Effect of Coating Parameters of the Buffer Layer on the Shape Ratio of TRISO-Coated Particles

Weon-Ju Kim, Jong Hoon Park, Ji Yeon Park, Young Woo Lee, Jonghwa Chang
Korea Atomic Energy Research Institute, P. O. Box 105, Yuseong, Daejeon 305-600, Korea
weonjkim@kaeri.re.kr

1. Introduction

Fuel for high temperature gas-cooled reactors (HTGR's) consists of TRISO-coated particles. Fluidized bed chemical vapor deposition (FBCVD) has been applied to fabricate the TRISO-coated fuel particles. The TRISO particles 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 porous PyC coating layer, called the buffer layer, attenuates fission recoils and provides void volume for gaseous fission products and carbon monoxide. The buffer layer, which has the highest coating rate among the coating layers, shows the largest variation of the coating thickness within a particle and a batch. This could be the most plausible source of an asphericity in the TRISO particles. The aspherical particles are expected to have an inferior fuel performance. Miller et al. [2] have predicted that a larger stress is developed within the coating layers and thus the failure probability increases in the particles with high aspect ratios. Therefore, the shape of the TRISO-coated particles should be controlled properly and has been one of the important inspection items for the quality control of the fabrication process [3].

In this paper, we investigated the effect of coating parameters of the buffer layer on the shape of the TRISO particles. The flow rate of coating gas and the coating temperature were varied to control the buffer layer. The asphericity of the TRISO-coated particles was evaluated for the various coating conditions of the buffer layer, but at constant coating parameters for the IPyC/SiC/OPyC layers.

2. Experimental Procedure

The TRISO coating is produced on the 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 the bed tube. 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. For the buffer coating, the concentration of C_2H_2 was set to 70%. The total gas flow rate was varied from 1600 to 2500 sccm at coating temperatures of 1250° and 1300°C. The IPyC/SiC/OPyC layers were coated using predetermined coating gas concentrations and gas flow rates at 1300° and 1500°C for the IPyC/OPyC and the SiC layers, respectively.

After deposition of coating layers, the TRISO-coated particles were mounted in an epoxy resin and polished down to close the middle plane of the particles. Microstructures of the coated particles were observed for the polished cross-sections using an optical microscopy. The shape ratios of the particles were measured from the ratio of the maximum to the minimum diameters using an image analyzer attached to the optical microscopy.

3. Results and Discussion

Fig. 1 shows the cross-sections of the TRISO-coated particles in which the buffer layers were coated at 1250°C with different gas flow rates. When the buffer layer was coated at the flow rate of 1600 sccm, the thickness of the buffer layer was thinner and the shape of the particles was more irregular than the case of 2500 sccm. As described earlier by the current authors [4], the coating rate of the buffer layer decreases as the flow rate decreases. The average coating rates of the buffer layers were 18.9 and 29.4 μm/min for the flow rates of 1600 and 2500 sccm, respectively.

To increase the thickness of the buffer layer at the same flow rate of 1600 sccm, the coating temperature

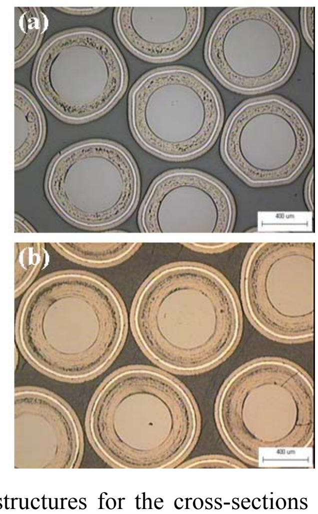

Fig. 1. Microstructures for the cross-sections of the TRISO-coated particles. The gas flow rate for the buffer coating was (a) 1600 and (b) 2500 sccm.

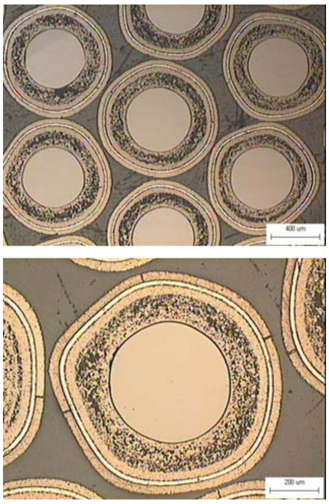

Fig. 2. Microstructures for the cross-sections of the TRISO-coated particles with the buffer layer coated at 1600 sccm and 1300°C.

was increased from 1250° to 1300°C. Fig. 2 shows the cross-sectional microstructures of the TRISO-coated particles with the buffer layer coated at 1300°C. The sphericity of the particle seems to show little change while the thickness of the buffer layer increases, being similar to Fig. 1(b). As can be seen in Figs. 1(a) and 2, the thickness of the buffer layer varies from point to point within a single particle and the asphericity of the whole particle originates from the inhomogeneity of the buffer thickness.

