Characteristics of SiC Layers in TRISO Particle Fuel Deposited at Low Temperatures

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1. Introduction

High-temperature gas-cooled reactors (HTGRs) utilize TRISO-coated particles as a fuel, consisting of UO_2 microspheres coated with layers of porous pyrolytic carbon (porous PyC), inner dense PyC (IPyC), silicon carbide (SiC), and outer dense PyC (OPyC) [1]. The porous PyC coating layer, the so-called buffer layer, attenuates fission recoils and provides a void volume for gaseous fission products and carbon monoxide. The IPyC layer acts as containment for gaseous products. The OPyC layer protects the SiC coating layer by inducing a compressive stress along with the IPyC layer and provides chemical compatibility with the graphite matrix in a fuel compact [1]. Among the TRISO coating layers the SiC layer is particularly important because it acts as a diffusion barrier to gaseous and metallic fission products and as a miniature pressure vessel for the particle. In order to insure the integrity of the SiC layer after fabrication and in use, the microstructure, mechanical properties, and chemical composition of the SiC layer should be controlled properly [2]. It has been known that the release rate of fission products through the SiC layer is higher in a large columnar structure than with small grain sizes [3]. The lower diffusivity of metallic fission products in the finer microstructure is due to the higher degree of tortuosity of the grain boundaries.

The SiC layer in TRISO-coated particles is normally deposited at temperatures between 1500° and 1650℃. In this study, we investigated various microstructural and chemical features of SiC layers deposited at lower temperatures between 1300° and 1400℃ to obtain a finer grain size while fixing the other deposition parameters.

2. Experimental Procedure

Coatings of the TRISO particles were conducted using ZrO_2 kernels in a FBCVD reactor. In this work, a graphite tube of 25 mm inner diameter with an inlet nozzle of 3 mm at the base of a 60° cone was used as a coating bed. Input gases for the depositions of the buffer, IPyC (OPyC) and SiC were C_2H_2/Ar, C_2H_2/C_3H_6/Ar and CH_3SiCl_3 (MTS)/H_2/Ar, respectively. For the deposition of the SiC layer, the deposition temperature was varied from 1300° to 1400℃ at a constant gas flow rate of 3000 cm^3/min. For the purpose of comparison, standard SiC TRISO particles deposited at 1500℃ were also prepared.

Microstructures of the SiC layer were characterized using SEM and TEM. For the SEM observation of the outer surface of the SiC layer, the OPyC layer was burned off at 850℃ for 2 h in air. The TEM samples were prepared on the cross-section of the SiC layer by a focused ion beam (FIB) technique. Phase purity and stoichiometry of the SiC layer were measured using X-ray powder diffraction (XRD) and Raman micro-spectroscopy, respectively. Raman microspectroscopy analyses were carried out at five points on the polished cross-section of the SiC layer. The analyses points were 5, 10, 15, 20, 25 μm apart from the IPyC/SiC interface along the growth direction. The existence of polytypes with noncubic structures such as 2H, 4H, 6H, 15R, etc. was characterized by a ^{29}Si magic-angle spinning nuclear magnetic resonance (MASNMR) method.

3. Results and Discussion

Fig. 1 shows XRD patterns of SiC layers deposited at 1300° and 1500℃. Only β-SiC phase with cubic structure (3C) is detected in the XRD analysis irrespective of deposition temperature.

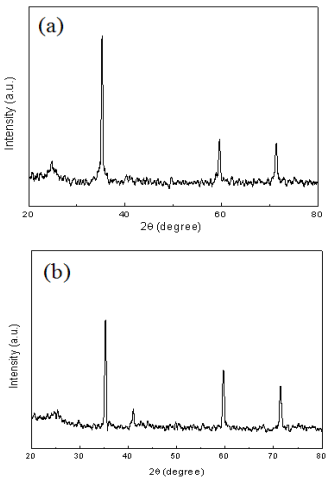

Fig. 1. XRD patterns of SiC layers deposited at 1300° (a) and 1500℃ (b).

Fig. 2 shows SEM microstructures for the surfaces of SiC layers deposited at different temperatures. The SiC layers deposited below 1400℃ have pebble-like structures which consist of much smaller crystallites. On the other hand, the SiC layer deposited at 1500℃ shows

a faceted structure with larger grains. The size of pebbles does not vary significantly with the deposition temperature. However, the size of crystallites within the pebbles decreases as the deposition temperature decreases.

