Density Measurement of Silicon Carbide Layer and Outer Pyrolytic Carbon layer of Simulated TRISO-Coated Particles by Using a Density Gradient Column under an IAEA CRP Program

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1. Introduction

The TRISO-coated fuel particle for a HTGR(high temperature gas-cooled reactor) is composed of a nuclear fuel kernel and outer coating layers[1,2]. The O-PyC coating layer protects the primary barrier, SiC layer. The density of SiC layer as well as the density of O-PyC layer is measured to evaluate the coating layers. For measuring densities, SiC layer fragments are acquired by oxidizing the broken coated particles. The O-PyC fragments are acquired from the broken coated particles. The fragments are so small and irregular that it is not easy to measure the weight and volume of the SiC and O-PyC fragments. The density gradient column and standard floats are usually used to measure such a small fragment[3-6]. In this experiment, the SiC specimens and the O-PyC specimens of 4 kinds of simulated TRISO-coated particles with ZrO_2 kernel were measured by a density gradient column with a density gradient solution under the IAEA CRP-6 (coordinated research project 6) program for characterizing TRISO-coated fuel particles for HTGR through the round robin tests with 4 different samples distributed from 4 member organizations to the 9 participating organizations.

2. Density Gradient Column

The model of the density gradient column used for this study is DC/02 made by LLOYD Instruments Ltd.[6] The density gradient solution should be made in consideration of the density of the sample to be measured. We tried to make a solution with density gradient ranging from 1.10 g/cc of lower density to 1.60 g/cc of higher density for sample A, a solution with density gradient ranging from 1.72 g/cc of lower density to 2.18 g/cc of higher density for sample B and C, and a solution with density gradient ranging from 1.60 g/cc to 1.98 g/cc for sample D.

The density of SiC layer is often less than 3.2 g/cc. We tried to make a solution with density gradient ranging from 3.10 g/cc of lower density to 3.30 g/cc of higher density for sample A, B, C and D.

200 cc of gradient solution for each sample was made as shown in Table 1. Table 2 shows the density of each liquid.

The density range of the density gradient solution for density measurement of O-PyC layers can be evaluated by using standard floats as shown in Fig. 1.

Table 1. Solutions to measure the density of each layer in a density gradient column.

Coating layer	OPyC			SiC
Sample	A	B, C	D	A, B, C, D
Lower density, g/cc	1.10	1.72	1.60	3.10
Upper density, g/cc	1.60	2.18	1.98	3.30
Lower density liquid	Ethanol 66 cc	Carbon-tetra-chloride 80 cc	Carbon-tetra-chloride 100 cc	Bromoform 50 cc
Upper density liquid	Carbon-tetra-chloride 134 cc	Dibromo-ethane 120 cc	Dibromo-propane 100 cc	Methylene iodide 150 cc

Table 2. Density of liquid.

Liquid	Density, g/cc
Ethanol	0.79
Carbon-tetrachlorid	1.60
Dibromopropane	1.98
Dibromoethane	2.18
Bromoform	2.90
Methylene iodide	3.30

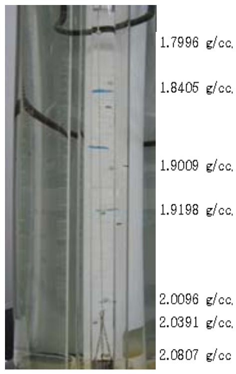

Fig.1. A gradient density solution for density measurement of O-PyC layers of coated particles for specimen B and C.

The density D_x of the sample is calculated by equation (1)[6].

$$D_x = a + \frac{(x-y)(b-a)}{(z-y)}, \qquad (1)$$

where a and b are the densities of the two standard floats, which are at distance y and z, and x is the distance of the unknown sample from the same datum, located between y and z.

3. Density measurement of O-PyC and SiC layer

O-PyC fragments were prepared to measure the densities from the simulated TRISO-coated particles with ZrO_2 kernel instead of UO_2 kernel. The fragments of the broken coating layers consist of SiC layer and PyC layer. The O-PyC layer fragments were selected carefully among these fragments. The prepared test samples were put into the density gradient solution of the column. The positions of specimens were measured after some time to come to equilibrium as shown in Fig. 2. The densities of specimens were 1.435 g/cc for sample A, 1.893 g/cc for sample B, 2.006 g/cc for sample C and 1.615 g/cc for sample D, respectively as shown in Table 3.

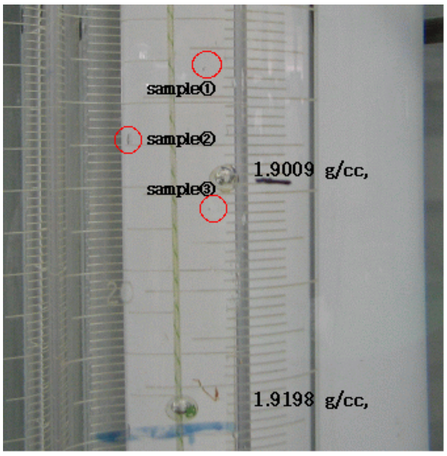

Fig.2. Standard float and O-PyC fragments of sample B floated in a density gradient column.

Table 3. Densities of O-PyC fragments floated in a density gradient column.

Samples	A	B	C	D
Average density	1.435	1.893	2.006	1.615
Standard deviation	0.004	0.009	0.027	0.017

The PyC layer was removed from the SiC layer by oxidizing the fragments at the temperature of 850 °C for 3 hours in an oxidation furnace. The prepared test samples were put into the density gradient solution of the column. The densities of specimens were 3.201 g/cc for sample A, 3.192 g/cc for sample B, 3.197 g/cc for sample C and 3.198 g/cc for sample D as shown in Table 4.

Table 4. Densities of SiC fragments floated in a density gradient column.

Samples	A	B	C	D
Average density	3.201	3.192	3.197	3.198
Standard deviation	0.003	0.002	0.002	0.002

4. Conclusion

In this experiment, the densities of O-PyC layer and SiC layer of the coated particles were measured by a density gradient column with various mixtures of density gradient solutions. The experimental results are as follows.

- Density gradient solution were prepared in the density gradient column for each O-PyC sample with different density range.
- The densities of the specimens were 1.435 g/cc for sample A, 1.893 g/cc for sample B, 2.006 g/cc for sample C and 1.615 g/cc for sample D.
- The density gradient solution ranging from 3.10 g/cc of lower density to 3.30 g/cc of higher density was prepare by mixing bromoform and diodomethane in a density gradient column.
- The densities of the SiC specimens ranged from 3.192 g/cc to 3.201 g/cc.
- The measured SiC densities were higher than the low limitation, 3.18 g/cc, for the HTR-10. The measured O-PyC densities for sample A and C were lower than the low limitation, 1.80 g/cc.

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