Optimized manufacturing process of large TRISO fuel particle using surrogate kernel

Doeun Kim[1,3], Soon Ik Hong[1], Hyeon-Geun Lee[2], Eung Sun Kim[3]*
[1] Materials Science & Engineering, Chungnam National University, Daejeon 34134, Korea
[2]Nuclear Materials Development Division, Korea Atomic Energy Research Institute, Daejeon 34051, Korea
[3] Next-Generation Fuel Technology Development Division, Korea Atomic Energy Research Institute, Daejeon 34051, Korea
hglee@kaeri.re.kr

1. Introduction

One of the main technologies for VHTR (Very High Temperature Reactor), TRISO (TRi-ISOtropic) particle is a spherical nuclear fuel surrounded by four layers. The TRISO particle is located in the center of the fuel and coated in a continuous layer of Buffer, I-PyC, Silicon Carbid (SiC) and O-PyC around it to prevent fission products from be release out of the particle. In experiment, the surrogate kernel uses zirconium oxide (500 μm diameter, ZrO_2) because the existing nuclear fuel, uranium oxide (UO_2), is at risk as a radioactive substance [1]. ZrO_2 has similar physical and thermal properties to UO_2 [2,3] and it has the advantage of being non-radioactive, making handling far easier.

TRISO particle can be used manufacture to FCM (Fully Ceramic Microencapsulated) for LWR's (Low Water Reactor) nuclear fuel. FCM is a nuclear fuel that has improved accident resistance, such as excellent oxidation resistance and ability to possess fission product, consisting of TRISO particles and SiC matrix.

The nuclear fuel may be applied to LWR, to increased loading or size of particles for the same efficiency and volume fraction as the existing nuclear fuel UO_2 [4]. The experimental conditions for manufacturing TRISO particle was confirmed by increasing the diameter of the surrogate kernel ZrO_2 to 800 μm. To investigate the fluidization of 800 μm ZrO_2 kernel at FB-CVD (Fluidized-Bed CVD), the required gas flow rate was identified compared to 500 μm ZrO_2.

The spouting height evaluation of spherical particles under various conditions newly defined the fluidized velocity relationship according to the density and diameter. To find a new surrogate kernel with conditions similar to UO_2, replacing ZrO_2 with the low density. The new surrogate kernel was used to identify the process optimization conditions for the manufacture of large TRISO particle.

2. Experimental

2.1 The Spouting Height Measurement Materials

When the kernels reach the hot zone in the chamber, deposition of each coating layer is possible. The visualization equipment of Fig 1 has been installed for use in evaluation, the gas flow rate and the spouting height required for each kernel.

The visualized experimental equipment used a conical bed, which is the similar form to a conical geometry coater. Fig 2 shows the geometric factor of these spouted bed. The dimensions of these bed are : diameter of the upper cylindrical section, D_c, 25 mm, the bed angle, r, 60°; the height of the conical section, H_c, 15 mm; the gas inlet diameter used are, D_0, 3 mm. and the base diameter D_i, 3.6 mm [5].

Figure 1. Visualization experiment equipment

After loading the particles into the bed, it supplied a certain amount of the fluid gas to prevent them from falling to the bottom of the bed. The particles start spouting by flow gas, measure the height of the bubbling of the particles through a laser pointer located at the top of the bed. The height evaluation of particles in a high temperature environment due to the heat supply system outside the column used particles of the same condition.

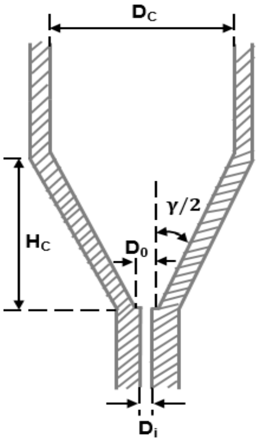

Figure 2. Geometry of the conical spouted bed reactor

The type and conditions of the surrogate kernel used in the experiment are as shown in Table 1. The gas flow rate and the height measurements required to spouting each kernel with the constant diameter to the same height were performed.