Fig. 3 shows the variation of the shape ratio of the TRISO-coated particles with the change of the coating conditions of the buffer layer. As can be expected from the microstructures, the shape ratio of the TRISO-coated particles decreases as the gas flow rate increases. The mechanism leading to the thickness inhomogenity of the buffer layer is not clear yet. Lackey et al. [5] observed that more uniform thickness of *dense* PyC layer (BISO type particles) could be obtained by coating in smaller charges, at higher gas flow rates and at lower temperatures. They ascribed the asphericity to

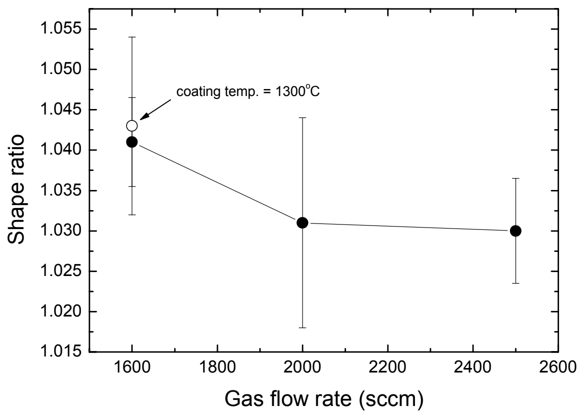

Fig. 3. Variation of shape ratio of the TRISO-coated particles by the coating parameters of the buffer layer.

the irregularity in the shape of the kernel. In this study describing the porous buffer layer, however, we could not find any relation of the kernel irregularity to the thickness variation of the buffer layer.

There is an optimum range of the gas flow rates leading to a normal spouting of particles during the coating operation. Lower and higher gas flow rates below and above the optimum range lead to a small spouting and a violent spouting of particles, respectively. One can expect that the coating reaction, which is induced by the gas-solid particle contacts during spouting, would occur most homogeneously in the normal spouting mode. The correlation between the shape ratio of the TRISO-coated particles and the simulation study of the spouting mode at room temperature using an acryl bed will be described in the presentation.

4. Conclusions

The shape ratio of the TRISO-coated particles was closely correlated with the thickness uniformity of the buffer layer. More uniform thickness of the buffer layer could be obtained at higher gas flow rates, which resulted in more spherical TRISO particles. The asphericity of particles seemed to be due to an improper control of the spouting mode during the coating operation.

REFERENCES

[1] H. Nabielek, W. Kühnlein, W. Schenk, W. Heit, A. Christ, and H. Ragoss, Development of Advanced HTR Fuel Elements, Nucl. Eng. Des., Vol. 121, p. 199-210, 1990.

[2] G. K. Miller, D. A. Petti, and J. T. Maki, Consideration of the Effects of Partial Debonding of the IPyC and Particle Asphericity on TRISO-Coated Fuel Behavior, J. Nucl. Mater., Vol. 334, p. 79-89, 2004.

[3] D. A. Petti, J. T. Maki, J. Buongiorno, R. R. Hobbins, and G. K. Miller, Key Differences in the Fabrication, Irradiation and Safety Testing of U.S. and German TRISO-Coated Particle Fuel and Their Implications on Fuel Performance, INEEL/EXT-02-00300, 2002.

[4] W. -J. Kim, J. H. Park, J. Y. Park, Y. W. Lee, and J. Chang, Effect of Fluidized Bed CVD Condition on the Microstructure of Buffer Layer in the TRISO Coated Particles, Proceedings of the Annual Meeting of the Korean Nuclear Society, May 26-27, 2005, Jeju, Korea.

[5] W. J. Lackey, W. H. Pechin, and J. D. Sease, Measurement and Control of Shape of Fuel Particles for High Temperature Gas-Cooled Reactors, Am. Ceram. Soc. Bull., Vol. 54, p. 718-724, 1975.