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Effects of FBCVD Conditions on the Properties of a SiC Layer in TRISO 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 (PyC), inner dense PyC (IPyC), silicon carbide (SiC), and outer dense PyC (OPyC) [1]. For a uniform coating of the microspherical particles, the TRISO coating is performed using a fluidized-bed chemical vapor deposition (FBCVD) method. Among the TRISO coating layers the SiC layer is particularly important because it acts as a diffusion barrier to gaseous/metallic fission products and as a miniaturized pressure vessel of the particle. In order to insure the integrity of the SiC layer after a fabrication and in use, the microstructure, mechanical properties, and chemical composition of the SiC layer should be controlled properly [2]. Characteristics of the coating layer depend largely on the FBCVD conditions such as the gas flow rate, concentration of the coating gas, coating temperature, etc [3,4]. In this study, we investigated the effects of the deposition temperature and the gas flow rate on the properties of the SiC layer. Microstructure, chemical composition, porosity, and mechanical properties of the SiC layer were characterized by various techniques.

2. Experimental Procedure

Coatings of the TRISO particles were conducted on ZrO_2 kernels as surrogates for UO_2 particles in a FBCVD coater. 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. At the deposition temperatures, 14 g of ZrO_2 particles were put into the coater from the top of the graphite tube in the presence of an Ar flow from the bottom of the coater. After assuring the fluidization of the particles through a quartz window, reactants were put into the coater to produce a coating layer on the particles fluidized in the coater. 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 coating temperature and the gas flow rate were controlled at 1460°~1550°C and 2500~4000 sccm, respectively, with an input gas ratio (Ar+H_2/MTS) of 100. All the TRISO layers were continuously coated without unloading the particles after each coating step.

Microstructure, phase purity, and chemical composition of the SiC layer were observed using a scanning electron microscopy (SEM), X-ray powder diffraction (XRD), and an auger electron spectroscopy (AES), respectively. Porosity of the SiC layer was measured from SEM micrographs using an image analyzer. Hardness and elastic modulus were evaluated by a nanoindentation method.

3. Results and Discussion

The SiC layers coated at 1460°~1550°C exclusively consisted of the β-SiC phase and contained no free Si phase at least within a resolution limit of the XRD technique. In order to take a closer look at the stoichiometry of the SiC layer, AES analyses were performed for the SiC layers coated at different temperatures (Fig. 1). It can be seen from this figure that a stoichiometric SiC is obtained at deposition temperatures below 1500°C while an excess C appears at 1550°C. This result is consistent with a previous report in which the β-SiC+C phase tended to be deposited at high temperatures [5].

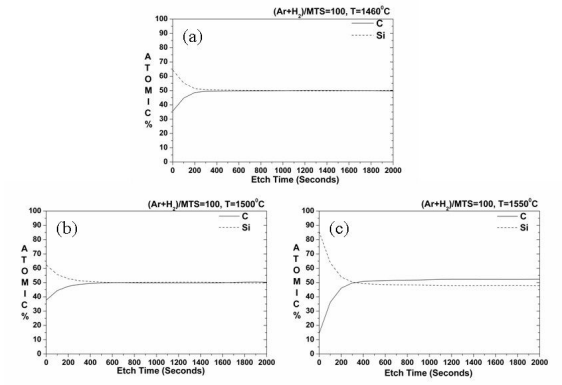

Fig. 1. AES analyses of the SiC layers deposited at (a) 1460°, (b) 1500°, and (c) 1550°C.

Fig. 2 shows the surface and the cross-sectional microstructures of the SiC layers deposited at different

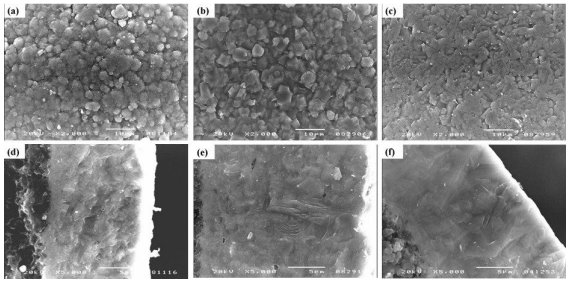

Fig. 2. SEM micrographs for the surfaces and the cross-sections of the SiC layers deposited at 1460°C ((a), (d)), 1500°C ((b), (e)), and 1550°C ((c), (f)).

temperatures. The SiC layer deposited at 1460°C shows an inhomogeneous grain distribution mixed with small and large grains. The grain size increases slightly and the homogeneity is improved as the deposition temperature increases. The SiC layers deposited at 1460° and 1500°C contain a little porosity while the SiC layer deposited at 1550°C shows the lowest porosity. The porosity was closely correlated with the deposition rate of the SiC layer [4].

