Evaluations of the Density and the Anisotropy of Pyrolytic Carbon Prepared by Fluidized Bed CVD for TRISO Fuel

Si-Hyung Kim, Jong-Hoon Park, Weon-Ju Kim, Choong-Hwan Jung, Ji-Yeon Park, Young-Woo Lee
Korea Atomic Energy Research Institute, P. O. Box 105, Yuseong, Daejeon 305-600, Korea, exodus@kaeri.re.kr

1. Introduction

The high temperature gas-cooled reactors have utilized a TRISO coated fuel, where the fuel core (kernel) is coated with porous carbon (buffer), inner dense pyrolytic carbon (I-PyC), silicon carbide (SiC) and outer dense PyC (O-PyC) layers. A PyC is the deposit formed on a hot substrate by a thermal decomposition of a gaseous hydrocarbon and has a hexagonal structure similar to that of graphite except that the stacking order of the basal plane is essentially random [1].

The density and the microstructures of the PyC vary greatly with the coating conditions such as the temperature, gas pressure and flow rate. Several kinds of microstructures for PyC have been observed, for example, isotropic, columnar and laminar structures. The isotropic structure exhibits little or no preferred orientation, but, a laminar coating layer shows a strong preferred orientation. I-PyC and O-PyC have to be isotropic and dense to minimize the neutron irradiation effects [2]. Like this, the density and the anisotropy of a PyC layer are very important properties which influence the performance behaviors of coated fuel particles during an irradiation.

In this study, the density and the anisotropy of PyC were investigated to evaluate the coating conditions established by KAERI.

2. Methods and Results

Spherical ZrO_2 microspheres (kernel) having an average diameter of 500 um were initially coated with a buffer and then I-PyC, SiC and finally O-PyC. These coating processes were sequentially conducted in a fluidized bed system developed by KAERI. Among these layers, both I-PyC and O-PyC were deposited from a mixture of acetylene, propylene and argon at 1300^{o}C for 10 minutes. The density measurement was conducted by the density gradient technique [3] and the anisotropy degree by the OPTAF (optical anisotropy factor) method, which was proceeded by making use of the polarizing microscope-photometer system.

Fig. 1 shows the ceramographic section of the coated fuel prepared in this study. The deposition rates of I-PyC and O-PyC were 3.7 and 4.6 ㎛/min, respectively. The thicknesses of these PyC layers could be, of course, controlled by an alteration of the coating conditions.

For the density measurement, a mixed solution composed of carbon tetrachloride and ethylene-dibromide was prepared at a specific ratio and then slowly poured into the column of the density gradient apparatus (Fig. 2). At that time, several standard floats were also immersed into the solution to identify the density gradients exactly. The PyC pieces separated from the above coated fuels were dropped into the solution and their densities were evaluated in terms of their position (X) in the solution by the following equation.

$$\textit{Density at } \mathrm{X} = \mathrm{a} + \frac{(\mathrm{x}-\mathrm{y})(\mathrm{b}-\mathrm{a})}{(z-y)} \qquad (1)$$

where a and b are the densities of the two standard floats, which are at distances y and z from the arbitrary datum and x is the distance of the unknown from the same datum, intermediate between y and z. The densities of the PyC pieces were observed to meet the density specification of 1.9±0.1 g/cm^3.

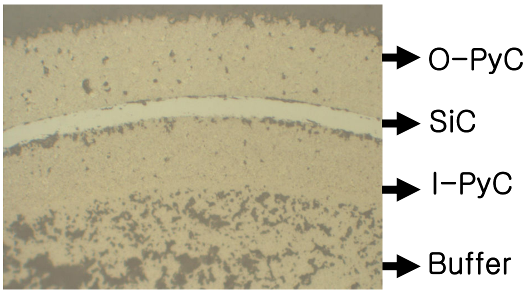

Figure 1. Ceramographic section of coated particle. The thickness of I-PyC and O-PyC layers is 37 and 46㎛, respectively.

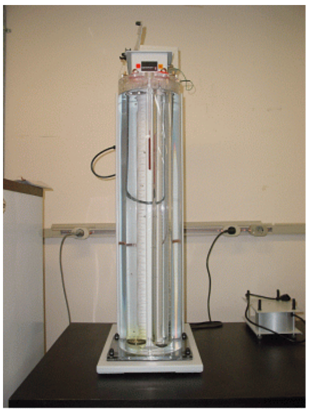

Figure 2. Density gradient apparatus.

A linear-polarized light was used for the measurement of the anisotropy degree. The polarized-light reflected from the PyC surfaces can be observed with crossed Nicols as well as without an analyzer. In the present paper, we measured the intensity values without an analyzer. The measurement of the anisotropy degree was carried out directly with the metallographic sections of the PyC layers. The sample stage of the microscope was rotated and at every 30 degrees a measurement was taken. The measurement region was always located in the center of the images during the rotation.

