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A Preliminary Study on the Manufacture of Spherical ZrO_2 Particle and Fuel Preform with Complex-shape

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1. Introduction

As 3D printing technology is generalized, interest in research to replace nuclear reactor parts produced by traditional manufacturing processes with parts manufactured by AMM (advanced manufacturing methods) is increasing in the nuclear fuel field [1]. In the nuclear fuel manufacturing field, research on manufacturing a fuel element having a small and complex structure using ceramic oxide powder is being actively researched. Basic research is underway to apply the BJ (binder jet) method as a way to manufacture Ceramic-Metal (CERMET) composites using oxides such as UO_2 powder as raw materials [2]. In this connection, research on the manufacture of fuel preforms with complex shapes using BJ 3D printing is being actively conducted with spherical ZrO_2 powder prepared by simulating uranium oxide powder.

In this study, the manufacturing of spherical ZrO_2 powder as a simulated powder for manufacturing complex-shaped fuel element preforms and the results of physical property analysis of the manufactured ZrO_2 powder were briefly presented. Also, the results of manufacturing a fuel element preform manufactured by a BJ 3D printer [3] using the prepared ZrO_2 powder are briefly shown.

2. Experiments

In this study, by using the spray drying process [4] which widely used in the food industry was selected as a method for producing spherical ZrO_2 powder of less than 100μm. The basic concept of spray drying process can be expressed as shown in the Fig. 1 [5]. Commercial nano-sized ZrO_2 powder was purchased, dissolved in distilled water, and thus a raw material solution for spray drying process was prepared to have a slurry concentration of 30 to 70%.

The spray drying equipment is simply shown on the right side of Fig.1, and a solution obtained by adding an appropriate amount of PVA and a small amount of carboxylic acid as a surfactant to the zirconia slurry solution to have a uniform composition was used in order to prepare the spherical fine ZrO_2 particles.

Table 1 briefly shows the preparing conditions of complex-shaped nuclear fuel element preforms using the BJ 3D printer and the manufacturing of spherical ZrO_2 particles obtained from prepared-intermediates and heat treatment from the manufacturing of spherical droplets.

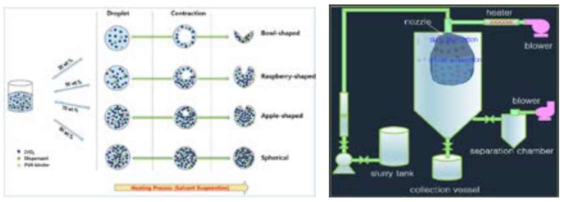

Fig. 1. Spray drying concept and simple equipment.

Table 1. Preparation conditions of the spherical fine ZrO_2 powder

Preparation parameters	Contents
- Binder	Poly Vinyl Alcohol - M.W. : PVA 205 - volume : ~1% of powder
- Surfactant(dispersion)	Carboxyl acid - volume : ~0.8% of powder
- Dryer	Diameter : 1,000Φmm - Inlet temperature : 200℃ - Outlet temperature : 100℃
- Disc rotation speed	rpm : ~10,000

3. Results and discussion

3.1 ZrO_2 powder preparation

In an initial study on the production of ZrO_2 powder using a spray drying equipment, spherical particles were prepared using zirconium acetate salt solution as a raw material. The zirconium hydrate intermediate was produced with an irregular shape, but the raw material was changed to a zirconium hydroxide salt solution due to the unpleasant odor generated by acetate during the preparing process. Fine spherical ZrO_2 powder was prepared under the preparation conditions of table 1 by using the 30 to 70% slurry solution and the spray drying apparatus.

In Fig. 2, the shape of the ZrO_2 particles obtained by changing the slurry concentration of the raw material solution to prepare the intermediate material and then going through the subsequent heat treatment process is briefly shown.

Looking at the state of the particles, it was found that in the case of 30%, the fine particle grains were loosely combined on the surface to produce fluffy particles. When the particles were produced by increasing the slurry concentration to 70%, it was found that almost spherical shape ZrO_2 particles were produced, and it was found that they were shown with mono-modal particle size distribution.

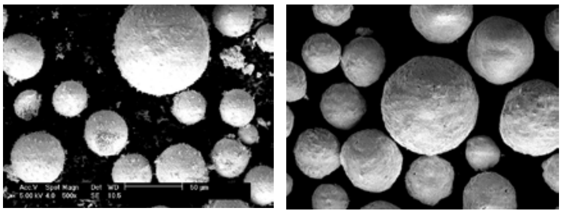

Fig. 2. Particle shape of ZrO_2 powder obtained from different slurry concentration (left: 30%, right:70%).

3.2 Powder characteristics

To identify the physical properties of spherical ZrO_2 particles manufactured by the spray drying process, the particle size distribution and porosity of the particles obtained under each manufacturing condition were analyzed and shown in Fig. 3. According to the analysis of the particle size distribution of spherical ZrO_2 particle prepared by changing the concentration of zirconia slurry solution to 30% to 70%, it was found that the bimodal size distribution was obtained when 30% solution was used. But in case of the 70% solution, it was prepared as particles having a mono-modal particle size distribution.

