

Error Analysis of Ceramographic Sample Preparation for Coating Thickness Measurement of Coated Fuel Particles

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Abstract – *The thicknesses of four coatings of HTR coated fuel particle are very important parameters. It is indispensable to control the thickness of four coatings of coated fuel particles for the safety of HTR. A measurement method, ceramographic sample-microanalysis method, to analyze the thickness of coatings was developed. During the process of ceramographic sample-microanalysis, there are two main errors, including ceramographic sample preparation error and thickness measurement error. With the development of microscopic techniques, thickness measurement error can be easily controlled to meet the design requirements. While, due to the coated particles are spherical particles of different diameters ranged from 850 to 1000 μ m, the sample preparation process will introduce an error. And this error is different from one sample to another. It's also different from one particle to another in the same sample. In this article, the error of the ceramographic sample preparation was calculated and analyzed. Results show that the error introduced by sample preparation is minor. The minor error of sample preparation guarantees the high accuracy of the mentioned method, which indicates this method is a proper method to measure the thickness of four coatings of coated particles.*

Key words: Coated fuel particle, coating thickness, ceramographic sample preparation, error analysis

I. INTRODUCTION

High temperature gas cooled reactor (HTR) has been considered as one of the most promising advanced nuclear energy systems because of its inherent safety features [1, 2]. The safety of pebble bed-type HTR is based on the quality of the spherical fuel elements, each of which contains tens of thousands of coated fuel particles. Coated fuel particles consist of a micro spherical kernel of uranium dioxide (UO₂), coating layers of porous pyrolytic carbon (buffer), inner dense pyrolytic carbon (IPyC), silicon carbide (SiC) and outer dense pyrolytic carbon (OPyC) [3-5]. The principal functions of these coating layers are to retain fission products within the particles. The porous pyrolytic carbon coating layer provides storage space for fission gas to keep the inner pressure in a certain range, control kernel expansion during fission process and prevent inner dense pyrolytic carbon from the damage of nuclear fission fragments. The SiC coating layer acts as a barrier against the

diffusive release of metallic fission products and provides mechanical strength for the particles. The inner and outer dense pyrolytic carbon layers protect the SiC coating layer and stop release of fission products. The parameters of coated fuel particles are very important to the safe operation of a pebble bed high temperature gas-cooled reactor. The thicknesses of four coatings of coated fuel particle are the important parameters of coated fuel particle. Therefore, it is of great significance to measure the thicknesses of four coatings rapidly and accurately. A measurement method, ceramographic sample-microanalysis method, to analyze the thickness of coatings has been developed [6, 7]. This method can measure the four coatings of coated fuel particles rapidly and efficiently. However, during the ceramographic sample preparing process and measurement process, errors would be introduced [8]. Both of the errors in sample preparation process and measurement process need to be evaluated. With the development of microscopic techniques,

thickness measurement error can be easily controlled to meet the design requirements. While, due to the coated particles are spherical particles of different diameters, the sample preparation process will introduce errors. And these errors can't be measured directly. In this paper, these errors were analyzed and calculated. Results show that the errors introduced by ceramographic sample preparation are minor. The minor errors guarantees high accuracy of ceramographic sample-microanalysis method, which indicates this method is a proper method to measure the thickness of four coatings of coated particles.

II. THE DESCRIPTION OF CERAMOGRAPHIC SAMPLE-MICROANALYSIS METHOD

Ceramographic sample-microanalysis method is a method to measure the thicknesses of four coatings for HTR coated fuel particle.

II.A. Ceramographic Sample Preparation

Before measurement, ceramographic sample of coated fuel particles is prepared first. The coated fuel particles are fixed to one plane of the columnar sample, then grinding and polishing the sample until most of the particles near to equatorial plane.

II.B. Measurement Method of the Thickness of Four Coatings

After the ceramographic sample is obtained, put the sample on the microscope stage, then, focus and photograph the particles in the sample. The obtained images are processed and measured by image analysis software, and the thicknesses are obtained.

III. ANALYSIS OF ERRORS IN CERAMOGRAPHIC SAMPLE-MICROANALYSIS METHOD

Both the sample preparing process and measurement process may introduce errors. These errors include, grinding damage at each layer edge, different diameter of particles, layer thickness variation in a same particle and between any two particles, and analysis error in identifying each layer edge. In this part, all of the above errors were analyzed.

