

Characterization of the SiC Layer in TRISO-Coated Particles

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1. Introduction

Fuel for high-temperature gas-cooled reactors (HTGRs) is featured by TRISO-coated fuel particles, which consist of UO_2 microspheres coated with layers of porous pyrolytic carbon (PyC), inner dense PyC (IPyC), SiC, and outer dense PyC (OPyC) [1]. The function of these coating layers is to retain fission products within the particle. In particular the SiC coating layer is the most important layer among TRISO coatings because it acts as a diffusion barrier to metallic fission products and a miniaturized pressure vessel of the particle. In order to insure the integrity of the SiC layer after fabrication and in use, the microstructure and chemical composition of the SiC layer should be controlled properly [2]. Characteristics of the coating layer depend largely on the various chemical vapor deposition (CVD) conditions in a fluidized-bed reactor, such as gas flow rate, concentration of the coating gas, coating temperature, etc [3,4].

In this paper, we investigated effects of the fluidized-bed CVD conditions especially on the characteristics of the SiC layer. Microstructure, chemical composition and other properties of the SiC layer were characterized by various techniques.

2. Experimental Procedure

The TRISO coatings were produced on the ZrO_2 kernels as surrogates for UO_2 particles in a fluidized CVD 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 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 depositions of buffer, IPyC (OPyC), and SiC were $\text{C}_2\text{H}_2/\text{Ar}$, $\text{C}_2\text{H}_2/\text{C}_3\text{H}_6/\text{Ar}$, and CH_3SiCl_3 (MTS)/ H_2/Ar , respectively. For the deposition of the SiC layer, the input gas ratio (H_2/MTS) was controlled to 50 or 100 with a total gas flow rate of 2000–5000 sccm. All the TRISO layers were continuously coated without unloading the particles after each coating step.

Microstructures of the coated particles were observed for the fractured and polished cross-sections using an optical microscopy (OM) and a scanning electron microscopy (SEM). Transmission electron microscopy (TEM) studies were also performed through a TEM

sample preparation by a focused-ion beam (FIB) technique. Chemical composition and impurities of the SiC layer were analyzed by an EDS attached to TEM and an auger electron spectroscopy (AES).

3. Results and Discussion

Equilibrium mole fractions of solid species were calculated using the SOLGASMIX-PV program in order to obtain deposition conditions for stoichiometric SiC phase. Fig. 1 shows the calculation results with variations of deposition temperatures and input gas ratios. It can be seen from this figure that a stoichiometric SiC is obtained at deposition temperatures above 1300°C if we control the dilution ratio to be higher than 20. No free Si phase is formed in this condition. We determined that the coating temperature and the input gas ratio should be higher than 1400°C and 50, respectively, according to the calculation result.

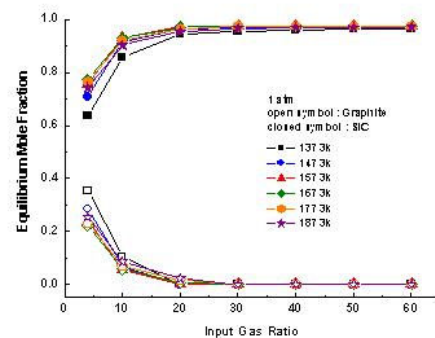


Fig. 1. Thermodynamic yield of SiC phase as a function of input gas ratio at various temperatures.

Fig. 2 shows the deposition rate of the SiC layer at 1500°C with a variation of the total gas flow rate. The

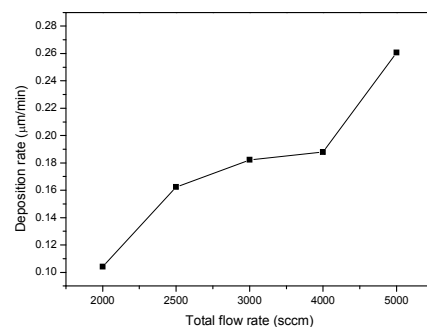


Fig. 2. Deposition rate of the SiC layer as a function of total gas flow rate at the coating temperature of 1500°C .

spouting mode of particles was in the state of normal at total flow rates between 2500 and 4000 sccm. At higher flow rates, the spouting would be in a violent mode, which could be an origin of the SiC failure [2]. Microstructures of the TRISO particles coated in this manner are shown in Fig. 3. The SiC layer contained little porosity and the thickness was $36.9 \pm 2.1 \mu\text{m}$. The coating rate at this condition was $0.13 \mu\text{m}/\text{min}$, being similar to the reported values [4]. The coating rate of the SiC layer should be controlled properly to obtain high quality coatings because the coating rate has been known to affect significantly the microstructures of the SiC layer [4].

