Small Size UO_2 Microsphere Preparation for a HTGR Nuclear Fuel

Jeong Kyung Chai+, Kim Yeon Ku, Oh Seung Chul, Kim Young Min, Kim Woong Ki,
Lee Young Woo, and Cho Moon Seoung,
HTGR Fuel Technology Development Division, Kaeri, Daejeon 305-353, Korea
kcjeong@kaeri.re.kr

1. Introduction

Generally, the nuclear fuel of a HTGR has used TRISO(TRistructural ISOtropic) coated fuel particles which is enclosed by the three layers of coating materials, such as pyrolytic carbon(PyC), as buffer layer, inner dense PyC(IPyC) layer, silicon carbide(SiC) layer, and outer PyC(OPyC) layer at the surface of spherical UO_2 kernels as shown in Figure 1.

Fig.1. The structure of the TRISO particle.

The key process in a HTGR nuclear fuel is a spherical UO_2 kernel preparation process[1]. Since the 1970s, many methods for a spherical UO_2 kernel particle preparation have been investigated based on the sol-gel process (internal and external gelations), and a suspension process with a temporary binder, and a mixed process(sol-gel process and suspension). We selected the gel-supported precipitation processes. Sol-gel processes in the nuclear fuel preparation field have many advantages at a high purity atmosphere and low processing temperature[2].

In this study, the preparation methods of the spherical liquid-ADU droplets, the first material of a HTGR nuclear fuel, were investigated first and then, the thermal characteristics of the ADU→UO_3 and UO_3→UO_2 conversions were studied through heat treatments.

2. Technical Background and Experimental

In the case of a HTGR nuclear fuel, the preparation process of a UO_2 kernel uses to a gel-supported precipitation, a kind of sol-gel process, method among the many chemical processes. After making a casting solution, a raw material solution, containing an uranyl nitrate[$UO_2(NO_3)_2$] as a uranium source, the spherical liquid-ADU droplets are made by using a vibrating nozzle system. Figure 2 shows a photograph of the experimental apparatus for making the spherical liquid-ADU droplets and the overall process for the TRISO coated particles production.

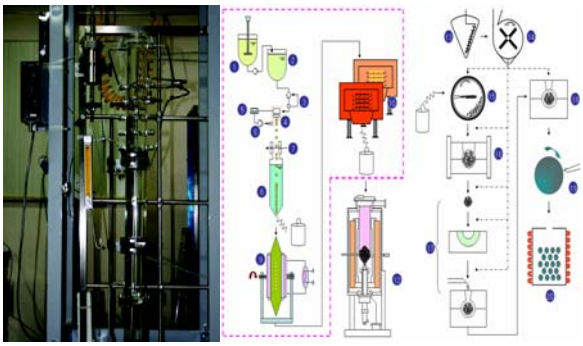

Fig.2. Experimental apparatus for ADU gel particles production and simple overall process for TRISO coated particle.

The ADU gel particles obtained from the above experiments are transported to the AWD (Aging-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 by a calcination furnace and a sintering furnace, respectively.

3. Results

3.1 FT-IR Characteristics of ADU Gel Particles

FT-IR has prove to be a most useful tool for a structural determination and identification of organic and inorganic compounds[3]. Figure 3 shows the FT-IR spectrum in short wave number of the dried ADU compound particles in the KBr pellet.

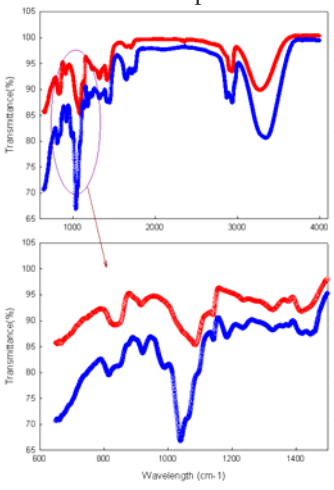

Fig.3. FT-IR spectra of the ADU gel particles.

Generally, the ADU compound particle exhibited absorption bands associated with a stretching and the bending vibrations of H-O-H in the coordinated water and hydrated water at 3370 and 3150 cm^{-1}, stretching and bending vibrations of N-H in NH_4^+(sharp) and U-O(middle), one sharp peak and another middle peak at 1400 and 1100 cm^{-1} respectively.