Kernel	Density(g/cm^3)	Diameter(μm)
Al_2O_3	3.8	500, 700, 800
Ti	4.5	
ZrO_2	5.7	
Steel	7.9	
Ag	10.5	
WC	15.6	

Table 1. Types and condition of spherical particles

2.2 TRISO Particle Coating

The manufacture of TRISO is generally applied by the FB-CVD to coat small spherical particles uniformly. A mixture of C_2H_2 or $C2H_2$+$C3H_6$ is mainly used as the reaction gas for PyC, and MethyTrichloroSilane (CH_3SiCl_3, MTS)+H_2 is used as the reaction gas for SiC. The chemical deposition reaction is carried out by injecting the Ar gas through the nozzle to maintain uniform fluidized bed formation in object to be coated [6,7,8]. Detailed experimental conditions for 800 μm ZrO_2 kernel and the new surrogate kernel, Tungsten Carbide (WC) are specified in Table2. In the experiment, all the surrogate kernels used 14g of the same mass.

3. Results and Discussions

3.1 Minimum Fluidization Flow Rate

Minimum spouting velocity is important operating parameters in spouted bed performance. The influence of temperature over the hydrodynamics of cylindrical spouted beds has been studied by several paper [9]. Through the visualization equipment, the characteristics of fluidization of particles at room temperature and high the fluidization of particles at room temperature and high temperature were explored.

In Fig 3, assessed has the flow rates for each mass to spout ZrO_2 of small diameter and large diameter at the same height. The evaluation was conducted at room temperature and the gas flow rate increased linearly with increasing diameter. 800 μm showed the same fluidization behavior only when it supplied twice the gas flow rate compared 500 μm ZrO_2. The results at RT are expected to look similar even at high temperatures, setting the experimental conditions as shown in Table 2(A).

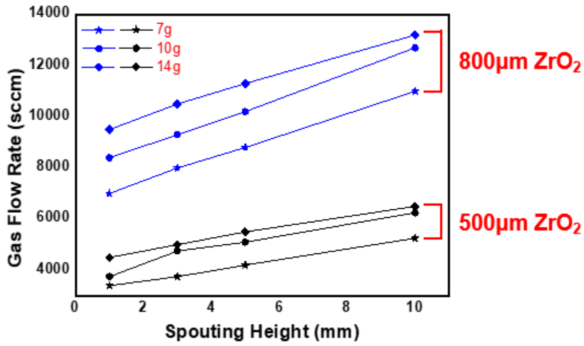

Figure 3. 500 μm and 800 μm ZrO_2 spouting height

The relationship of fluidization characteristic according to the density and the diameter of various particles were identified. In Fig 4, Measured by two standard, the degree of gas flow required linearly increases as the diameter or density of the kernel used increases. The linear regression analysis was shown to define a new flow relationship between spherical particles.

Based on the data, the required gas flow rate for 800 μm ZrO_2 and the new surrogate kernel WC bubbling tot the same point as the existing 500 μm ZrO_2 is set. The difference in flow rate is about four times greater. Other detailed experimental conditions are as shown in Table 2(B).

A	Ar (sccm)	H_2 (sccm)	MTS (sccm)	C_2H_2 (sccm)	C_3H_6 (sccm)	T_{dep} (℃)	time (min)
Buffer	800	-	-	3200	-	1460	45s
I-PyC	2800	-	-	600	600	1410	3m
SiC	2664	1998	54	-	-	1560	13m
O-PyC	3800	-	-	815	815	1410	50s

B	Ar (sccm)	H_2 (sccm)	MTS (sccm)	C_2H_2 (sccm)	C_3H_6 (sccm)	T_{dep} (℃)	time (min)
Buffer	1800	-	-	7200	-	1450	15s
I-PyC	5600	-	-	1200	1200	1450	2m
SiC	7400	5500	150	-	-	1560	20m
O-PyC	8400	-	-	1800	1800	1450	2m

Table 2. Detailed experimental conditions (A) 800 μm ZrO_2 (B) 700 μm WC

3.2 The Surrogate Kernel Coating

800 μm ZrO_2 kernel confirmed through the height evaluation results that it needs two times the gas flow rate compared to the small ZrO_2 kernel. By applying this, large TRISO particles were manufactured and the cross section is in Fig 5. In order to find the optimized process conditions, the deposition temperature, time and the reaction gas ratio were changed several time. The three inner layers were deposited with the proper porosity and thickness.

