Effect of Fluidized Bed CVD Condition on the Microstructure of Buffer Layer in the TRISO Coated Particles

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1. Introduction

Fluidized bed chemical vapor deposition (FBCVD) of carbon and silicon carbide (SiC) has been applied to TRISO-coated fuel particles for high-temperature gas-cooled reactors (HTGRs). The TRISO-coated fuel particle consists 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. The porous PyC coating layer, called the buffer layer, attenuates fission recoils and provides void volume for gaseous fission products and carbon monoxide. The IPyC layer acts as a containment to gaseous products. The SiC layer provides mechanical strength for the particle and acts as a diffusion barrier to metallic fission products, which diffuse easily through the IPyC layer. The OPyC layer protects the SiC coating layer by inducing a compressive stress and provides chemical compatibility with a graphite matrix in the fuel compact. In order to insure the integrity of each layer after fabrication and in use the microstructure of the PyC and the SiC layers should be controlled properly [2]. The microstructure of the coating layer depends largely on the various FBCVD conditions in a fluidized-bed reactor, such as gas flow rate, concentration of the coating gas, coating temperature, etc.

In this paper, the current progress of the TRISO coating development in KAERI would be described. We performed some preliminary experiments on the TRISO coating using a fluidized bed reactor designed internally. Effect of the FBCVD conditions especially on the microstructure of the buffer layer was investigated. The concentration of the coating gas and the gas flow rate were varied at a fixed coating temperature.

2. Experimental Procedure

The TRISO coating is produced on the ZrO_2 kernels as surrogates for UO_2 particles in a fluidized CVD coater. A schematic diagram of the FBCVD reactor used in this study is shown in Fig. 1. 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 the coater.

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 C_2H_2/Ar, C_2H_2/C_3H_6/Ar, and CH_3SiCl_3/H_2/Ar, respectively. After deposition of coating layers, the reactant gas was replaced by Ar and the fluidizing state was maintained until the particles were cooled down. Microstructures of the coated particles were observed for the fractured and polished cross-sections using optical and scanning electron microscopy. Thickness of the coated layer was measured by a micrometer or from the cross-sectional micrographs.

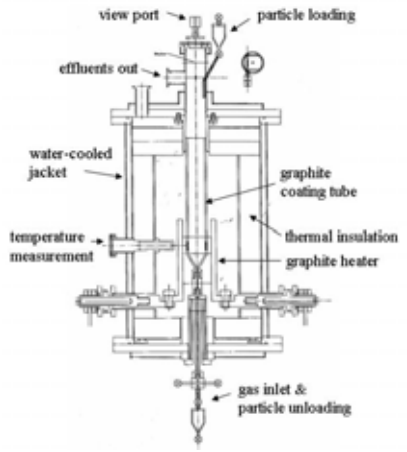

Fig. 1. Schematic diagram of the FBCVD coater.

3. Results and Discussion

Fig. 2 shows the result of the TRISO coating for all layers using the designed coater. The FBCVD conditions of each layer were as follows: buffer, 600 sccm Ar/1400 sccm C_2H_2, 1250°C, 5 min; IPyC (OPyC), 1400 sccm Ar/300 sccm C_2H_2/300 sccm C_3H_6, 1300°C, 10 min; SiC, 1980 sccm Ar/1980 sccm H_2/40 sccm MTS, 1500°C, 70 min. Coating rates of the buffer, IPyC, SiC, and OPyC layers were about 18.6, 3.2, 0.2, and 3.5 μm/min, respectively. In the standard process of the German TRISO-coated particle fuel fabrication, the coating rates of each layer were 6–10, 4–6, 0.2, and 4–6 μm/min [2]. When comparing with the German process, IPyC (OPyC) and SiC layers have similar coating rates while the coating rate of the buffer layer has a large discrepancy. The coating rate should

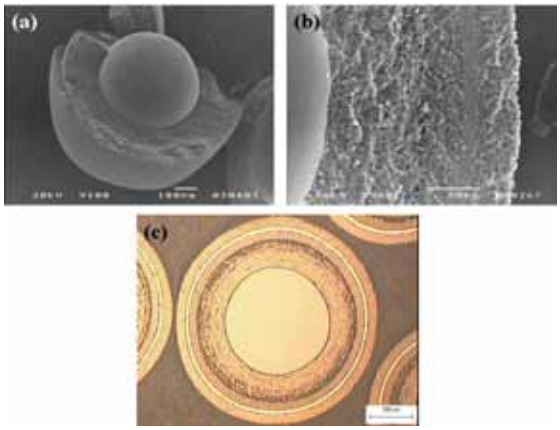

Fig. 2. Fractured ((a) and (b)) and polished (c) cross-sectional micrographs of the TRISO coated particles.

