduction

Silicon carbide (SiC) is an important material due to its excellent thermal conductivity, wear resistance, oxidation resistance, and high-temperature mechanical properties [1–10]. SiC ceramics have been used for several industrial applications. Capsule materials for nuclear fuel, burner nozzles, rocket nozzles, heater and heater plates, and other high-temperature applications all take advantage of its excellent high-temperature mechanical properties as well as its excellent thermal conductivity.

Thermal conductivity of liquid-phase sintered SiC (LPS-SiC) ceramics has been intensively investigated over the past three decades [11–16]. Polycrystalline SiC ceramics demonstrate a wide range of thermal conductivity values, from 30 W (m K)$^{-1}$ to 270 W (m K)$^{-1}$, depending on the specific chemistry of sintering additives and post-heat treatment conditions. For example, hot-pressed SiC sintered with BeO yielded a thermal conductivity of 270 W (m K)$^{-1}$ [11]. A SiC ceramic sintered with Al_2O_3–Y_2O_3 had a conductivity of 55–90 W (m K)$^{-1}$ [12,13], whereas a SiC ceramic sintered with Al_2O_3–C had a conductivity of 30–45 W (m K)$^{-1}$ [14]. Hot-pressed SiC ceramic sintered with Al_2O_3–Y_2O_3–CaO additives at 1750 °C for 40 min at a pressure of 25 MPa yielded a conductivity of 32 W (m K)$^{-1}$. Post annealing of the hot-pressed SiC ceramic at 1850 °C for 4 h increased its thermal conductivity to 106 W (m K)$^{-1}$ [15]. Hot-pressed SiC ceramic with 5 vol% Y_2O_3–La_2O_3 had a thermal conductivity of 167 W (m K)$^{-1}$ [16]. When the SiC ceramic was further annealed at 2000 °C for 4 h in an argon atmosphere, the thermal conductivity increased from 167 W (m K)$^{-1}$ to 211 W (m K)$^{-1}$. Unfortunately, mechanical properties of the above ceramics were not reported simultaneously with thermal conductivity data.

The flexural strength of SiC ceramics sintered with various additives has also been studied at high temperatures [2,17–26]. SiC ceramics sintered with Al_2O_3–Y_2O_3 exhibited rapid degradation in flexural strength at temperatures above 1300 °C, with strength values of 200–300 MPa at 1300 °C [2,17]. A SiC ceramic with 5.6 wt% Al–B–C also demonstrated severe degradation in flexural strength at high temperatures, with a strength of ~100 MPa at 1300 °C [18]. Higher strength values were obtained in SiC ceramics sintered with AlN–RE_2O_3 (RE = Lu, Yb, Er, Y): ~400 MPa at 1400 °C in SiC with 10 vol% AlN–Y_2O_3 [19], ~500 MPa at 1400 °C in SiC with 10 vol% AlN–Yb_2O_3 [20], ~500 MPa at 1500 °C in SiC with 10 vol% equimolar AlN–Lu_2O_3 [21], and ~550 MPa at 1600 °C in SiC with 10 vol% AlN–Er_2O_3 in a 3:2 molar ratio [22]. Improved strength values were obtained in SiC ceramics sintered with 10 vol% AlN–Lu_2O_3 in a 2:3 molar ratio (~600 MPa at 1600 °C) [23] and SiC ceramics with 10 vol% AlN–Sc_2O_3 in a 1:4 molar ratio (~620 MPa at 1600 °C)

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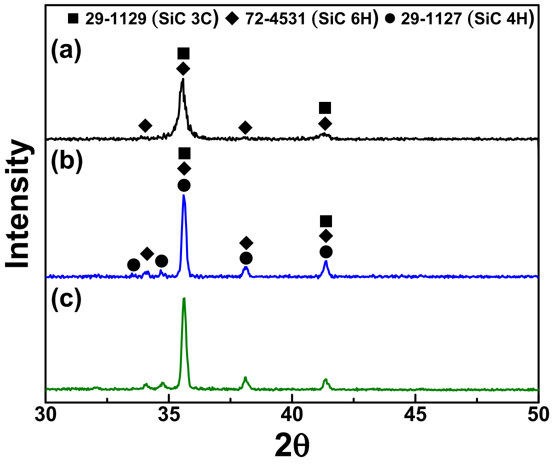

Fig. 1. X-ray diffraction patterns: (a) starting SiC powder, (b) as-sintered SiC ceramic with 1 vol% Y_2O_3–Sc_2O_3 additives, and (c) SiC ceramic tested at 1800 °C.

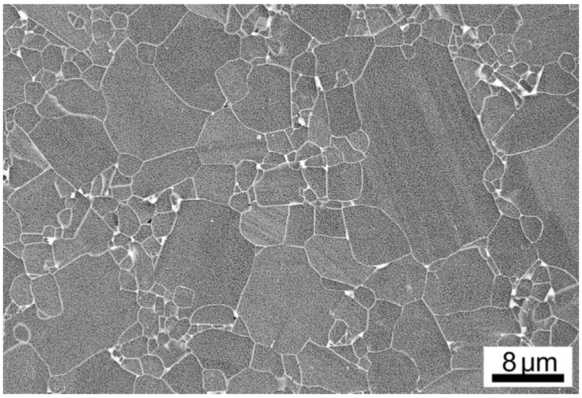

Fig. 2. Typical microstructure of a highly thermally conductive SiC ceramic.

