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Microstructure and high-temperature strength of silicon carbide with 2000 ppm yttria

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ABSTRACT

A dense silicon carbide (SiC) ceramic with a very high flexural strength at 2000 °C (981 ± 128 MPa) was obtained by conventional hot-pressing with extremely low additive content (2000 ppm Y_2O_3). Observations using high-resolution transmission electron microscopy (HRTEM) showed that (1) homophase (SiC/SiC) boundaries were clean without an intergranular glassy phase and (2) junction pockets consisted of nanocrystalline Y-containing phase embedded in an amorphous Y-Si-O-C-N phase. The excellent strength at 2000 °C was attributed to the clean SiC/SiC boundary and the strengthening effect of plastic deformation.

1. Introduction

Silicon carbide (SiC) ceramics are important high-temperature structural materials because of their unique combination of superior properties, including their high thermal conductivity, excellent mechanical properties, and excellent resistance to wear and oxidation at high temperatures [1–9]. Most SiC ceramics are densified via liquid phase sintering using metal oxides as sintering additives because of their superior fracture toughness of liquid-phase-sintered SiC (LPS-SiC) ceramics compared to solid-state-sintered SiC ceramics. During liquid phase sintering, the metal oxide additives form a melt, reacting with SiO_2 and SiC at high temperatures and forming an amorphous intergranular film (IGF) at the SiC/SiC boundaries as well as an amorphous or crystalline junction phase. Nearly all LPS-SiC ceramics reported thus far have an IGF with a characteristic thickness on the order of 0.4–2.0 nm [1,3,10–14]. However, the thermally weak IGF leads to the degradation of flexural strength at high temperatures. Thus, the IGF of LPS-SiC is a major weakness in high temperature applications.

To overcome this obstacle for high temperature applications, several strategies have been investigated to improve high temperature properties: (1) crystallization of the IGF by post-sintering heat treatment [1], (2) removal of the IGF by post-sintering heat treatment [15], and (3) judicious selection of sintering additive compositions, leading to a highly refractory IGF [16–19].

To improve the high temperature strength of LPS-SiC ceramics, many combinations of additives, such as combinations of Al_2O_3, Y_2O_3, CaO, MgO, or AlN with an RE_2O_3 (RE = Sc, Nd, Sm, Gd, Dy, Ho, Er, Yb, and Lu), have been investigated [11,15–24]. Among those tested, the combinations of AlN-RE_2O_3 and Y_2O_3-RE_2O_3 were found to be the most effective in improving the high temperature properties of SiC ceramics. Strength values reported in SiC ceramics sintered with AlN-RE_2O_3 additives are as follows: 400 MPa at 1400 °C in SiC ceramics sintered with 10 vol% AlN-Y_2O_3 [17]; 550 MPa at 1600 °C in SiC ceramics sintered with 10 vol% AlN-Er_2O_3 [11]; 500 MPa at 1400 °C [19] and 550 MPa at 1500 °C [18] in SiC ceramics sintered with 10 vol% AlN-Yb_2O_3; 620 MPa at 1600 °C in SiC ceramics sintered with 10 vol% AlN-Sc_2O_3 [20]; 500 MPa at 1500 °C [24] and 600 MPa at 1600 °C [16] in SiC ceramics sintered with 10 vol% AlN-Lu_2O_3 and 633 MPa at 1600 °C in SiC ceramics sintered with 1 wt% AlN-Lu_2O_3 [22]. In contrast, limited data are available for SiC ceramics sintered with Y_2O_3-RE_2O_3. Seo et al. [23] reported flexural strength values of 501 MPa at 1600 °C and 345 MPa at 1800 °C in SiC ceramics sintered with 1 vol% Y_2O_3-Sc_2O_3.

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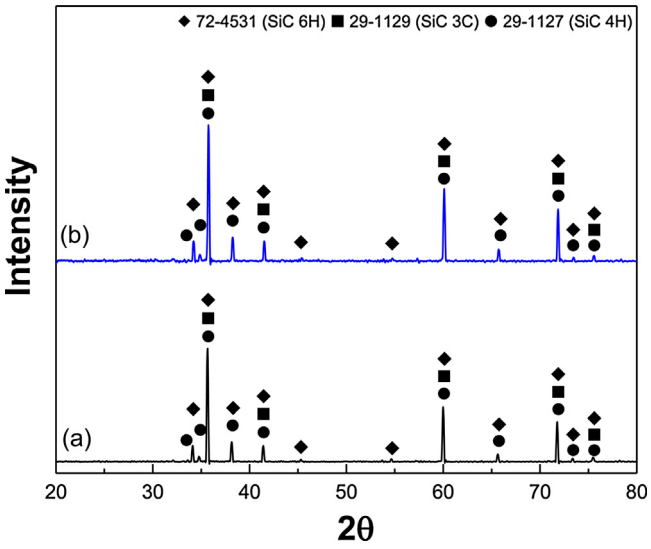

Fig. 1. X-ray diffraction patterns: (a) as-sintered SiC ceramic with 2000 ppm Y_2O_3 and (b) SiC ceramic tested at 2000 °C.