III.A. Analysis of Errors in Sample Preparing Process

During sample preparing process, errors are introduced by grinding damage at each layer edge and size distribution. During grinding process, damage may appear at each layer edge. This damage would introduce error. However, by controlling grinding parameters, particles with grinding damage

would be rare, though it can't be completely avoided. Thus, this source of error could be eliminated by only choosing the particles without grinding damage as measurement samples.

Different diameter of particles is another source of error during sample preparation process. As shown in Fig. 1, because of different diameter of particles, it's impossible to grind and polish all the particles to equatorial plane simultaneously. Error would be directly introduced while the grinding surface has a distance from equatorial plane. This error can't be avoided and also can't be measured. Thus this error was calculated by assuming the particles were ideal sphere.

When the particles were ideal sphere, either grinding surface haven't reach equatorial plane or grinding surface have been over equatorial plane would introduce error to measurement process. Because each particle is plane symmetry, introduced error is determined by the distance of grinding surface to equatorial plane. No matter whether grinding surface hasn't reach equatorial plane or grinding surface have been over equatorial plane, error has been determined as long as the distance between grinding surface and equatorial plane is certain. Therefore, we take the situation of grinding surface having not reached equatorial plane to discuss the error in sample preparing process.

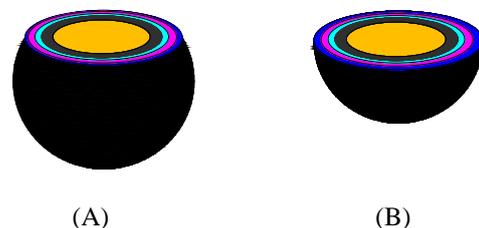


Fig. 1: Schematic diagram of prepared sample (A. grinding surface haven't reach equatorial plane; B. grinding surface and equatorial plane were coincide)

As Fig.2 shows, when the measurement surface had a distance with equatorial plane of coated particle, the measure value would be different with true value. The measurement value could be calculated using equation1:

$$t'_i = \sqrt{r_i^2 - d^2} - r'_{i-1} \quad (\text{eq. 1})$$

where t'_i is the measurement value of the thicknesses of four coatings ($i=1, 2, 3, 4$ represent porous pyrolytic carbon, inner dense pyrolytic carbon, silicon carbide layer, outer dense pyrolytic carbon, respectively.); d is the distance of grinding surface to equatorial plane; r_i and r'_i are true radius and measurement radius of coated particle, respectively. ($i=1, 2, 3, 4$ represent monolayer coated particle, double layer coated particle, three layer coated particle and four layer coated particle, respectively.)

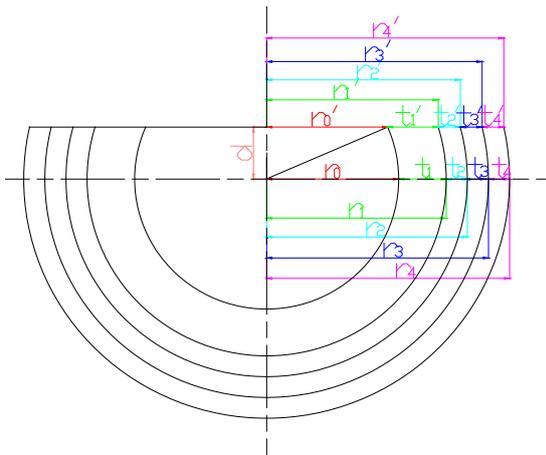


Fig.2: Schematic diagram of sample preparing error

Errors of the thicknesses for coatings are calculated as follows:

$$e_i = |t_i - t'_i| \quad (\text{eq. 2})$$

Where e_i is the errors of the thicknesses for four coatings; t_i is the real value of the thicknesses of four coatings ($i=1, 2, 3, 4$ represent pyrolytic carbon, inner dense pyrolytic carbon, silicon carbide layer, outer dense pyrolytic carbon, respectively).

To simplify analysis process, particle with kernel diameter of 500 μm , porous pyrolytic carbon (buffer) of 90 μm , inner dense pyrolytic carbon (IPyC) of 40 μm , silicon carbide (SiC) of 35 μm , outer dense pyrolytic carbon (OPyC) of 40 μm , was taken as specimen to calculate the introduced errors. Calculated errors were listed in Table 1.

