Transactions of the Korean Nuclear Society Autumn Meeting
Pyeongchang, Korea, October 30-31, 2014

Effect of Starting Powder Polytype on Thermal Conductivity of Liquid-Phase Sintered Silicon Carbide Ceramics

Kwang-Young Lim[a], Seung-Jae Lee[a], and Young-Wook Kim[b]
[a]*Materials Development Group, KEPCO Nuclear Fuel, Daejeon 305-353, Republic of Korea*
[b]*Functional Ceramics Laboratory, Department of Materials Science and Engineering, The University of Seoul, Seoul 130-743, Republic of Korea*
E-mail: kylim@knfc.co,kr

1. Introduction

Silicon carbide (SiC) is an important candidate for Particle-Based Accident Tolerant (PBAT) fuel, fission, and fusion power applications because of its superior physical and mechanical properties such as low specific mass, low neutron cross section, excellent radiation stability, excellent corrosion resistance, low coefficient of thermal expansion, and high thermal conductivity [1].

Thermal conductivity of PBAT fuel is one of very important factors for plant safety and energy efficiency of nuclear reactors. PBAT fuels encapsulated with SiC enable a nuclear reactor to reduce the stored energy in the core and produce more thermal energy. For this reason, improvement in the thermal conductivity of SiC matrix is an important research issue for PBAT fuel. According to the previous reports, the thermal conductivity of high-purity SiC single crystal was 490 W/m·K [2] and maximum experimental value of hot-pressed polycrystalline SiC ceramics was 270 W/m·K [3]. The SiC ceramic was obtained by hot-pressing SiC powders with BeO additive. However, the application of the material has been limited because of the toxicity of Be. Therefore, developments of new additive compositions for polycrystalline SiC is essential for developing PBAT fuels encapsulated with SiC with high thermal conductivity. Recently, a fully dense 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].

In the present study, the effect of initial α-SiC content on the microstructure and thermal properties of the hot-pressed SiC ceramics have been investigated.

2. Experimental Procedure

Commercially available α-SiC (FCP15C, Norton AS, Lillesand, Norway), β-SiC (Ultrafine, Betarundum, Ibiden Co. Ltd., Ogaki, Japan, and RE_2O_3 (RE=Y, Sc, 99.9% pure, Kojundo Chemical Lab Co. Ltd.) were used as the starting powders. Three batches of powder mixtures were prepared, each containing 99 vol% SiC (α and/or β) and 1 vol% Y_2O_3-Sc_2O_3 additives. The relative content of α-SiC powder in batches with 1 vol% Y_2O_3-Sc_2O_3 was 10, 50, and 100 vol%. 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 (XRD) 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 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. The average phonon mean free path of each sample was calculated from the sound velocity, the measured heat capacity, and the measured thermal conductivity .

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

Sample	Batch Composition (vol%)	Relative Density (%)
SY10	9.9% α-SiC + 89.1% β-SiC + 1% (Y_2O_3+Sc_2O_3)	99.9
SY50	49.5% α-SiC + 49.5% β-SiC + 1% (Y_2O_3+Sc_2O_3)	99.9
SY100	99% α-SiC + 1% (Y_2O_3+Sc_2O_3)	99.9

3. Results

The relative densities of the hot-pressed samples were 99.9% for all samples. The addition of 1 vol% Y_2O_3–Sc_2O_3 was enough for densifying SiC ceramics fully at the present processing conditions. The XRD patterns of the samples are shown in Fig. 1 [5]. The SY10 and SY50 samples consisted of 3C as a major phase and 6H and 4H phases as minor phases. In contrast, the SY100 sample consisted of 6H and 4H phases.

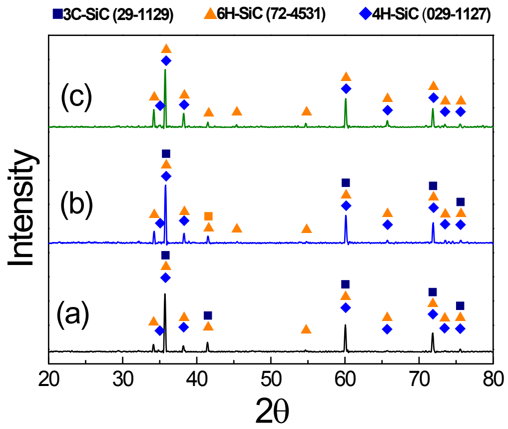

Fig. 1. X-ray diffraction patterns of SiC ceramics: (a) SY10, (b) SY50, and (c) SY100 (refer to Table 1) [5].

