Status of UO_2 Sphere Preparation Technology Development for HTGR Fuel in Korea

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1. Introduction

The HTGR(High Temperature Gas Reactor) will play a dominant role in the worldwide fleet of nuclear reactors of the next decade for electricity production and high temperature heat for hydrogen gas production[1]. HTGR have two reactor types, pebble or prismatic, which use the basic fuel concept based on the dispersion of TRISO coated particles in graphite powder[2,3]. The inner part of this TRISO coated particle has a UO_2 sphere of various sizes, which was prepared with a sol-gel method. An external gelation method for UO_2 spheres was designed and developed from a modified GSP(gelation supported precipitation) process as shown in Fig.1[4].

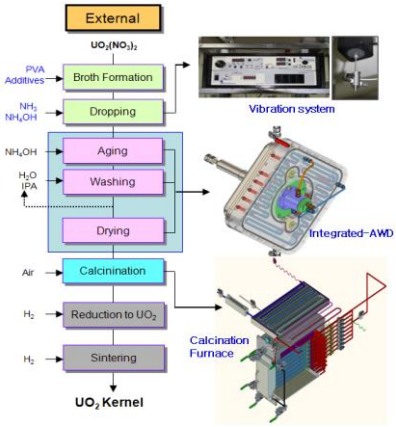

Fig.1. Process flow and equipment for UO_2 sphere production.

2. UO_2 sphere preparation

The process for the spherical UO_2 sphere mainly consists of a broth solution preparation, droplet formation from a vibrating dropping method, an AWD (Ageing-Washing-Drying), and thermal treatment systems. The material and process flow are as below:

- dissolving UO_3 powder and organics : U conc., viscosity of the solution
- broth preparation : pH for pre-neutralization
- sphere formation : vibration frequency and amplitude, pre-hardening of liquid droplets
- ageing, washing, and drying : medium, temperature, and vacuum degree
- calcining : atmosphere and heating profile
- reduction and sintering : atmosphere, heating profile, and max. temperature

In the pre-hardening of liquid droplets among the above processes, the most important factors are the pre-hardening method and contacting equipment of ammonia gas uses at the surface of the liquid droplets. Fig.2 shows the ammonia contacting method and improved-equipment for this treating procedure. The equipment was applied to the double wall concept for effective contacting of ammonia gas at the surface of falling liquid droplets. The contacting method and improved-equipment have already gained a patent in Korea.

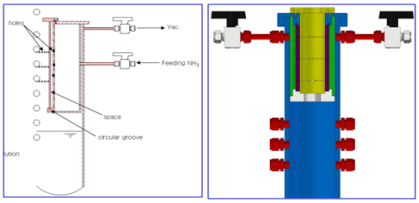

Fig.2. Ammonia gas contacting method and improved-equipment for pre-hardening of the liquid droplet' surface.

UO_2 spheres were prepared using the above process flow and improved-equipment of the lab.-scale experimental apparatus. The procedure for the UO_2 sphere preparation is in due order, that is, broth solution preparation, pre-neutralization of broth, formation of liquid ADU droplets, aged-ADU, dried-ADU, calcining to UO_3, and reduction and sintering to UO_2 finally.

Fig.3 shows that the intermediated-particles and TRISO coated particles were obtained from each unit process. In the initial experiments, the shape of the liquid droplets was made with a non-spherical shape, but from the final trial, the shape of the UO_2 spheres were made with nearly perfect sphere, and its sphericity was measured to below of 1.15 and the sintered density was measured above 10.60g/cm^3. These values are in a good state in comparison with other countries.

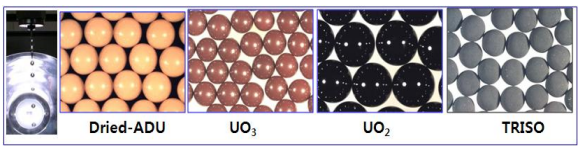

Fig. 3. ADU gels, UO_3 and UO_2 particles, and TRISO coated particles.

Various experiments for improving the sphericity, sintered density, non-crack particles, and internal structure through the cross section analyses of UO_2 spheres were conducted. Here, various experiments included the changes of uranium concentration, viscosity in broth solution, vibration frequency control

during liquid droplets dropping, drying condition, calcining profile, reduction temperature, and sintering conditions.

The optimum conditions for our specifications were determined. The main factors from these preparation experiments are as follow:

- uranium conc. of broth : 0.5~0.6 mol-U/L
- vibration frequency : 80~120 Hz
- ammonia conc. of gelation medium : 7M NH_4OH
- washing method : NH_4OH → H_2O(2) → