Spherical ADU and UO_3 Intermediate Particles Preparation

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1. Introduction

Nowadays, nuclear energy has been spotlight as the countermeasure for an immense electricity supply and a clean hydrogen gas production over the next a few decades. With these circumstances, concentrated studies and discussions have already been progressed on the R&D of a HTGR[1]. Generally, the nuclear fuel of a HTGR is TRISO(TRistructural ISOtropic) coated fuel particles which are enclosed by three layers of coating materials, such as pyro-carbons and silicon carbide, at the surface of spherical UO_2 kernels.

The production processes of a spherical UO_2 kernel consists of the following steps (Figure.1)[2,3]:

1. $UN(UO_2(NO_3)_2)$ solution preparation from a dissolution of uranium oxide powder (UO_3 or U_3O_8) with HNO_3
2. pre-neutralization of UN solution with NH_4OH solution
3. broth solution preparation by mixing of UN and the organic additives
4. liquid-ADU droplets preparation using the broth solution into a vibrating nozzle system
5. ADU gelation
6. conversion from dried-ADU to UO_3 by a calcination furnace
7. heat treatments for the reduction and sintering processes from UO_3 to UO_2 by a sintering furnace

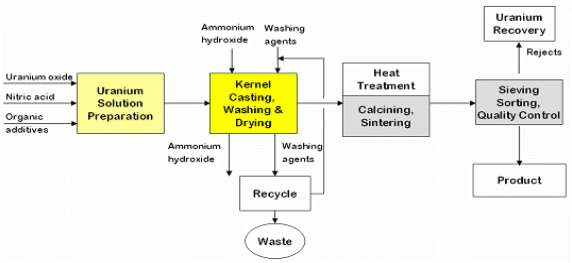

Fig.1. Flow diagram for a UO_2 kernel preparation.

In this study, the preparation methods of spherical ADU gel particles *via* liquid-ADU droplets, and the intermediate of a HTGR nuclear fuel, were investigated and then, the thermal characteristics of an ADU→UO_3 conversion were studied through heat treatments in a calcination furnace. The shapes of the liquid droplets, ADU gels and UO_3 particles were observed with a Streoscope.

2. Technical Background and Experimental

The production of a spherical UO_2 kernel can be carried out by wet chemical processes such as a sol-gel method, based on a solidification of spherical liquid-ADU droplets prepared by a vibrating nozzle system.

Generally, in the case of a HTGR nuclear fuel, the production process of a UO_2 kernel uses to the gel-supported precipitation method among the many techniques. After making a broth solution containing an uranyl nitrate as a uranium source, the spherical liquid-ADU droplets are prepared by using a vibrating nozzle system. Figure 2 shows the experimental apparatus for the preparation of the spherical liquid-ADU droplets and the ADU gel particles.

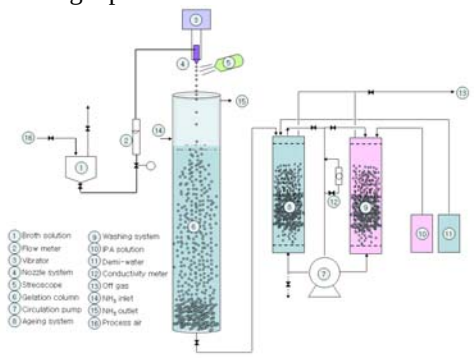

Fig.2. Experimental apparatus for ADU gel particles production.

The ADU gel particles obtained from the above experiments are transported to the AWD (Ageing-Washing-Drying) tank for the ageing and washing steps, by using an ammonia solution, distilled water, and isopropyl alcohol, respectively. Finally, the dried-ADU particles are obtained from a slightly vacuumed condition in an air atmosphere.

Then the dried-ADU gel particles are converted to UO_3 and UO_2 particles respectively, by heat treatments in calcination and sintering furnaces.

3. Results

3.1 Preparation of Spherical Liquid-ADU Droplets and ADU Gel Particles

The main units for the preparation of spherical liquid-ADU drops are:

- broth solution storage vessel and feeding system
- flow meter (single) and vibrating nozzle system
- air/NH_3-inlet and air/NH_3-outlet for a pre-hardening of a liquid droplet surface
- gelation column

By providing air pressure to the broth storage vessel, the broth solution enters the vibrating nozzle system with a constant flow. After achieving a round shape in an air atmosphere, the spherical liquid-ADU droplets fall through an ammonia gas atmosphere in which a

pre-hardening occurs. Then the spherical liquid-ADU droplets fall into the ammonium hydroxide solution in the gelation column, here the solidification of the spherical liquid-ADU droplets is completed.

To keep the ammonia concentration required for a constant stream in the gelation column, a fresh ammonium hydroxide solution is fed into the gelation column when the liquid-ADU droplets are falling. An expansion reservoir accommodates the overflow solution. The kernels are collected in the collecting vessel, then transferred to the ageing, washing and drying steps.