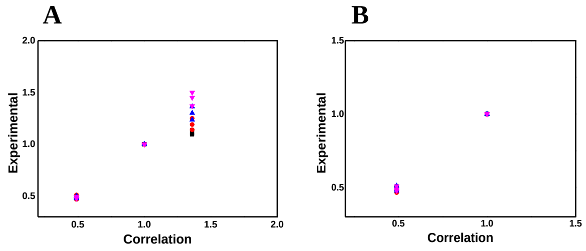

Figure 4. (A) Spouting height (B) Total height

However, the Fig 5 shows that the O-PyC layer has a rather high porosity and poor coating quality. Along with the increased size of the ZrO_2 kernel and the weight of particles increases as the SiC layer is deposited, the flow rate required for smooth flow is insufficient. Due insufficient fluidization, the low-quality coating layers are expected to be depleted as particle do not reach the area where coating takes place properly.

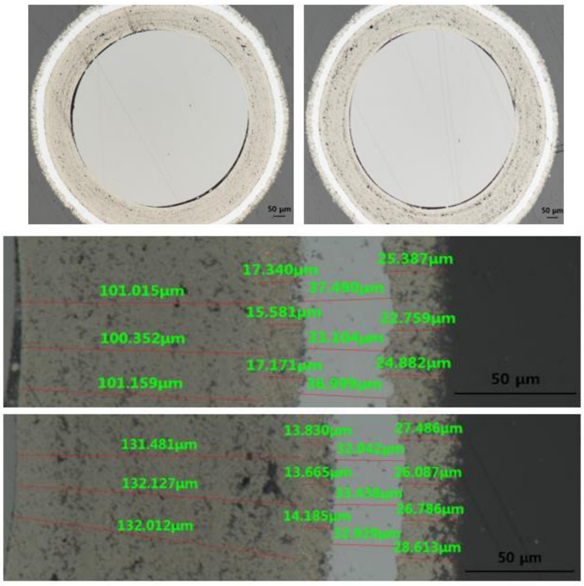

Figure 5. OM image of 800 μm ZrO_2 kernel TRISO

WC (700 μm diameter), the most similar condition among spherical particles with the same volume ratio as the existing UO_2, has been adopted as the new surrogate kernel. WC with density 15.6 g/cm^3 has a heavier weight than ZrO_2 and UO_2, so more gas is needed for fluidization.

Based on the flow characteristic experiment, the gas flow rate was about four times higher than 500 μm ZrO_2, and each layer was deposited to coat TRISO particle. Section are shown in Fig 6. Due to the high density of the particle, a small amount exists in the area of the co-mmon angle when the same mass as ZrO_2 kernel is charged. Therefore, the gas supply amount is increased, it is determined that the behavior of the kernel in a high-temperature environment is not the same. The particles are coated evenly throughout, but the thickness deviation exists.

Because the boundary between the innermost layers is not clear, variables need to be adjusted, such as consideration of this particle and changes in the deposition time or the response gas rate.