Fig. 3 shows the hardness and elastic modulus of the SiC layers deposited at different temperatures. The SiC layer deposited at 1460°C shows the lowest hardness and elastic modulus even though the sample has a stoichiometric composition as previously shown in Fig. 1. The low mechanical properties seem to be due to the inhomogeneity of the grains and the high porosity. The hardness and the elastic modulus show a maximum at 1500°C and then decrease again at 1550°C. The SiC layer deposited at 1550°C shows lower mechanical properties than the sample at 1500°C in spite of the lower porosity. This is likely to be due to the existence of excess carbon.

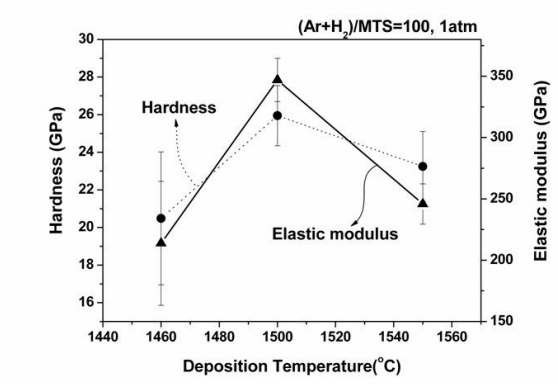

Fig. 3. Hardness and elastic modulus of the SiC layers deposited at different temperatures.

One of the most important variables in the FBCVD process is the gas flow rate because it affects the stability of a particle fluidization. The cross-sectional SEM microstructures of the SiC layers deposited at different gas flow rates are shown in Fig. 4. It can be seen that the porosity increases as the gas flow rate increases. Fig. 5 shows the relation between the deposition rate and the porosity and the pore size distribution of the SiC layers. The deposition rate and the porosity show low values at 2500 sccm and rapidly increase at 3000 and 4000 sccm. Even though the SiC layer deposited at 4000 sccm has a similar deposition

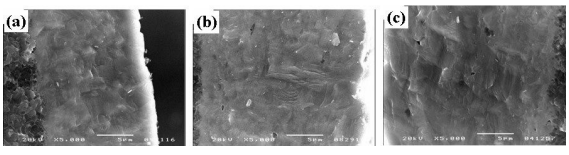

Fig. 4. SEM micrographs for the cross-sections of the SiC layers deposited at 1500°C with the gas flow rates of (a) 2500, (b) 3000, and (c) 4000 sccm.

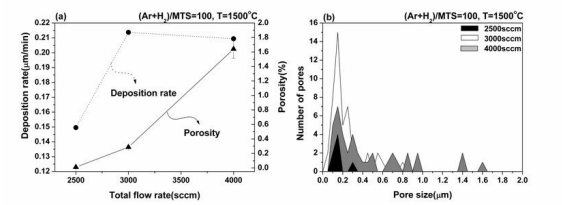

Fig. 5. Variation of (a) deposition rate and porosity and (b) pore size distribution of SiC layers as a function of the gas flow rate.

rate to the sample at 3000 sccm, it contains a much higher porosity and pores with sizes larger than 1 μm. It is ascribed to a violent spouting of particles at 4000 sccm, leading to a deposition at an inhomogeneous region and a formation of defects due to collisions between the tube wall and particles.

4. Conclusions

TRISO coatings on ZrO_2 surrogate kernels were conducted by a FBCVD method. Effects of the deposition temperature and the gas flow rate on the properties of the SiC layer were investigated in the TRISO-coated particles. At the deposition temperature of 1550°C the SiC layer contained an excess carbon, whereas the SiC layers had stoichiometric compositions at 1460° and 1500°C. Hardness and elastic modulus measured by a nanoindentation method were the highest in the SiC layer deposited at 1500°C. The SiC layer deposited at a gas flow rate of 4000 sccm exhibited a high porosity and contained large pores of more than 1 μm, due to a violent spouting of the particles. On the other hand, the SiC layer deposited at 2500 sccm revealed the lowest porosity.

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