Distanc e/ μm	Buffer / μm	IPyC/ μm	SiC/ μm	OPyC/ μm
5	0.01	0	0	0
10	0.05	0.02	0.01	0.01
15	0.12	0.03	0.02	0.02
20	0.21	0.06	0.04	0.04
25	0.33	0.10	0.07	0.07
30	0.48	0.14	0.10	0.10
35	0.66	0.19	0.14	0.13
40	0.86	0.25	0.18	0.17
45	1.09	0.32	0.23	0.22
50	1.35	0.39	0.28	0.27

Table 1: Measurement errors of four coatings with various distances of grinding surface to equatorial plane

In practical production process, particle size ranges from 850 to 1000 μm . After ceramographic sample is obtain, the particles in the sample had different distances of grinding surface to equatorial plane for different particles. The error in sample preparing process was calculated by analyzing percentage of each particle diameter range and the error corresponding to each particle diameter range.

The percentage of each particle diameter range for batch 1 and batch 2 was listed in table 2 and shown in Fig. 3. As shown in Fig. 3, both of particle size for batch 1 and batch 2 distributed in nearly normal distribution. Most particles concentrated in the range of 880-950 μm for batch 1 and most particles concentrated in the range of 900-950 μm for batch 2. Particle size distribution influences sample preparing error directly. The sample preparing error was weighted average of maximum error corresponding to each particle diameter range.

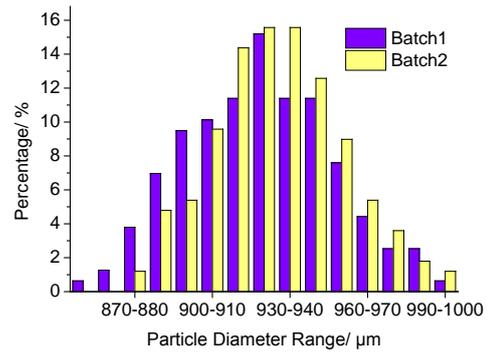


Fig. 3: Particle size distribution of two batches

Diameter Range/ μm	Batch 1/%	Batch 2/%
850-860	0.6	0
860-870	1.3	0
870-880	3.8	1.2
880-890	7.0	4.8
890-900	9.5	5.4
900-910	10.1	9.6
910-920	11.4	14.4
920-930	15.2	15.6
930-940	11.4	15.6
940-950	11.4	12.6
950-960	7.6	9.0
960-970	4.4	5.4
970-980	2.5	3.6
980-990	2.5	1.8
990-1000	0.6	1.2

Table 2: Particle size distribution of two batches

In ideal status, all of the particles have the same diameter, and every particle is tangency to all the particles around it in equatorial plane. However, during practical process, only most of particles are tangency to particles around it in grinding surface. During sample preparing process, microscope is employed to observe particles in sample. By microscope observation during grinding and polishing process, tangent particles are obtained as much as possible. From statistical data in Table 2, we can find that grinding depth could be easily controlled in the range of 450 μm to 470 μm by observing particle tangency condition. Therefore, errors of sample preparation for two batches were

analyzed with grinding depth ranged from 450 μm to 470 μm .

The maximum distances of grinding surface to equatorial plane for each range of particle diameter at grinding depth of 450 μm , 460 μm and 470 μm were listed in Table 3.

Diameter Range/ μm	Grinding depth/ μm		
	450	460	470
850-860	25	35	45
860-870	20	30	40
870-880	15	25	35
880-890	10	20	30
890-900	5	15	25
900-910	5	10	20
910-920	10	5	15
920-930	15	5	10
930-940	20	10	5
940-950	25	15	5
950-960	30	20	10
960-970	35	25	15
970-980	40	30	20
980-990	45	35	25
990-1000	50	40	30

Table 3: The maximum distances of grinding surface to equatorial plane at different grinding depth

Item	Grinding depth/ μm	Error/ μm	Max Error/ μm
Buffer	450	0.23	0.23
	460	0.14	
	470	0.18	
IPyC	450	0.07	0.07
	460	0.04	
	470	0.05	
SiC	450	0.05	0.05
	460	0.03	
	470	0.04	
OPyC	450	0.04	0.04
	460	0.03	
	470	0.04	

Table 4: Sample preparing errors for batch 1 at different grinding depth

Weighted average of sample preparation errors is calculated by corresponding data in Table 1, Table 2 and Table 3. Obtained weighted averages of four coatings at grinding depth of 450 μm , 460 μm and 470 μm were listed in Table 4 and Table 5, of which Table 4 listed calculated results for batch 1 and Table 5 listed calculated results for batch 2.