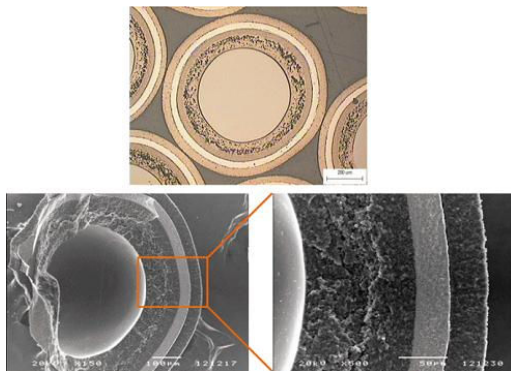


Fig. 3. OM and SEM images for the cross-sections of the TRISO particles.

Fig. 4 shows TEM microstructure at the interface between IPyC and SiC and the result of EDS analysis for the SiC layer. The interface between two layers is rough and not clearly defined. This is a feature of the continuous coating without loading and unloading after each coating step. The discontinuous coating process results in a smooth and sharp interface, which is not beneficial for the interface strength. The SiC layer consists of grains with sizes of $1\text{--}2 \mu\text{m}$ and contains no impurities such as oxygen or chlorine.

In XRD analyses for the surface of the SiC layer after burning out the OPyC layer, the SiC coating only consisted of cubic $\beta\text{-SiC}$ without any second phases. For a more detailed analysis of the existence of free Si

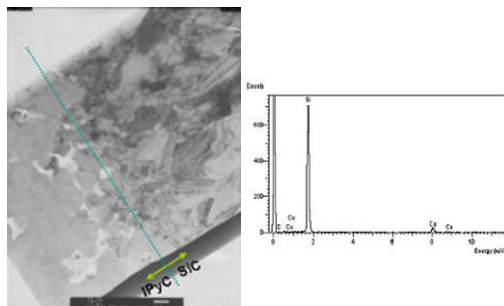


Fig. 4. TEM microstructure for the interface between IPyC and SiC layers and the result of EDS analysis of the SiC layer.

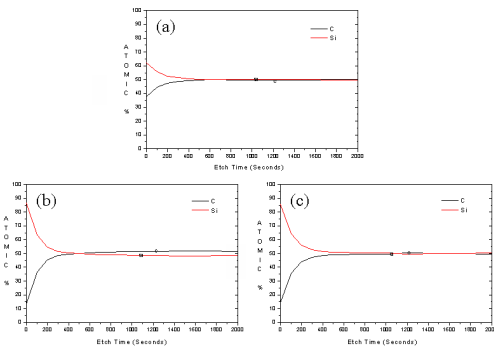


Fig. 5. Results of AES analyses for the SiC layers coated at different temperatures and input gas ratios (α). (a) 1500°C , $\alpha = 50$, (b) 1500°C , $\alpha = 100$, and (c) 1550°C , $\alpha = 50$.

or free C, however, AES analyses for the SiC layer were performed as well (Fig. 5). A stoichiometric SiC layer could be obtained at the coating temperature of 1500°C and the input gas ratio of 50. Variation of the coating condition, however, resulted in a slightly C-rich or a Si-rich SiC layer. These results are contrary to the calculation in Fig. 1, which may be due to an assumption of a thermodynamic equilibrium in the SOLGASMIX-PV program.

4. Conclusions

TRISO particles consisting of buffer/IPyC/SiC/OPyC layers were fabricated using a fluidized bed CVD reactor. The SiC layer consisted of $1\text{--}2 \mu\text{m}$ $\beta\text{-SiC}$ grains and contained no pores and impurities. The CVD condition should be controlled properly to obtain a stoichiometric SiC layer because the existence of free C or free Si would adversely affect the high-temperature and irradiation behaviors.

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