Optimized manufacturing process of large TRISO fuel particle using surrogate kernel

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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 Carbide (SiC) and O-PyC around it to prevent fission products from 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, $\alpha$, 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].
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.

### 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$_2$H$_2$ or C$_2$H$_4$+C$_3$H$_6$ is mainly used as the reaction gas for PyC, and MethylTrichloroSilane (CH$_3$SiCl$_3$, 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 Table 2. 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 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).

#### Table 1. Types and condition of spherical particles

<table>
<thead>
<tr>
<th>Kernel</th>
<th>Density(g/cm$^3$)</th>
<th>Diameter(μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al$_2$O$_3$</td>
<td>3.8</td>
<td>500, 700, 800</td>
</tr>
<tr>
<td>Ti</td>
<td>4.5</td>
<td></td>
</tr>
<tr>
<td>ZrO$_2$</td>
<td>5.7</td>
<td></td>
</tr>
<tr>
<td>Steel</td>
<td>7.9</td>
<td></td>
</tr>
<tr>
<td>Ag</td>
<td>10.5</td>
<td></td>
</tr>
<tr>
<td>WC</td>
<td>15.6</td>
<td></td>
</tr>
</tbody>
</table>

#### 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 to 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).

#### Table 2. Detailed experimental conditions (A) 800 μm ZrO$_2$ (B) 700 μm WC

<table>
<thead>
<tr>
<th>A</th>
<th>Ar (sccm)</th>
<th>H$_2$ (sccm)</th>
<th>MTS (sccm)</th>
<th>C$_2$H$_2$ (sccm)</th>
<th>C$_3$H$_6$ (sccm)</th>
<th>T$_{dep}$ (℃)</th>
<th>time (min)</th>
</tr>
</thead>
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<tr>
<td>Buffer</td>
<td>800</td>
<td>-</td>
<td>-</td>
<td>3200</td>
<td>-</td>
<td>1460</td>
<td>45s</td>
</tr>
<tr>
<td>I-PyC</td>
<td>2800</td>
<td>-</td>
<td>-</td>
<td>600</td>
<td>600</td>
<td>1410</td>
<td>3m</td>
</tr>
<tr>
<td>SiC</td>
<td>2664</td>
<td>1998</td>
<td>54</td>
<td>-</td>
<td>-</td>
<td>1560</td>
<td>13m</td>
</tr>
<tr>
<td>O-PyC</td>
<td>3800</td>
<td>-</td>
<td>-</td>
<td>815</td>
<td>815</td>
<td>1410</td>
<td>50s</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>B</th>
<th>Ar (sccm)</th>
<th>H$_2$ (sccm)</th>
<th>MTS (sccm)</th>
<th>C$_2$H$_2$ (sccm)</th>
<th>C$_3$H$_6$ (sccm)</th>
<th>T$_{dep}$ (℃)</th>
<th>time (min)</th>
</tr>
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<tbody>
<tr>
<td>Buffer</td>
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<td>-</td>
<td>-</td>
<td>7200</td>
<td>-</td>
<td>1450</td>
<td>15s</td>
</tr>
<tr>
<td>I-PyC</td>
<td>5600</td>
<td>-</td>
<td>-</td>
<td>1200</td>
<td>1200</td>
<td>1450</td>
<td>2m</td>
</tr>
<tr>
<td>SiC</td>
<td>7400</td>
<td>5500</td>
<td>150</td>
<td>-</td>
<td>-</td>
<td>1560</td>
<td>20m</td>
</tr>
<tr>
<td>O-PyC</td>
<td>8400</td>
<td>-</td>
<td>-</td>
<td>1800</td>
<td>1800</td>
<td>1450</td>
<td>2m</td>
</tr>
</tbody>
</table>
3.2 The Surrogate Kernel Coating

800 μm ZrO₂ kernel confirmed through the height evaluation results that it needs two times the gas flow rate compared to the small ZrO₂ 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.

Based on the flow characteristic experiment, the gas flow rate was about four times higher than 500 μm ZrO₂, 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 common angle when the same mass as ZrO₂ 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 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 ZrO₂ 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 ZrO₂ kernel TRISO

WC (700 μm diameter), the most similar condition among spherical particles with the same volume ratio as the existing UO₂, has been adopted as the new surrogate kernel. WC with density 15.6 g/cm³ has a heavier weight than ZrO₂ and UO₂, so more gas is needed for fluidization.

### 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.

Based 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 \( \mu m \) ZrO\(_2\) kernel confirmed that the gas flow rate is about twice 500 \( \mu m \) 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 \( \mu m \) 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\(^\circ\)C.

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