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, small satellite droplets occurred. 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 of 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.

Figure 4 shows the photographs of the spherical liquid-ADU droplets and ADU gel particles obtained from our vibrating nozzle system.

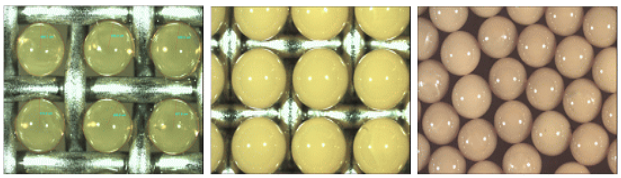

Fig.4. Spherical ADU gel particles.

3.2 Thermal Treatments

Calcination and sintering furnaces are commonly used in the dried-ADU→UO_3 and UO_3→UO_2 conversion steps, respectively. Our heat treatment experiments for a calcination of the dried ADU particles were carried out by raising the temperature till 450 °C in an air atmosphere. Heating rate is a very important factor for obtaining no cracked spherical particles[4]. If the heating rate is faster than 3 °C /min., the dried-ADU gel particles are cracked or broken. This is caused by the exothermic heat which occurs from a rapid thermal decomposition of the PVA among the ADU constituents. So the heating rate in the calcination process must be kept below 3 °C /min.

Otherwise, the heat treatment for the sintering process was carried out till 1650°C in a 4%-H_2 atmosphere with the sintering furnace. The sintering furnace reduces UO_3 to UO_2 using a hydrogen reduction atmosphere. Furthermore, the furnace sinters the UO_2 in order to achieve dense UO_2 kernels.

The following chemical reduction reaction takes place :

$UO_3(s) + H_2(g) \rightarrow UO_2(s) + H_2O(g)$

To obtain the optimum condition of the sintering process, pre-sintering experiments are carried out by TG/DTA instrument. Test conditions are in a 4%H_2-Ar mixture gas atmosphere, and 3 step heating modes which were progressed till 1650 °C.

Figure 5 shows the heating modes for a calcination and a sintering of our heat treatment experiments and the shapes of the UO_3 and UO_2 particles converted according to changes of the heat treating modes.

- 1'st reduction : 25 → 700 °C (5°C/min. heating) and 1 hr isotherm
- 2'nd heating and sintering : 700 → 1650 °C (3 °C/min. heating) and 4 hrs isotherm
- 3'rd cooling : 1650 °C → room temperature (5°C/min. cooling rate)

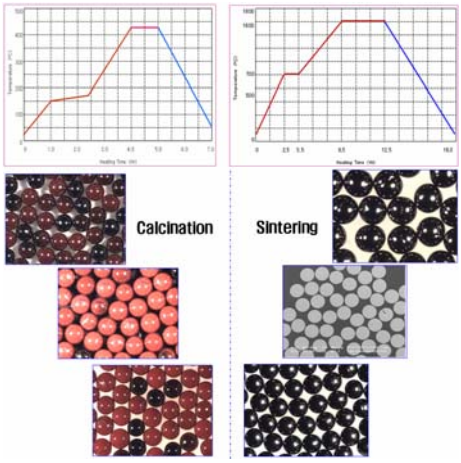

Fig.5. Heating modes and UO_3 /UO_2 particles.

4. Conclusion

In this study, to obtain a spherical UO_2 kernel, the most important factors for the droplets preparation are the composition ratio of the broth solution, and the viscosity of the broth solution, and the frequency/amplitude of the vibrator. Also the heating rate in the calcination process must be kept below 3 °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 1650 °C.

REFERENCES

[1] 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.

[2] M.Kadner and J.Baier, Production of Fuel Kernels for High-Temperature Reactor Fuel Elements, Kerntechnik, 18, p.413, 1976.

[3] A.R.Nyquist and R.O.Kagel, Infrared Spectra of Inorganic Compounds, Academic press, Newyork, 1971.

[4] P.Naefe and E.Zimmer, Preparation of Uranium Kernels by External Gelation Process, Nucl. Tech., 42, p.163, 1979.