The microstructures of the hot-pressed samples are shown in Fig. 2. The microstructure of all samples consisted of equiaxed SiC grains. The grain size decreased with increasing the α-phase content in the starting composition, owing to the increased impingement of the growing α–SiC grains. The XRD patterns and microstructure observation indicate that the β→α phase transformation was not completed in the present samples.

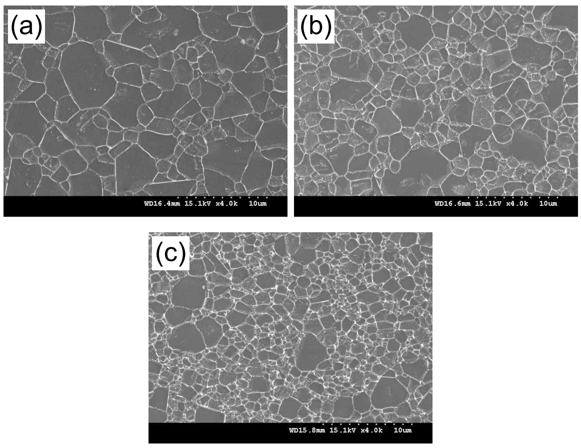

Fig. 2. Typical microstructures of SiC ceramics: (a) SY10, (b) SY50, and (c) SY100 (refer to Table 1).

Thermal conductivities of the SY10 and SY100 samples were 131 W/m·K and 159 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. Both thermal diffusivity and mean free path decreased with increasing α-phase content in the starting composition. An increase in α-phase content in the starting composition increased impingement between growing SiC grains, resulting in a decreased grain size. The finer grain size made more grain boundaries per unit volume, resulting in more phonon scattering. Thus, the increase of α-phase content in the starting composition led to a detrimental effect on improving the thermal conductivity of SiC ceramics under the present processing conditions.

4. Conclusions

The effect of crystalline phase (α-phase content) in starting powders on microstructure and thermal properties of SiC ceramics has been investigated by using submicron-sized α- and/or β-SiC starting powders. The addition of small amount (1 vol%) of Y_2O_3-Sc_2O_3 additives resulted in 99.9% densification of submicron SiC powder mixtures by hot-pressing at 2050°C for 6 h. The thermal conductivity decreased with increasing α-SiC content in the starting composition. Such results suggest that both the presence of α/β phase boundaries and the occurrence of β→α phase transformation of SiC are detrimental for improving the thermal conductivity of polycrystalline SiC ceramics. A maximal thermal conductivity of 159 W/m·K was obtained when 10% α-SiC was added in the starting composition.

REFERENCES

[1] L. L. Snead, T. Nozawa, Y. Katoh, T.S. Byun, S. Kondo, and D. A. Petti, Handbook of SiC properties for fuel performance modeling, J. Nucl. Mater., Vol. 371, pp. 329-377, 2007.
[2] G. A. Slack, Thermal conductivity of pure and impure silicon, silicon carbide, and diamond, J. Appl. Phys., Vol. 3, pp. 3460-3466, 1964.
[3] Y. Takeda, Development of high-thermal-conductive SiC ceramics, Ceram. Bull., Vol. 67, pp. 1961-1963, 1988.
[4] Y.-W. Kim, K.-Y. Lim, and W.S. Seo, Microstructure and thermal conductivity of silicon carbide with yttria and scandia, J. Am. Ceram. Soc., Vol. 97, No. 3, pp 923-928, 2014.
[5] K.-Y. Lim, T.Y. Cho, Y.-W. Kim, and S.J. Lee, Effect of initial alpha-SiC content on thermal conductivity of silicon carbide ceramics, Key Eng. Mater., Vol. 616, pp. 23-26, 2014.