Effect of Gas Flow Rate on the Microstructure and Properties of Silicon Carbide Layers in TRISO-Coated Particles

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1. Introduction

TRISO-coated fuel particles for high-temperature gas-cooled reactors consist 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) [1,2]. 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 [3].

For a uniform coating of the microspherical particles, the TRISO coating is performed using a fluidized-bed chemical vapor deposition (FBCVD) method. In the method, the process conditions such as the gas flow rate, concentration of the coating gas, coating temperature, etc., largely affect the characteristics of the coating layer [2,3]. Among the deposition parameters the gas flow rate mainly determines the fluidization behavior of microspherical particles. In this study, we investigated the effect of the gas flow rate on the microstructure and properties of the SiC layer 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 gas flow rate was varied between 2000 and 7000 sccm (cm^3/min) at a constant coating temperature of 1500℃ and an input gas ratio ((Ar+H_2)/MTS) of 100. Microstructures of the SiC layer were characterized using optical microscopy (OM) and scanning electron microscopy (SEM). For observation of the outer surface of the SiC layer, the OPyC layer was burned off at 800℃ for 2 h in air. Phase purity and stoichiometry of the SiC layer were measured using X-ray powder diffraction (XRD) and Auger electron spectroscopy (AES), respectively. The XRD was carried out after burning off the OPyC layer. The AES analysis was performed on the polished cross-sections of the coated particles. The native oxide layer on the SiC surface was sputtered using an Ar ion beam before the analysis. Pore size distribution and porosity of the SiC layer were measured from twenty SEM micrographs for each specimen using an image analyzer. Hardness and elastic modulus were evaluated by a nano-indentation tester (Nano Indenter XP, MTS) equipped with a Berkovich diamond tip. Indentations were applied along the middle plane of the SiC layer on the diamond-polished cross-section to exclude an interference of the adjacent IPyC and OPyC layers. Five particles were measured for each coating condition and ten indentations were made for each particle.

3. Results and Discussion

Fig. 1 shows XRD patterns of the coated particles after burning off the OPyC layers. There is no appreciable difference in the XRD results with variation of the gas flow rate. The SiC layers formed at gas flow rates consist exclusively of the β-SiC phase, at least within the resolution limit of the XRD technique. The small carbon peaks were due to the IPyC layer beneath the SiC layer, which could be confirmed by AES analysis.

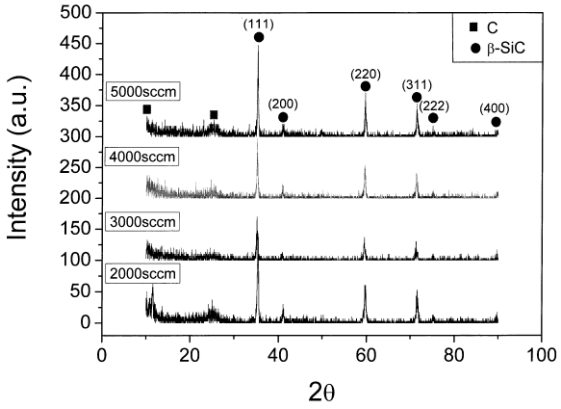

Fig. 1. XRD patterns of SiC layers deposited at various gas flow rates.

Fig. 2 shows surface microstructures of the SiC layers deposited at various gas flow rates. Grain size and shape of SiC grains do not show a large difference with variation of the gas flow rate, inferring a mass transport process through a boundary layer as a rate-controlling mechanism of the deposition reaction.

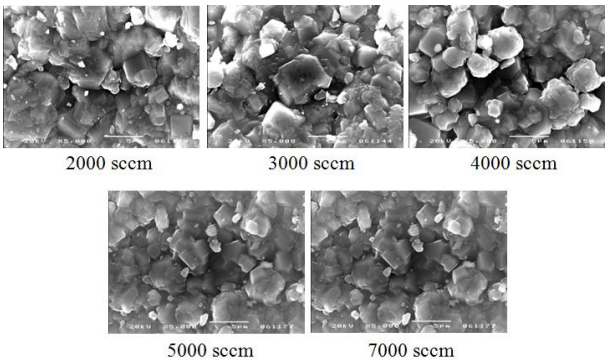

Fig. 2. SEM microstructures for the surfaces of SiC layers deposited at various gas flow rates.

Pore size distribution and porosity of the SiC layers deposited at various gas flow rates are shown in Figs. 3 and 4, respectively. At gas flow rates lower than 3000 sccm, most pores are less than 1.5 μm in size whereas pores larger than 2.0 μm start to appear at gas flow rates higher than 4000 sccm. The porosity also starts to increase largely at gas flow rates higher than 4000 sccm. A possible reason for the porosity increase would be a change in the mode of particle fluidization. At an optimum gas flow rate, the mode of particle fluidization is characterized as normal spouting. That is, a stream of the particles rises rapidly in the center as the spout, reaches a maximum level, then falls back onto the annular space around the spout and travels downward uniformly as a packed bed. As the gas flow rate increases, the particles can be fluidized more vigorously. If the particle fluidization is in the violent mode, the particles will be blown out of the normal deposition zone and the probability of the formation of internal flaws will increase, leading to an increase of the pore size and porosity of the SiC layer.

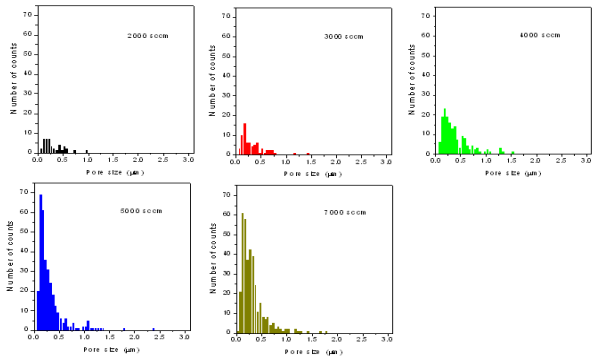

Fig. 3. Pore size distribution of SiC layers deposited at various gas flow rates.

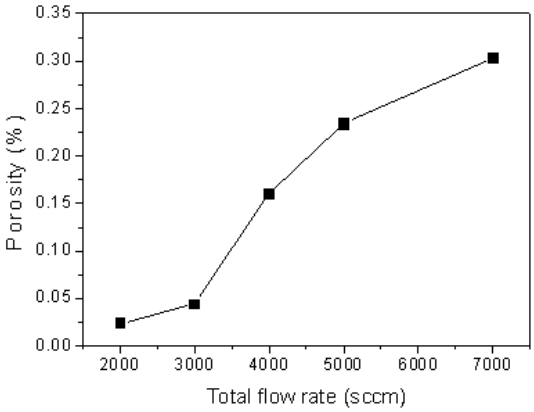

Fig. 4. Porosity of SiC layers deposited at various gas flow rates.

In summary, the phase purity and the surface microstructure of SiC layers did not show an appreciable difference with variation of the gas flow rate. The porosity of SiC layer, however, largely increased at gas flow rates higher than 4000 sccm. This could be attributed to the violent spouting of particles as the gas flow rate increased. In the violent mode of particle fluidization, the deposition can occur out of the constant deposition zone, leading to the higher porosity of the SiC layer.

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