From calculated results of sample preparation, we can find that sample preparation errors of four coatings are all small. Errors for buffer layer are maximum. For batch 1, it's 0.23. And for batch 2, it's 0.25. While, errors of the other three layers are all smaller than 0.1 μm . The results show that during

sample preparing process, errors are introduced, while the errors are small.

Item	Grinding depth/ μm	Error/ μm	Max Error/ μm
Buffer	450	0.25	0.25
	460	0.13	
	470	0.13	
IpyC	450	0.07	0.07
	460	0.04	
	470	0.04	
SiC	450	0.05	0.05
	460	0.02	
	470	0.02	
OPyC	450	0.05	0.05
	460	0.02	
	470	0.02	

Table 5: Sample preparing errors for batch 2 at different grinding depth

III.B. Analysis of Errors in Measurement Process

During measurement process, errors are introduced by layer thickness variation in a same particle and between any two particles, and analysis error in identifying each layer edge. Layer thickness variation in a same particle and between any two particles is a source of error performed during measurement process. To minimize this error, thicknesses of different directions for hundreds of particles are measured. Average thickness value and standard deviation are performed to show thickness range of each layer. Different sample number is corresponding to different standard deviation expression. By measuring enough samples and using corresponding standard deviation expression, the effect of this error can be ignore.

Analysis error in identifying each layer edge is another source of error performed during measurement process. Figure 4 shows a measurement image of a coated fuel particle. As shown in figure 4, thickness is the distance between internal boundary and outer boundary. During measurement process, the distance is performed by distance of two pixels, which are at internal boundary and outer boundary, respectively. Both the unclear layer edge and pixel size may introduce analysis error in identifying each layer edge. It's obvious that each layer edge of the particle is clear. Thus this error was directly affected by pixel sizes.

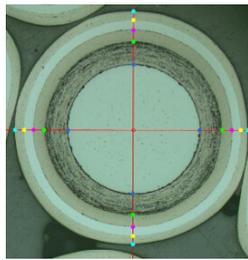


Fig.4: Measurement image of a coated fuel particle

Figure 5 is a schematic diagram of pixel sizes at different magnifications. At lower magnification, square pixel size is larger than that at higher magnification. As the magnification increases, square pixel size decreases. Table 6 shows pixel sizes of images at different magnifications. Pixel sizes of 100 times magnification, 200 times magnification, 500 and 1000 times magnification were $0.54\mu\text{m}$, $0.27\mu\text{m}$, $0.11\mu\text{m}$, $0.05\mu\text{m}$, respectively. So measurement accuracy increases with the microscope magnification increasing.

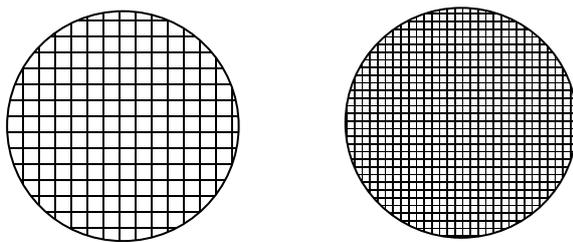


Fig. 5: Schematic diagram of pixel sizes at different magnifications

Magnification	Pixel size/ μm
100	0.54
200	0.27
500	0.11
1000	0.05

Table 6: Pixel sizes at different magnifications

IV. CONCLUSION

Ceramographic sample-microanalysis method was developed to analyze the thickness of coatings. This method can measure the thickness of coatings visual and fast. However, during the process of ceramographic sample-microanalysis, there are two main errors, including ceramographic sample preparation error and thickness measurement error. Both of them were analyzed.

(1) Two batches of coated particles, with particle size ranged from 850 to $1000\mu\text{m}$, were used to analyze sample preparing errors of four coatings. Analyzed data displays that errors for buffer layer are maximum. For batch 1 and batch 2, they are 0.23 and 0.25 , respectively. While, errors of the other three layers are all smaller than $0.1\mu\text{m}$. The results

show that during sample preparing process, introduced errors are smaller and acceptable.

(2) During measurement process, measurement errors decreases with the microscope magnification increasing. At 100 times magnification, 200 times magnification, 500 and 1000 times magnification, measurement errors were $0.54\mu\text{m}$, $0.27\mu\text{m}$, $0.11\mu\text{m}$, $0.05\mu\text{m}$, respectively.

Ceramographic sample-microanalysis method have high accuracy. Errors introduced in sample preparing process and measurement process are both smaller and acceptable. Therefore, this method can meet the requirements of accuracy and efficiency. It is a proper method for engineering application to measure the thickness of the four coatings for coated particles.

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