Effects of Processing Parameters on the Density and Microstructure of Pyrolytic Carbon

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1. Introduction

Chemical vapor deposition (CVD) of pyrolytic carbon (PyC) and silicon carbide (SiC) has been applied to TRISO-coated fuel particles for high-temperature gas-cooled reactors (HTGR). The porous PyC coating layer, called the buffer layer, attenuates fission recoils and provides void volume for gaseous fission products and carbon monoxide. The inner PyC layer acts as a containment to gaseous products. The outer PyC layer protects the SiC coating layer by inducing a compressive stress and provides chemical compatibility with a graphite matrix in the fuel compact [1]. The PyC layers undergo shrinkage due to neutron irradiation, affecting the design and modeling of fuel particles [2]. Because the dimensional change of PyC depends on the detailed microstructure of PyC, it differs from one fabrication route to another one [3]. This requires a new design of irradiation experiment applicable to spherical objects and leads to an international collaborative work called PYCASSO (PYrocarbon irradiation for Creep And Swelling/Shrinkage of Objects). KAERI proposed four different types of PyC layers coated on ZrO_2 particles, buffer with a density of 1.0 and dense PyCs with densities of 1.7, 1.9 and 2.1 g/cm^3, for the irradiation experiment.

In this study, we fabricated PyC-coated particles with various coating densities for supporting the PYCASSO experiment. We also investigated effects of processing parameters such as temperature, hydrocarbon concentration and gas flow rate on the density and microstructure of the PyC layer.

2. Experimental Procedure

PyC coatings were conducted on ZrO_2 kernels in a fluidized-bed 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 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 deposition of buffer and dense PyC were C_2H_2/Ar and C_2H_2/C_3H_6/Ar, respectively. For the buffer coating, the concentration of C_2H_2 was set to 50~80%. The total gas flow rate was varied from 1600 to 2500 sccm at coating temperatures between 1250° and 1450°C. The concentration of C_2H_2/C_3H_6, the gas flow rate and the coating temperature for the dense PyC were 20~40%, 2000 sccm and 1250°~1400°C.

Microstructures of PyC coatings were observed using an optical microscopy. Density of the coating layer was calculated by subtracting the volume and weight of kernel particles from the volume and weight of coated particles. The volume of particles was calculated from the equivalent diameter of particles measured from optical micrographs using an image analyzer.

3. Results and Discussion

Fig. 1 shows the effects of some processing parameters on the density of buffer layer. The coating temperature and the concentration of hydrocarbon significantly affect the density of buffer layer. The buffer density sharply decreases with the increase of the temperature and the C_2H_2 concentration. However, the gas flow rate has a marginal effect on the buffer density.

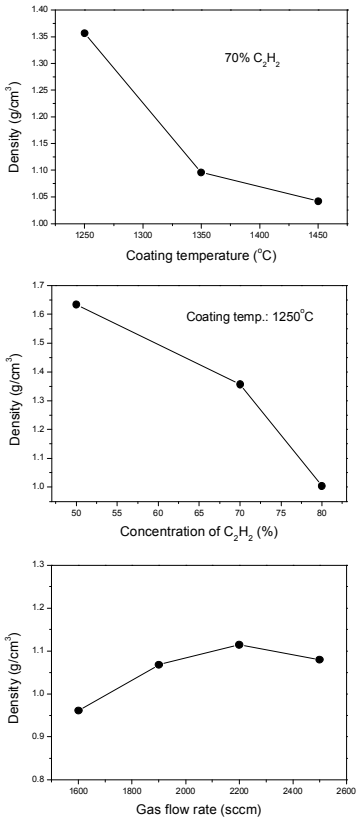

Fig. 1. Variation of the density of buffer layer as a function of the coating temperature, the concentration of C_2H_2 and the gas flow rate.

As the gas flow rate increases, the buffer density increases slightly and then decreases again above 2200 sccm.

Fig. 2 shows the density change of dense PyC with a variation of the coating temperature and the C_2H_2/C_3H_6 concentration. The two processing parameters have a similar effect on the density of the PyC layer to the case of the buffer layer. Cross-sectional optical micrographs of the PyC-coated particles with coating thicknesses of ~100 μm are shown in Fig. 3. The particles will be supplied for the PYCASSO experiment.

