

Characterization of SiC Layers in TRISO Coated Particles at Various Temperatures

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1. Introduction

TRISO-coated fuel particles for high-temperature gas-cooled reactors consist of UO_2 microspheres coated with multilayers 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, a fluidized-bed chemical vapor deposition (FBCVD) method is utilized.

Among the TRISO coating layers, the SiC layer is particularly important because it acts as a diffusion barrier to gaseous/metallic fission products and a miniaturized pressure vessel of the fuel particle. In order to insure the integrity of the SiC layer after a fabrication, the microstructure, mechanical properties, and chemical composition of the SiC layer should be properly controlled[2]. Properties of the coating layer depend largely on the FBCVD conditions such as a flow rate, concentration of the coating gas, coating temperature, etc[3,4].

In this study, we investigated the influence of deposition temperatures on the property of SiC layer. Microstructure, chemical composition, and other properties of the SiC layer were characterized by various techniques.

2. Experimental procedure

The TRISO coating layers were coated on the ZrO_2 kernels regarded as surrogates for UO_2 particles in a fluidized CVD coater. In this work, a graphite tube with inner diameter of 25mm and an inlet nozzle of 3 mm at the base of a 60° cone was used as a coating bed. At the deposition temperatures, ZrO_2 particles with the amount of 14g were inserted into the coater through the top of the graphite tube while Ar gas was flowed through the bottom of the coater. After assuring the fluidization of the particles through a quartz window, reactants were inserted into the coater to produce a coating layer on the fluidized particles. For the proper deposition of coating layers, the coating temperature and the gas flow rate were controlled at 1350°C~1500°C and 3000 sccm, respectively, with an input gas ratio (Ar+H₂/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 auger electron spectroscopy

(AES), Raman spectroscopy, nuclear magnetic resonance (NMR), respectively.

3. Results and Discussion

Raman spectroscopy and nuclear magnetic resonance (NMR) are two non-destructive characterisation techniques which provide more detailed information of the structure of SiC coating layer. Fig. 1 shows the Raman spectra of the SiC layer deposited at 1350°C~1500°C. Bands for silicon and carbon elements were not detected, so it was assumed that the coating was stoichiometric SiC.

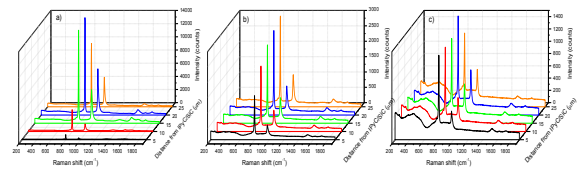


Fig. 1. Raman spectra of the SiC layers deposited at various temperatures; a)1500°C, b)1400°C, c)1350°C.

Two strong signals were observed at around 790 and 971 cm^{-1} . In 3C-SiC, only two bands are present, the transverse optical (TO) and longitudinal optical (LO) bands at 790 and 971 cm^{-1} , respectively. However, acoustic SiC bands were observed in the SiC layer coated at 1350°C due to an increased grain boundary area caused by fine grains and stacking faults[5].

Fig. 2 shows NMR spectra of the SiC layers deposited at 1350°C~1500°C. The SiC coating produced at 1500°C only showed the presence of 3C-SiC. This has the classic abc (kk) stacking sequence of the face-centered cubic crystal structure. The spectrum of SiC layer coated at 1400°C~1350°C indicated hexagonal and/or rhombohedral nature.

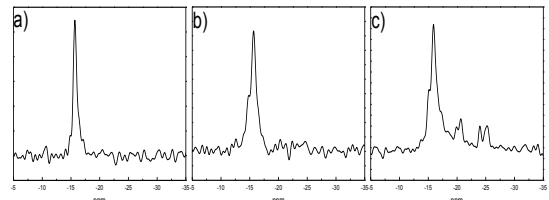


Fig. 2. NMR spectra of the SiC layers deposited at various temperatures; a)1500°C, b)1400°C, c)1350°C.

The main features of the spectrum are the three groups of peaks clustered around the three positions for type AB and C-site peaks. The largest peak is the peak

of 3C SiC (single A-type peak) at -16.0 ppm. Also present in region A is a shoulder at -15.2 ppm. B type peak is observed at -20 ppm and two major C-type peaks at -24 ppm and -25.2 ppm [6].

Fig. 3 shows surface microstructures of the SiC layers deposited at various temperatures. The size of SiC grains increased and the grain shape became more faceted with increasing coating temperature.

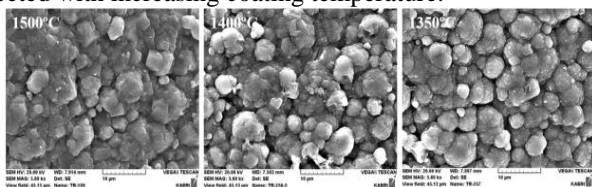


Fig. 3. SEM microstructures for the surfaces of SiC layers deposited at various temperatures.

Fig. 4 shows XRD patterns of the coated particles after burning off OPyC layers. There is no appreciable difference in the XRD results with variation of the temperatures. The observed small carbon peaks were due to the IPyC layer beneath the SiC layer, which could be confirmed by AES analysis (Fig. 5).

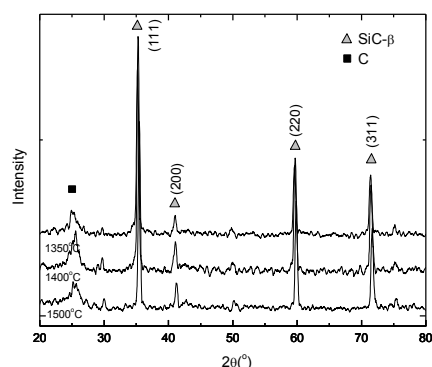


Fig. 4. XRD patterns of SiC layers deposited at various temperatures.

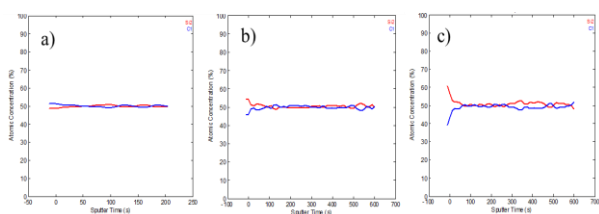


Fig. 5. AES analysis of SiC layers deposited at a) 1500°C, b) 1400°C, c) 1350°C.

4. Conclusions

TRISO coatings on ZrO_2 surrogate kernels were conducted using a FBCVD method. The influence of the deposition temperature on the properties of the SiC

layer were investigated. The SiC layers coated at 1350 °C contained a small amount of excessive carbon, while the stoichiometric composition of SiC coating layer was obtained at 1400°C~1500°C. Moreover, it was verified that the observed acoustic SiC bands in Raman spectra was due to an increased grain boundary caused by fine grains and stacking faults.

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