Fig. 3 shows the intensity values, measured by a photometer (MSP 2000, Leica), of the linear-polarized lights reflected from the PyC layers. OPTAF values were obtained by calculating the ratio of the maximum and the minimum intensity in Fig. 3. The OPTAF values of I-PyC and O-PyC were 1.03 and 1.04, respectively. According to Sawa et al., OPTAF in a high density PyC layer should be less than 1.04 to prevent an excessive deformation by a fast neutron irradiation [2].

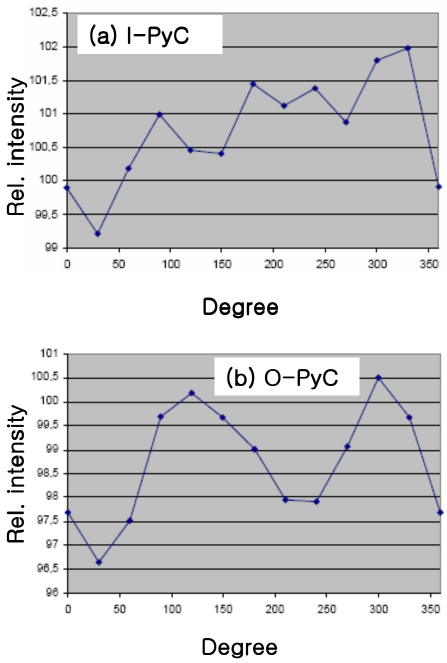

Figure 3. The relative intensity of the polarized light reflected from the I-PyC and O-PyC layers. The detected area is 10x10 microns and the interference filter, 546 nm, was used to exclude the effect of light source.

The anisotropy of the PyC layers is commonly characterized by the BAF factor, but it is not proper for a spherical coated fuel because a measurement of the preferred orientation by X-ray needs flat specimens. So, optical methods have been widely used because it is directly measured from a section of the spherical particles and experimentally simpler.

Gruebmeier derived a relation between BAF and OPTAF [4].

$$BAF = \frac{2(1-\gamma)\cdot(1/2+OPTAF)}{(1-\gamma\cdot OPTAF)} - 2 \qquad (2)$$

where the value of γ is 0.25 which is acquired from a graphite. According to Eq. (2), the BAF values of I-PyC and O-PyC were 1.09 and 1.12, respectively. U.S. demanded that the BAF values of the PyC layers should be below 1.2.

It is generally known [5] that high temperatures and high coating-gas concentrations result in high coating rates and thereby produce PyC layers with a low density and isotropic structures. On the other hand, low temperatures and low coating rates tend to create a PyC layer which has an anisotropic laminar structure and a high density. Therefore, it is considered that the coating conditions for the PyC were excellent according to our results such as the density and the anisotropy.

3. Conclusion

The evaluation techniques for measuring the density and the anisotropy degree of PyC layers were established in this study. The densities of the PyC layers were measured by the density gradient technique and they were confirmed to meet the density specification ($1.9\pm0.1 g/cm^3$). The results of the anisotropy measurement showed that the PyC layers had isotropic structures. From these results, it was considered that the coating conditions (coating temperatures, coating gas concentrations et al.) for the PyC were successfully established.

Acknowledgement

This work has been performed under the Nuclear R&D program by the Ministry of Science and Technology.

REFERENCES

[1] R.J. Gray and J. V. Cathcart, Polarized Light Microscopy of Pyrolytic Carbon Deposits, Journal of Nuclear Materials, Vol.19, pp.81-89, 1966.

[2] K. Sawa, J. Sumita, S. Ueta, S. Suzuki, T. Tobita, T. Saito, K. Minato, T. Koya and H. Sekino, Integrity Confirmation Tests and Post-irradiation Test Plan of the HTTR First-Loading Fuel, Journal of Nuclear Materials, Vol. 38, No. 6, pp. 403-410, 2001.

[3] ASTM Designation : D1505-03, Standard Test Method for Density of Plastics by the Density-Gradient Techniques.

[4] H.B. Grubmeier and G.P. Scheidler, An Optical Method for the Determination of the Local Anisotropy of Pyrolytic Carbon Layers and Graphite, ORNL-TR-2127.

[5] D.A. Petti, J. Buongiorno, J.T. Maki, R.R. Hobbins, and G.K. Miller, Key Differences in the Fabrication, Irradiation and High Temperature Accident Testing of US and German TRISO-Coated Particle Fuel and Their Implications on Fuel Performance, Nuclear Engineering and Design, Vol. 222, pp. 281-297, 2003.