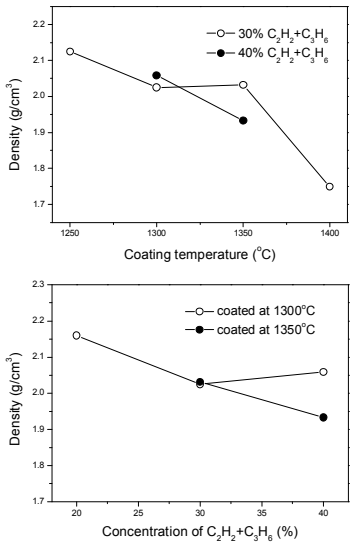

Fig. 2. Variation of the density of dense PyC as a function of the coating temperature and the hydrocarbon concentration.

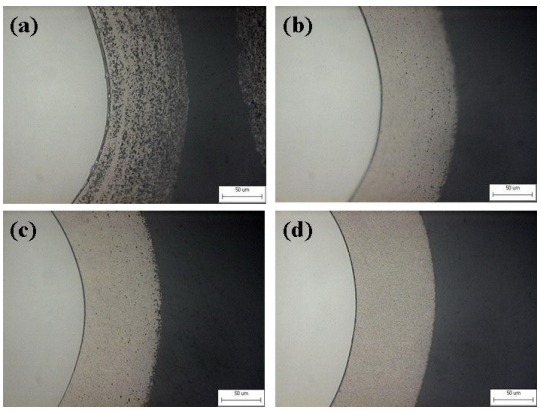

Fig. 3. Optical micrographs for the polished cross-sections of PyC-coated particles.

The effects of processing parameters on the density of PyC observed in this study are related to the mechanism of the deposition of PyC. The PyC is known to be formed from the deposition of hydrocarbon molecular species and solid spherical particles generated in the gas stream [4]. At a high degree of supersaturation of hydrocarbon molecules, where the coating rates are high, they form more spherical particles and more and larger particles are incorporated in the deposit. This leads to a lower density due to the interstices between the solid particles. On the other hand, at a low degree of supersaturation, where the coating rates are lower, there is a low probability for the formation of particles. Therefore, most of the PyC deposited on the kernel will come from molecular deposition, which will result in a higher density. Higher coating temperature and hydrocarbon concentration lead to a higher degree of supersaturation and thus a higher coating rate. As a result, the density of PyC is in inverse proportion to the coating rate as shown in Fig. 4.

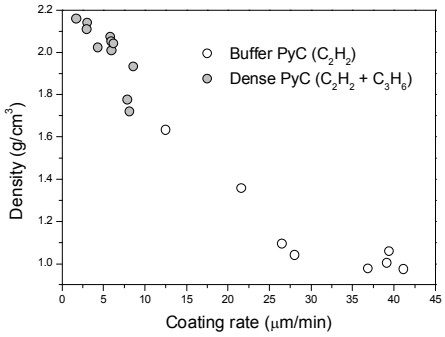

Fig. 4. Relationship between density and coating rate of PyC layers deposited in this study.

4. Conclusions

PyC-coated particles with various coating densities were successfully produced to supply particles for the irradiation study of PyC layers. Processing parameters such as coating temperature and hydrocarbon concentration had large effects on the density and microstructure of PyC, being closely related to the mechanism of the deposition of PyC.

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, pp. 199-210, 1990.
[2] J. L. Kaae, D. W. Stevens, and C. S. Luby, Prediction of the Irradiation Performance of Coated Particle Fuels by Means of Stress-Analysis Models, Nucl. Technol., Vol. 10, pp. 44-53, 1971.
[3] J. C. Bokros, Deposition, Structure, and Properties of Pyrolytic Carbon, Chem. Phys. Carbon, Vol. 5, 1-118, 1969.
[4] J. L. Kaae, The Mechanism of the Deposition of Pyrolytic Carbons, Carbon, Vol. 23, pp. 665-673, 1985.