Core Material Preparation of TRISO Coated Particle in HTGR Fuel

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

1. Introduction

A VHTR(Very High Temperature Reactor) is being conducted by many countries mainly promoted for electricity production and high temperature process heat. The proposed nuclear fuel for the preliminary reactor concept on these purposes is a TRISO(Tri-Isotropic or multi-layered structure) coated particle prepared by pyro-carbon and silicone coatings on a spherical UO_2 kernel surface as a fissile material.

Generally, UO_2 kernels are prepared by using the modified sol-gel process, wet process, known as the GSP(gel supported precipitation) method. This chemical route was well-known to the potential kernel fabrication process. HTGR nuclear fuel production processes have been classified in five categories of research and development[1,2] :

- Spherical UO_2 kernel preparation step
- Pyro-carbon(PyC) and silicon carbide(SiC) coatings
- Pebble or Prismatic block preparation by using graphite matrix powder
- Fuel performance including a fission products release
- Advanced and improved fuel development

The well-known GSP process is one of the modified processes for an external gelation method developed NUKEM of Germany. As shown in Figure 1, a spherical UO_2 kernel particle was prepared by using a modified external gelation process.

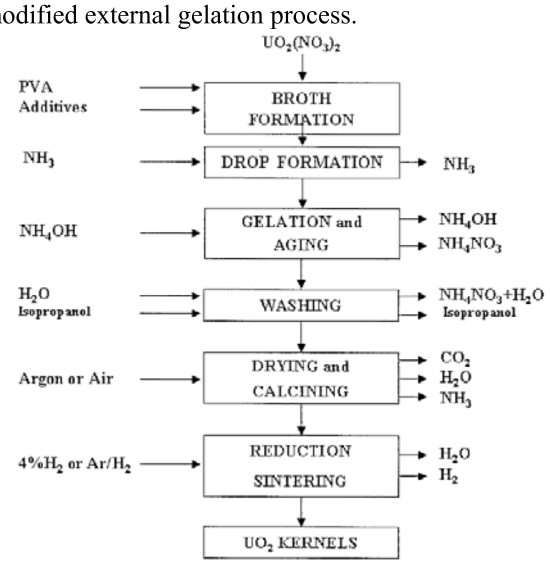

Fig.1. UO_2 kernel fabrication diagram.

UO_2 kernels are highly dense sintered microsphere of stoichometric UO_2 with a nominal diameter of about 500 μ m. The raw material for UO_2 kernel preparation is nuclear grade U_3O_8 powder which will be dissolved with nitric acid to obtain the UN solution. The procedures are formed by mixing the UN solution with organic additives and transferring the aqueous solution to spherical droplets, which are on the surface of liquid droplets slightly hardened by a chemical reaction with ammonia. The necessary viscosity and shape of liquid droplets are achieved by the addition of PVA. THFA solution is added to achieve a controlled shrinkage of the ADU gel particles during gelation, ageing, and washing processes.

After ageing in ammonia water and washing with demi-water and IPA solution, the drying process was carried out by increasing the temperature to ~80°C and applying a slightly vacuum condition. The dried kernels are calcined at a temperature of ~500°C in air atmosphere. The calcined kernels are then subsequently reduced to UO_2 kernels in a hydrogen atmosphere. Due to the pyrophoric nature of porous UO_2, the UO_2 kernels have to be sintered directly thereafter, at a temperature of ~1700°C in a sintering furnace.

2. UO_2 kernel preparation

The experimental apparatus for the spherical liquid droplets preparation mainly consisted of a broth solution storage tank, a flow-meter, a nozzle and vibrating system, a gas supply system, and a gelation column, as shown in Figure 2. After ADU gelation, the AWD apparatus or FBR method was used for ageing, washing, and drying. Also calcination and sintering furnaces were used for calcining of dried-ADU gel particles and reducing of UO_3 particles and densification of UO_2 particles, respectively.

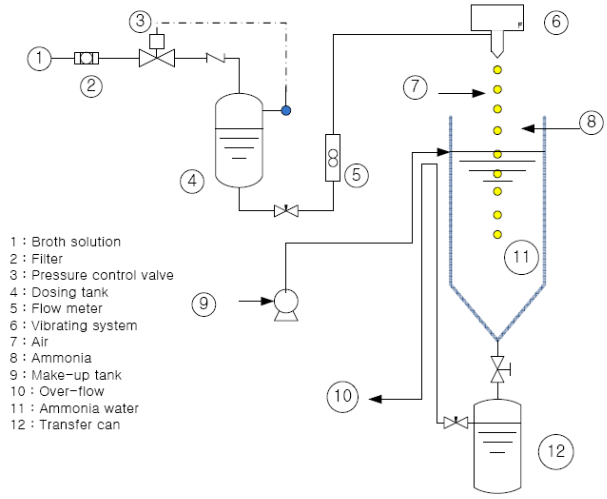

Fig.2. Experimental apparatus.