Figure 3 show photographs of the spherical liquid-ADU droplets and ADU gel particles obtained from our vibrating nozzle system.

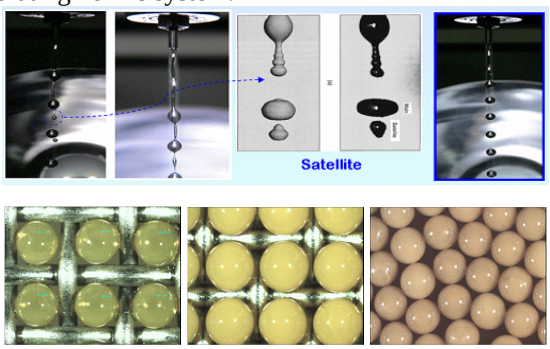

Fig.3. Spherical liquid-ADU droplets and ADU gel particles.

In this step, if the relation between the flow (feeding) rate of the broth solution and the frequency/amplitude of the vibrator is not discordant, the small satellite droplets are formed as shown in the left photograph of Figure 3[4]. This phenomenon of a different size of the droplets is because the feeding rate of the broth solution is so high that the natural laminar jet flow of a steady state was broken at the nozzle tip.

As a result, the sizes of the spherical liquid-ADU droplets and dried-ADU gel particles were obtained at about 1900~1950 μm and 950~1000 μm, respectively, at nearly the same size spheres. This droplets size is about four times bigger than that of the final UO_2 particle, and this value is suitable according to other discussions.

3.2 Heat Treatments and Its Characteristics

A calcination furnace is commonly used in the dried-ADU→UO_3 conversion step. Our heat treatment experiments for a calcination of the dried ADU particles were carried out by raising the temperature till 430 °C in an air atmosphere. Heating rate is a very important factor for obtaining no cracked spherical particles. If the heating rate is faster than 5 °C /min., the dried-ADU gel particles are cracked or broken. This is caused by the exothermic heats which occur from a fast thermal decomposition of the PVA among the ADU constituents. So the heating rate in the calcination process must be kept below 5 °C /min. Figure 4 shows the heating mode for a calcination of our heat treatment experiments and the shapes of the UO_3 particles converted according to changes of the heat treating mode.

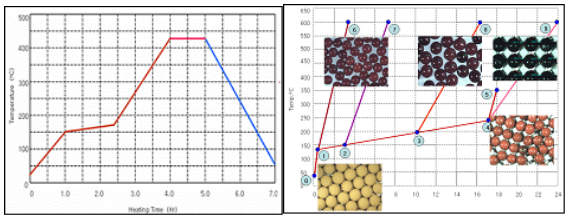

Fig.4. Heating mode and UO_3 particles.

Otherwise, to obtain the optimum condition of sintering process, pre-sintering experiments are carried out by TG/DTA instrument. Test conditions are in 4%H_2-Ar mixture gas atmosphere, and 3 step heating modes were progressed till 1600 °C.

- 1'st reduction : 25 → 700 °C (5°C/min. heating) and 1 hr isotherm
- 2'nd heating and sintering : 700 → 1600 °C (3 °C/min. heating) and 4 hrs isotherm
- 3'rd cooling : 1600 °C → room temperature (5°C/min. cooling rate)

Figure 5 shows the heating mode for a sintering from UO_3 to UO_2 of our heat treatment experiments and the particle shapes of the sintered-UO_2 particles obtained according to the optimum heat treating mode.

Fig.5. Sintering mode and UO_2 particles.

4. Conclusion

In this study, to obtain a spherical UO_2 kernel, the most important factors in the droplets preparation are the composition ratio of the broth solution, and the harmony of the flow (feeding) rate of the broth solution and the frequency/amplitude of the vibrator. Also the heating rate in the calcination process must be kept below 5 °C /min., and the optimum conditions of the sintering were in a 4%H_2-Ar mixture gas atmosphere and a 3 steps heating mode till 1600 °C.

REFERENCES

[1] INEEL/EXT-05-02581, Next Generation Nuclear Plant Research and Development Program Plan, Idaho National Engineering and Environmental Laboratory, p.1, 2005.
[2] G. Brambilla, P. Gerontopulos, and D. Neri, The SNAM Process for the Preparation of Ceramic Nuclear Fuel Microspheres : Laboratory Studies, Energia Nucleare, 17(4), p. 217, 1970.
[3] M.Kadner and J.Baier, Production of Fuel Kernels for High-Temperature Reactor Fuel Elements, Kerntechnik, 18, p.413, 1976.
[4] P. A. Hass, Formation of Uniform Liquid Drops by Application of Vibration to Laminar Jets, Industrial Engineering Chem. Res., 31, p. 959, 1992.