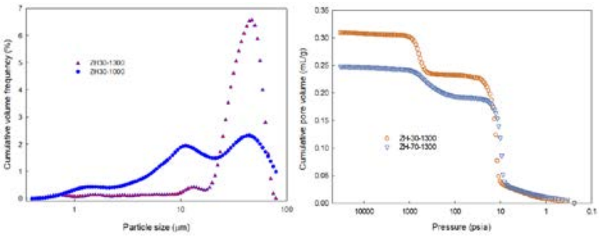

Fig. 3. Particle size distribution and porosity of ZrO_2 powder obtained from different condition.

Meanwhile, looking at the pore distribution of the prepared-ZrO_2 particles, it was analyzed that the total pore volume decreased when the ZrO_2 slurry concentration of the raw material solution was increased. Additional experiments are needed to prepare particles with a particle size distribution and pore volume suitable for the BJ 3D printing process.

3.3 Preform manufacture

Nuclear fuel element preforms of complex-shapes were manufactured using a BJ 3D printer. 3D drawings STL files were made and preform specimens were manufactured using the ExOne innovent+ 3D printer [6] using simulated ZrO_2 powder. The size of the specimen is 160x65x65 mm, the print resolution is X/Y: 63.5/60 ㎛, Z: 100㎛, and the layer thickness is 100㎛. The simple specifications of printing system and equipment are shown in the Fig. 4.

It was judged that the ZrO_2 powder used for preform manufacturing had good preform manufacturability when using D_{50} = 25~50μm particle size with almost no pores inside. When printing to a layering thickness of about 100μm, it is determined that powder pushing-phenomena in the printer can be prevented. Also, in order to manufacture a preform having a sound state, setting the saturation degree of the printing binder to 50% showed good results.

Manufacture	ExOne(USA)
Equipment	Innovent+
Specimen size	160x65x65 mm
Resolution	X/Y:63.5/60.0㎛, Z:100㎛
Layer thickness	100㎛

Fig. 4. BJ 3D printer specification and equipment.

In this study, the shape of the preform obtained in the preliminary manufacturing experiments for a fuel element preform using simulated spherical ZrO_2 powder is briefly shown in the Fig. 5.

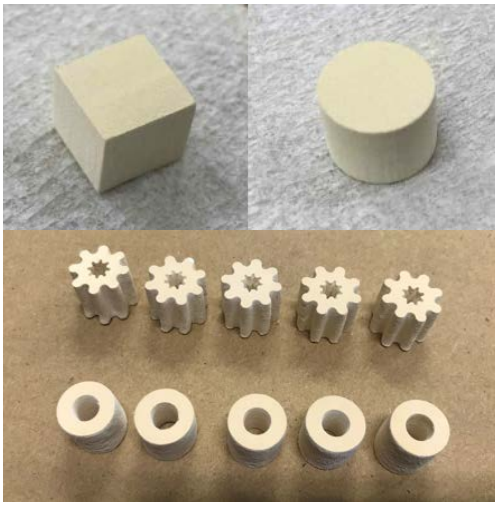

Fig. 5. Simulated ZrO_2 fuel element preforms with cubic, cylindrical, and complex shapes.

3.4 Waste powder and preform recycle process from the BJ printing

A large amount of UO_2 waste is generated in the process of manufacturing complex-shaped nuclear fuel element using the BJ process. A large amount of waste-powder and waste-preform with binder are generated, and their recycling is very meaningful from an economic and environmental point of view.

The actual manufacturing process of UO_2 powder could be prepared using the existing sol-gel process or spray drying process. In this research, the basic concept of a spray drying process was developed to recycle raw material powder or waste preform, which is a valuable material, from the BJ 3D printing. Fig. 6 shows the basic concept and a simple flow diagram for recovering and recycling raw material powder from waste powder and waste preform generated in the BJ 3D printing process in this study.

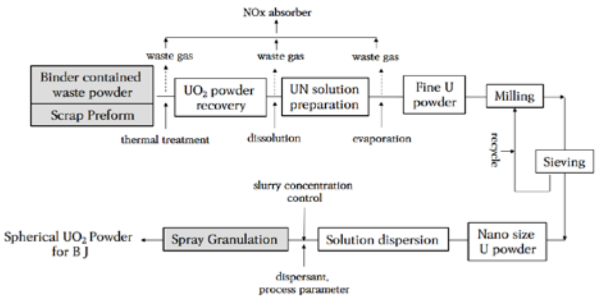

Fig. 6. Simple recycling process on waste powder and preform from BJ 3D printing fabrication.

4. Conclusion

Preliminary waste recycle studies were conducted to manufacture complex-shaped nuclear fuel element preform using oxide raw material powder. Considering domestic and foreign research environments, a technical analysis of BJ 3D printing technology was performed, and ZrO_2 powder was selected to simulate UO_2 powder. In addition, a spray drying process was selected to recycle waste generated during the manufacture of oxide preforms using BJ 3D printing.

A spherical ZrO_2 fine powder having good properties was prepared using the process, and the properties of the prepared powder were analyzed. The results of preliminary manufacturing complex-shaped fuel preform using the simulated ZrO_2 powder by the BJ 3D printing were also well prepared according to the purpose of this research.

In the future, based on the preliminary research results of this study, additional research on the development of the optimal ZrO_2 powder manufacturing process and the manufacturing of complex-shaped preforms is needed.

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