Most LPS-SiC ceramics are sintered with binary or ternary additives. However, the addition of a single additive is beneficial in terms of high temperature properties because there will be no melting point depression, in contrast with binary or ternary additive systems. Kim et al. [25] and Noviyanto et al. [26] reported successful densification of SiC ceramics with 4.95 wt% Y_2O_3 and 5 wt% Sc_2O_3, respectively, by conventional hot-pressing. Both additives were added in the form of nitrates. Kim et al. [27] also reported a successful densification of SiC with 6.29 wt% Y_2O_3 by conventional hot-pressing. One approach for improving high temperature properties of LPS-SiC ceramics is to reduce overall sintering additive content. Previous efforts to reduce the additive content have been focused on Y_2O_3 additive in SiC ceramics. Hot pressing submicron β-SiC powders containing 2.98 wt% Y_2O_3 at 2000 °C for 6 h under 40 MPa in a nitrogen atmosphere reached 98.9% of the theoretical density [28], and 81.7% of the theoretical density was obtained by spark plasma sintering submicron β-SiC powders containing 1.0 wt% Y_2O_3 at 1950 °C for 10 min under 60 MPa in a nitrogen atmosphere [29]. Almost fully densified SiC ceramics (99.9% of the theoretical density) have been achieved by hot-pressing submicron β-SiC powders containing 0.2 wt% Y_2O_3 at 2200 °C for 4 h under an applied pressure of 40 MPa in a nitrogen atmosphere [30]. However, the high temperature strength of the SiC ceramics with low additive content (<1 wt%) has not yet been reported. Furthermore, the high temperature strength of LPS-SiC ceramics at temperatures above 1800 °C has never been reported.

In the present study, LPS-SiC ceramics sintered with 2000 ppm Y_2O_3 were fabricated using a hot-pressing technique. Their grain boundary structure and junction pockets were characterized using scanning transmission electron microscopy (STEM), and the high temperature strength of the ceramic was examined for the first time at temperatures up to 2000 °C in a nitrogen atmosphere.

2. Experimental procedure

To prepare SiC with 2000 ppm Y_2O_3, β-SiC (~0.5 μm, grade BF-17, H.C. Starck, Berlin, Germany) and yttrium nitrate ($Y(NO_3)_3{\cdot}4H_2O$, 99.99% pure, Sigma-Aldrich Co., St. Louis, MO, USA) were mixed via ball milling using SiC balls and a polypropylene jar for 6 h in ethanol. The chemical composition of the powder mixture was designed to yield a SiC ceramic with 2000 ppm Y_2O_3. The mixture was dried, pyrolyzed at 800 °C for 1 h in a nitrogen atmosphere, and finally hot-pressed at 2200 °C for 4 h under an applied pressure of 40 MPa in a nitrogen atmosphere. Discs 150 mm in diameter and 10 mm in thickness were prepared using this method.

The relative density of the hot-pressed specimen was determined using the Archimedes method. The theoretical density of the specimen (3.218 g/cm^3) 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). X-ray diffraction (XRD) using Cu Kα radiation was performed on ground powders for the hot-pressed specimen and the specimen tested at 2000 °C. XRD data were analyzed using the Rietveld refinement method for quantitative phase analysis of SiC polytypes. The thin foils for STEM observation were prepared via conventional methods, including mechanical thinning to ~10 μm and ion beam milling at an acceleration voltage of 2–3 kV using an Ar ion beam. Z-contrast HAADF-STEM images were taken using a scanning transmission electron microscope (JEM-2100F, JEOL, Tokyo, Japan) at 200 kV with a spherical aberration corrector (Corrected Electron Optical Systems GmbH, Heidelberg, Germany). The optimum size of the electron probe was 0.9 Å. The collection semi-angles of the HAADF detector were adjusted from 90 to 220 mrad in order to exploit large-angle elastic scattering of electrons for clear Z-sensitive images. The obtained raw images were band-pass filtered

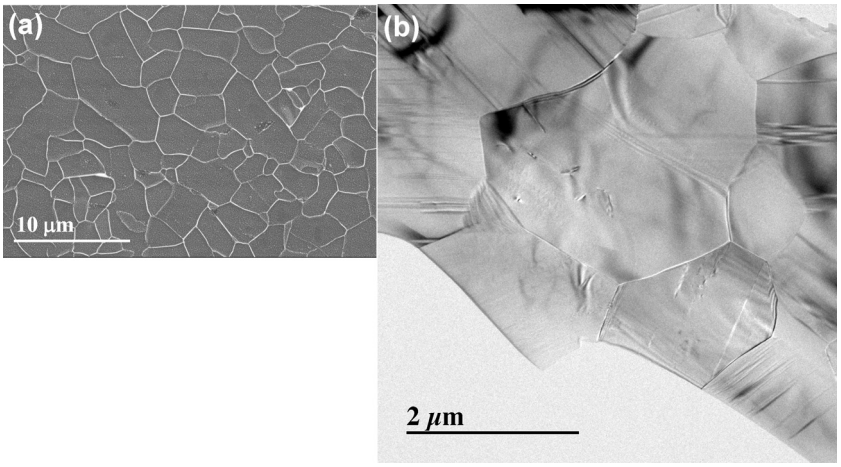

Fig. 2. Typical microstructure of SiC ceramic sintered with 2000 ppm Y_2O_3: (a) SEM and (b) TEM images. The SiC ceramic consists of slightly elongated grains and liquid pockets at the triple junctions. Liquid pockets are not frequently observed.

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