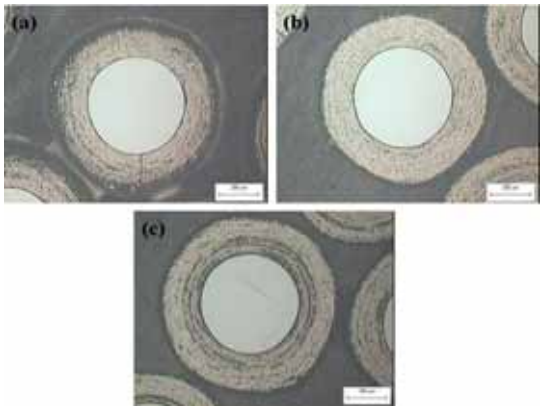

Fig. 4. Optical micrographs for the cross-sections of the buffer coated particles with the flow rates of 1700 (a), 2000 (b), and 2500 sccm (c).

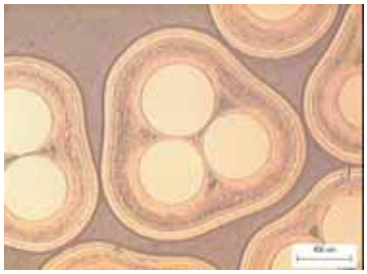

Fig. 3. Optical micrograph for the cross-sections of the agglomerated particles.

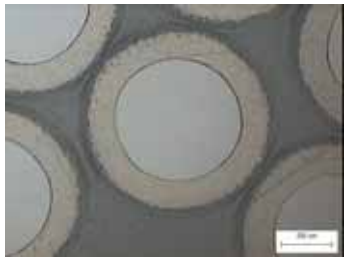

Fig. 5. Optical micrograph for the cross-section of the buffer coated particles with the C_2H_2 concentration of 50%.

be controlled properly to obtain high quality coatings because the coating rate has been known to affect significantly the microstructures of the TRISO layers [2].

The high coating rate of the buffer layer is supposed to induce the agglomeration of some particles as shown in Fig. 3. It can be seen in the microstructure that the dense PyC and SiC layers were coated in the periphery of the agglomerated particles, inferring the agglomeration had occurred during the buffer coating step. The agglomeration should be avoided because it affects detrimentally the fluidization state of the later coating steps and results in an inferior quality of the TRISO coating. In order to control the buffer coating process, the total flow rate and the concentration of the coating gas were varied at a fixed coating temperature of 1250°C.

The flow rate was varied to 1700, 2000, and 2500 sccm with a constant C_2H_2 concentration of 70%. At the flow rate of 2500 sccm, many agglomerated particles were found while the agglomeration did not occur below 1700 sccm. The coating thickness decreased as the flow rate decreased. Fig. 4 shows the microstructure of the buffer layer coated at 1250°C for 10 min with different flow rates. The coating rates were 15.2, 17.6, and 20.0 μm/min at the flow rates of 1700, 2000, and 2500 sccm, respectively. The C_2H_2 concentration was varied to 50% at the flow rate of 2000 sccm. The microstructure is shown in Fig. 5. Although the coating rate was decreased to 11.5 μm/min, the density of the buffer layer was too high. In order to accommodate the swelling of the kernel and provide void volume for gaseous fission products, the buffer layer should have enough porosity about 50%. Therefore, it is expected that the concentration of the coating gas should be higher than 70%. More detailed results of the optimization efforts for the buffer coating would be described in the presentation.

4. Conclusions

TRISO particles consisting of buffer/IPyC/SiC/OPyC layers were fabricated using a fluidized bed CVD reactor. Although most of the particles had a good coating quality, some particles were agglomerated due to an inadequate control of the buffer coating step. Optimization of the buffer coating was required to control the later coating steps properly.

REFERENCES

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