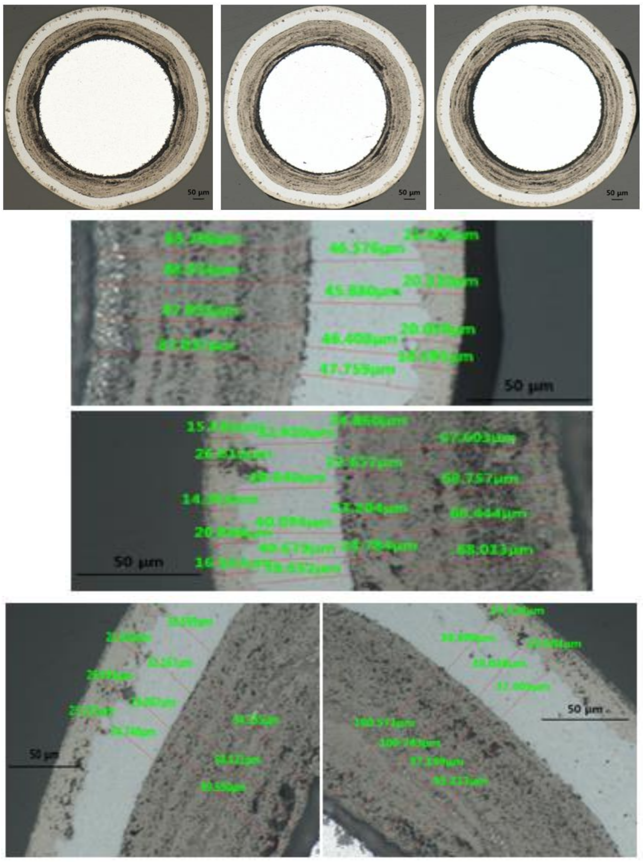

Figure 6. OM image of 700 μm WC kernel TRISO

4. Summary

The Spouting height evaluation was conducted according to the gas flow rate for the spherical kernels with different densities by constant diameter. The gas flow rate for each kernel required to form spouting to the same location was recorded. The change in gas flow rate for the variable of diameter or density was identified and the fluidized relationship in conical spouted bed was newly defined.

Baesd on Minimum spouting velocity [9] and research results, it is planning to supplement the fluidization relationship and conduct various experiments with TRISO particle coating using the large kernel in various direction.

Through the results of the spouting experiment, 800 μm ZrO_2 kernel confirmed that the gas flow rate is about twice 500 μm ZrO_2. The thickness and quality of each coating layer were evaluated after coating large TRISO particles by applying the increase the gas flow rate.

WC, which has been adopted as a new surrogate kernel, must supply about four times the gas flow rate compared to 500 μm ZrO_2 due to the high density. The wide increase in weight resulted in some differences in the fluidized behavior at high temperatures in the range of 1400 to 1500℃.

Both the increased size kernel and the new surrogate kernel generally performed successful deposition test. For the more successfully compensate for the thickness and coating quality, plan to experiment by considering various variables in the course of further process.

REFERENCES

[1] V.T. Gotovchikov, V.A. Seredenko, V.V. Shatalov, V.N. Kaplenkov, A.S. Shulgin, V.K. Saranchin, M.A. Borik, C.W. Forsberg, Melted and Granulated Depleted Uranium Dioxide for Use in Containers for Spent Nuclear Fuel, IHLRWM 2006, Las Vegas, NV, April 30–May 4, 2006.

[2] Zirconium oxide properties, http://www.accuratus.com/zirc.html

[3] J.K. Fink, Thermo-physical properties of uranium dioxide, J. Nucl. Mat. 279 (2000) 1–18.

[4] Powers, Jeffrey J., et al. "Fully ceramic microencapsulated (FCM) replacement fuel for LWRs." Oak Ridge: ORNL (2013).

[5] Sukarsono, R., S. Riyadi, and D. Husnurrofiq Sri Rinanti. "The Selection of Geometry and Flow Rate on The Fluidized Bed Reactor for Coating Particle." Journal of Physics: Conference Series. Vol. 1198. No. 2. IOP Publishing, 2019.

[6] K. Minato, T. Ogawa and K. Fukuda, "Review of experimental studies of zirconium carbide coated fuel particles for high temperature gas-cooled reactor," JAERI-Review 95-004 (1995).

[7] S.Ueta, H.Ino, M.Takahashi and K.Sawa, "Plan of Development of ZrC-TRISO coated fuel particle and construction of ZrC coater,"JAERI-Tech 2002-085 (2002)

[8] K.Minato, K.Fukuda, "Chemical vapor deposition of SiC for coated fuel particles," J. Nucl. Mater., 149, 233-246 (1987)

[9] Olazar, M., et al. "Minimum spouting velocity under vacuum and high temperature in conical spouted beds." The Canadian Journal of Chemical Engineering 87.4 (2009): 541-546.