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Effect of Liquid Phase Content on Thermal Conductivity of Hot-Pressed Silicon Carbide Ceramics

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1. Introduction

Silicon carbide (SiC) is a promising material for Particle-Based Accident Tolerant (PBAT) fuel, fission, and fusion power applications due to its superior physical and thermal properties such as low specific mass, low neutron cross section, excellent radiation stability, low coefficient of thermal expansion, and high thermal conductivity [1-3].

Thermal conductivity of PBAT fuel is one of very important factors for plant safety and energy efficiency of nuclear reactors. Recently, a fully dense polycrystalline SiC ceramic with a high thermal conductivity (234 W/m·K) was reported by hot-pressing β-SiC powders with 1 vol% Y_2O_3-Sc_2O_3 as a sintering additive [4]. The Y_2O_3-Sc_2O_3 additive system led to serve as an oxygen getter within SiC lattice by forming fully crystallized secondary phase in multigrain junctions. Thus, optimizing of the thermal conductivity of SiC ceramics with Y_2O_3-Sc_2O_3 by adjusting the additive contents would be of interest.

In the present work, the effect of Y_2O_3-Sc_2O_3 content on the microstructure and thermal properties of the hot-pressed SiC ceramics have been investigated.

2. Experimental Procedure

Commercially available β-SiC (0.5 μm, 99.9%, Grade BF-17, H. C. Starck, Berlin, Germany) and RE_2O_3 (RE=Y, Sc, 99.9% pure, Kojundo Chemical Lab Co. Ltd.) were used as the starting powders. Five batches of powder mixtures were prepared. The mixture of the raw materials was β-SiC and 0.25 to 3 vol% Y_2O_3-Sc_2O_3 in the 1:1 molar ratio. Table 1 shows the batch composition and sintered density of liquid-phase sintered SiC ceramics. All individual batches were mixed by ball milling using SiC media in a polypropylene jar for 24 h in ethanol. The mixture was dried, sieved (60 mesh), and hot pressed at 2050°C for 6 h under 40 MPa of an applied pressure in a nitrogen atmosphere.

The relative density of the hot-pressed specimen was determined using the Archimedes method. Using Cu Kα radiation, X-ray diffraction data was obtained for the ground powder. The hot-pressed specimen was polished and etched with CF_4 plasma containing 10% oxygen. The morphology of the etched microstructure was examined by scanning electron microscopy (SEM, S4300, Hitachi Ltd., Hitachi, Japan).

Thermal diffusivity at room temperature was measured using the laser flash method. Differential scanning calorimetry (DSC, Model Q200, TA Instrument Inc. New Castle DE) and thermal diffusivity measurement equipment (Model LFA 447, NETZSCH GmbH, Selb, Germany) were used for the heat capacity (C_p) and thermal diffusivity (D) measurements, respectively. The thermal conductivity κ was calculated according the equation, $\kappa = D\ \rho\ C_p$, where ρ is the density of the sample.

Table 1. Batch composition and sintered densities of liquid-phase sintered SiC ceramics

Sample	Batch Composition (vol%)	Relative Density (%)
SY0.25	99.75% β-SiC + 0.25% (Y_2O_3+Sc_2O_3)	> 91
SY0.5	99.5% β-SiC + 0.5% (Y_2O_3+Sc_2O_3)	> 99
SY1 [4]	99% β-SiC + 1% (Y_2O_3+Sc_2O_3)	> 99
SC2	98% β-SiC + 2% (Y_2O_3+Sc_2O_3)	> 99
SC3	97% β-SiC + 3% (Y_2O_3+Sc_2O_3)	>99

3. Results

The relative densities of the hot-pressed samples were > 99% for all samples except SY0.25 sample. This result suggested that even though extremely small amount 0.5 vol% of Y_2O_3-Sc_2O_3 additive were sufficient to fully densify SiC ceramics by conventional hot-pressing at 2050◦C for 6 h under an applied pressure of 40 MPa in flowing nitrogen, indicating that the Y_2O_3-Sc_2O_3 system is very effective for the densification of SiC. The XRD patterns of the samples are shown in Fig. 1. The XRD analysis for all specimens showed the presence of β-SiC (3C) as a major phase and α-SiC (6H, 4H) as a minor phase, indicating the occurrence of the β→ α phase transformation of SiC during hot-pressing. The 1 vol%

content in Y_2O_3-Sc_2O_3 system is critical content in order to suppress the β to α phase transformation of SiC.