3. Results

Process parameters for a broth solution making such as [U] concentration of the feeding solution, PVA/THFA mole ratio, the viscosity, and feeding rate of the broth, the treatments conditions of ageing, washing, and drying step, the heating profiles of calcining, reducing, and sintering processes have been studied extensively.

In the initial liquid spherical droplets preparation process, the most important factors influencing the spherical shape are the broth feed rate, viscosity of broth solution, and the vibrating frequency when using a droplet forming nozzle size of a 1 mm diameter.

Figure 3 shows the FT-IR spectra of the intermediate compound and ADU particles prepared by a gel-supported precipitation method in our experiment, in the KBr pellet. Generally, the ADU compound particles exhibited absorption bands associated with a stretching and the bending vibrations of H-O-H in the coordinated water and hydrated water at nearly 3470 and 3225 cm^{-1}, also, stretching bands of the UO_2^{2+} ion, peaks by NH_3 rock, stretching bands of U←O or U←N appeared at nearly 930, 828, 450 cm^{-1}, respectively.

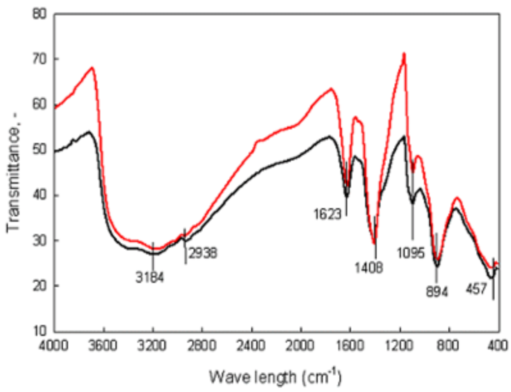

Fig.3. FT-IR spectra of dried-ADU gel particles.

According to these bands of a general ADU, an ADU compound obtained from our experiments is different to that of the one mentioned above. In our case, the obtained 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 nearly 3370 and 3150 cm^{-1}, and the stretching and bending vibrations of N-H in NH_4^+(sharp) and U-O(middle), one sharp peak and another middle peak at nearly 1400 and 1090 cm^{-1}, respectively. Also, a stretching vibration of U-O(strong) and a bending vibration of O-H(middle) containing a PVA precipitate appeared at 450 cm^{-1} and 1627 cm^{-1}, respectively.

Otherwise, the aged, and dried ADU gel particles, UO_3, and UO_2 particles obtained in our laboratory are shown in Figure 4.

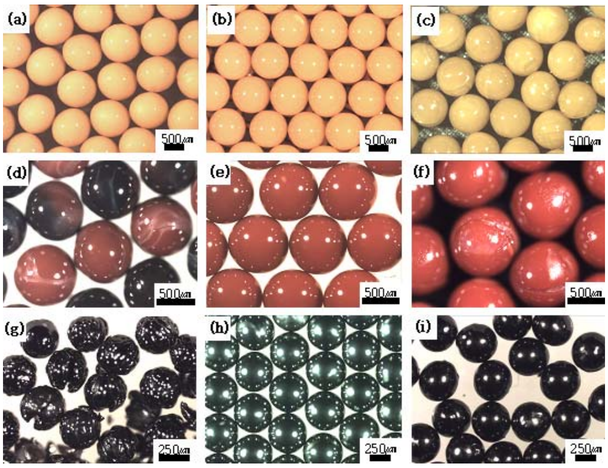

Fig.4. Photograhs of spheres obtained by our experiments : (a)~(c); dried ADU spheres, (d)~(f) ; calcined UO_3 spheres, (g)~(i) ; sintered UO_2 spheres.

The shape of all the UO_2 particles were observed in spherical form and the sphericity was within 1.0+0.15 from the ceramograph observation. The relationship between the shrinkage factor and the uranium concentration in the final broth solution is reported. Also, our heat treatment experiments for a calcination of the dried ADU particles were carried out by raising the temperature until it reached 450 ℃ in an air atmosphere. The heat treatment for the sintering process was carried out until it reached 1700 ℃ in a H_2 atmosphere with the sintering furnace.

4. Conclusion

In this study, to obtain a spherical UO_2 kernel, the most important factors for the droplets preparation are the composition ratio and viscosity of the broth solution, and the frequency/amplitude of the vibrator. Uranium concentration of the broth is 0.5~0.8mol/L, viscosity is 50~80 c.P, vibration frequency is about 100~120Hz. Also the heating rates in the calcination and sintering processes must be kept below 5°C/min. in air condition and 3~5 steps heating/cooling mode until it reached 1700°C in a H_2 gas atmosphere, respectively.

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