[24]. However, the additive content in the above investigations was greater than 5 wt%. High-temperature strength data of SiC ceramics with a small amount of additives (<2 wt%) was quite limited. Lim et al. [25] reported 100% retention of room temperature strength (~630 MPa) at 1600 °C in SiC sintered with 1 wt% AlN–Lu_2O_3 in a 3:2 molar ratio. All of the above SiC ceramics contained Al or Al-compounds as sintering additives. We were unable to find any reports regarding high-temperature strength of SiC ceramics sintered without Al or Al-compounds.

Recently, a highly thermally conductive LPS-SiC ceramic with a thermal conductivity of 234 W $(m K)^{-1}$ was successfully fabricated by hot-pressing a SiC powder mixture containing Y_2O_3–Sc_2O_3 as sintering additives [26]. The high thermal conductivity was attributed to (i) the reduction of oxygen content in SiC surface and/or lattice by forming $(Sc,Y)_2Si_2O_7$ phase, (ii) the lack of solubility of Y and Sc in SiC lattice, (iii) forming clean or crystallized SiC–SiC boundaries, and (iv) growth of nitrogen-doped SiC grains. In semiconductors, the conduction electrons can also contribute to thermal conduction as well as phonons [27]. In the present study, the highly thermally conductive SiC ceramic with 1 vol% (1.37 wt%) Y_2O_3–Sc_2O_3 was fabricated using a hot-pressing technique. Its grain boundary structure was characterized using high resolution transmission electron microscopy (HRTEM), and the high-temperature strength of the ceramic was examined at temperatures up to 1800 °C in a nitrogen atmosphere.

2. Experimental procedure

To prepare SiC with 1 vol% Y_2O_3–Sc_2O_3 additives (Y_2O_3:Sc_2O_3 = 1:1 in molar ratio), 98.63 wt% β-SiC (~0.5 μm, Grade BF-17, H. C. Starck, Berlin, Germany), 0.85 wt% Y_2O_3 (99.99% pure, Kojundo Chemical Lab Co., Ltd., Sakado-shi, Japan), and 0.52 wt% Sc_2O_3 (99.99% pure, Kojundo Chemical Lab Co., Ltd.) were mixed by ball milling using SiC media in a plastic jar for 24 h in ethanol. The mixture was dried, sieved, and hot pressed at 2050 °C for 6 h under an applied pressure of 40 MPa in a nitrogen atmosphere.

The relative density of the hot-pressed specimen was determined using the Archimedes method. The theoretical density of the specimen was calculated according to the rule of mixtures. The hot-pressed specimen was cut, polished and etched with CF_4 plasma containing 10% oxygen. The etched microstructure and fracture surface morphology were observed using scanning electron microscopy (SEM, S4300, Hitachi Ltd., Hitachi, Japan). SiC disks (3 mm in diameter) were prepared for transmission electron microscopy (TEM) using a standard method that included slicing, ultrasonic drilling, polishing, and ion milling. HRTEM (400 kV, JEM-4010, JEOL, Tokyo, Japan) was used to observe SiC–SiC phase boundaries and SiC-junction phase boundaries. X-ray diffraction (XRD) using Cu Kα radiation was performed on a starting SiC powder and ground powders for the hot-pressed specimen and the specimen tested at 1800 °C. XRD data were analyzed using the Rietveld refinement method for quantitative phase analysis of SiC polytypes.

Bar-shaped specimens were ground to a size of 2 × 1.5 × 25 mm for strength measurements. The tensile surface of the bars was polished to a 1 μm diamond finish, and edges were beveled to avoid stress concentrations and edge flaws caused by sectioning. Bend tests were performed in a nitrogen atmosphere at temperatures between room temperature (RT) and 1800 °C on five to seven specimens for each condition using a four-point method with inner and outer spans of 10 and 20 mm, respectively. Tests at high temperatures were performed by heating specimens (in a manner that limited the potential for thermal shock) with a dwell time of 10 min in order to ensure homogeneous temperature during testing. Specimens were loaded at a constant crosshead speed of 0.2 mm/min. The hardness of the polished specimens was measured by Vickers indentation (Model AVK-C2, Akashi Corp., Yokohama, Japan) under a load of 9.8 N with a dwell time of 15 s.

3. Results and discussion

The relative density of the hot-pressed specimen was 99.9%. This result suggests that a small amount (1 vol%) of Y_2O_3–Sc_2O_3 was sufficient to densify SiC to a relative density greater than 99% by conventional hot-pressing at 2050 °C for 6 h under 40 MPa in a nitrogen atmosphere. XRD analyses of the starting SiC powder, the sintered specimen, and the specimen tested at 1800 °C are shown in Fig. 1. Quantitative phase analyses of SiC polytypes (using the Rietveld refinement method) showed that: (1) the starting SiC powder consisted of 87% 3C (β-SiC) and 13% 6H (α-SiC) phases; (2) the sintered specimen consisted of 60% 3C, 26% 6H, and 14% 4H (α-SiC) phases; and (3) the specimen tested at 1800 °C consisted of 60% 3C, 25% 6H, and 15% 4H (α-SiC) phases. The results suggest that both the 3C → 6H and the 6H → 4H phase transformations for SiC occurred during sintering at 2050 °C. However, no meaningful transformations took place during flexural strength testing at 1800 °C.

The specimen sintered with 1 vol% Y_2O_3–Sc_2O_3 consisted of equiaxed large grains (size range 10–50 μm) and equiaxed small grains (size range 1–7 μm) (Fig. 2). The equiaxed morphology of SiC grains indicates that the β → α phase transformation of SiC was never completed. It has been well documented that the β → α phase