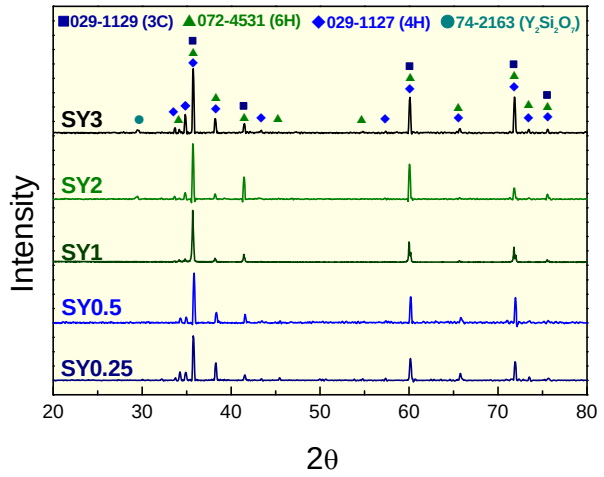

Fig. 1. X-ray diffraction patterns of SiC ceramics: (a) SY0.25, (b) SY0.5, (c) SY1 , (d) SY2, and (e) SY3 (refer to Table 1).

The microstructures of the hot-pressed samples are shown in Fig. 2. The grain growth of SiC increased with increasing additive content and large equiaxed grains grew in the small matrix grains of the microstructure by Ostwald-ripening mechanism. The XRD patterns and microstructure observation indicate that the β→α phase transformation was not completed in the present samples.

The thermal conductivity of the SC0.5, SC1, SC2 and SC5 specimens was 138, 234, 208, and 201 W/m·K, respectively. This high thermal conductivity was attributed to the beneficial effect of the Y_2O_3-Sc_2O_3 additives in decreasing the lattice oxygen content [4] and the confinement of poor-conducting RE-containing phases in the junction areas. The 1 vol% additive is an optimal content for maximizing the thermal conductivity of SiC ceramics sintered with Y_2O_3-Sc_2O_3 additive system. The Y_2O_3-Sc_2O_3 additives were very effective in achieving high thermal conductivity in SiC ceramics due to the following reasons: (1) an oxygen getter in the SiC lattice by forming crystallized $(Sc,Y)_2Si_2O_7$ phase in multigrain junctions, (2) Y and Sc were not soluble in the SiC lattice because of large differences in ionic size of the elements, and (3) forming clean or crystallized SiC-SiC boundaries. Further addition of additives exceeding 1 vol% decreased the thermal conductivity of SiC ceramics gradually, due to the enhanced phonon scattering at the β/α-SiC phase boundaries and greater amount of secondary phases which has lower thermal conductivity than SiC.

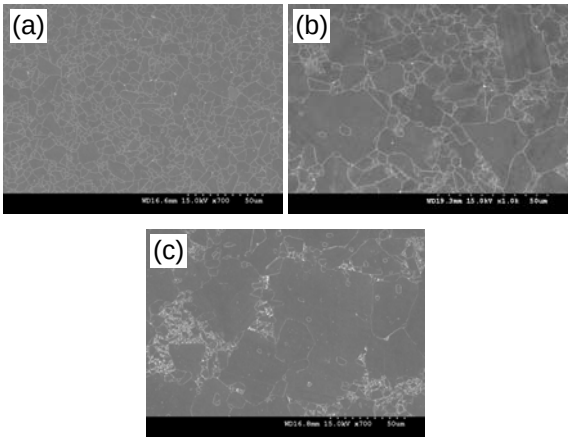

Fig. 2. Typical microstructures of SiC ceramics: (a) SY0.5, (b) SY1, and (c) SY3 (refer to Table 1).

4. Conclusions

SiC ceramics with 0.5~3 vol% Y_2O_3-Sc_2O_3 additives could be fully densified by conventional hot pressing at 2050°C for 6 h under an applied pressure of 40 MPa. Thermal conductivity of SiC ceramics sintered with Y_2O_3-Sc_2O_3 showed a maximum (234 W/m·K) when 1 vol% additives were added and, then, decreased with increasing the additive content. Suppressing the β to α phase transformation of SiC ceramics is beneficial in increasing the thermal conductivity of liquid-phase sintered SiC ceramics. Developed SiC ceramics with Y_2O_3-Sc_2O_3 additives are very useful for thermal conductivity on matrix material of the PBAT fuel.

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