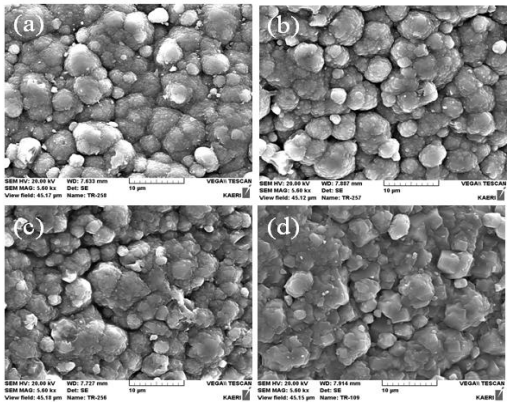

Fig. 2. SEM microstructures for the surfaces of the SiC layers deposited at 1300° (a), 1350° (b), 1400° (c), and 1500℃ (d).

The effect of deposition temperature on the grain size of SiC layer can be clearly seen in the TEM microstructure of Fig. 3. The SiC layer deposited at 1300℃ consists of very fine grains with a size of 0.1-0.5 μm. On the other hand, the grain size increases larger than 1.0 μm as the deposition temperature increases up to 1400℃.

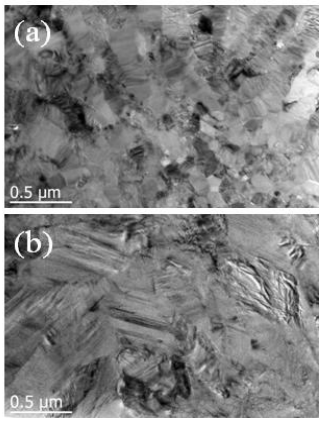

Fig. 3. TEM microstructures of the SiC layers deposited at 1300° (a) and 1400℃ (b).

Fig. 4 shows the results of Raman microspectroscopy analyses for the SiC layers deposited at 1300° and 1500℃. All SiC layers show stoichiometric compositions without any presence of free Si or free C. There are transverse optical (TO), longitudinal optical (LO), and second order SiC bands at 796, 972, and 1522 cm^{-1}, respectively, for both temperatures. However, the SiC layer deposited at 1300℃ shows a broad acoustic SiC band at around 450 cm^{-1} which are due to a large amount of stacking faults exists within SiC crystallites.

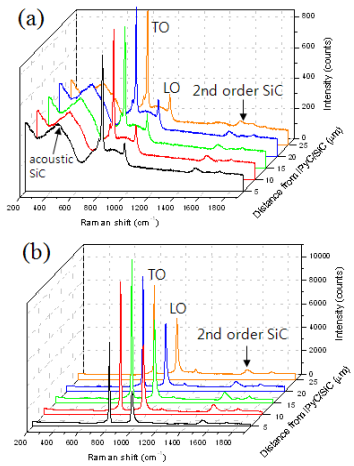

Fig. 4. Raman spectra of the SiC layers deposited at 1300° (a) and 1500℃ (b).

Fig. 5 shows MASNMR spectra of the SiC layers deposited at 1300° and 1500℃. Although the SiC layer deposited at 1500℃ shows only 3C cubic structure, the SiC layer deposited at 1300℃ consists of 3C and a mixture of polytypes. The effect of the existence of such a fine scale polytypes on the performance of TRISO particles needs to be elucidated through irradiation studies.

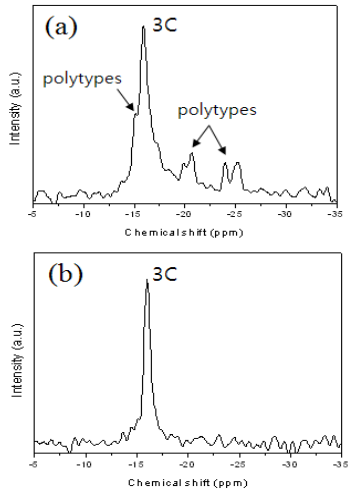

Fig. 5. MASNMR spectra of the SiC layers deposited at 1300° (a) and 1500℃ (b).

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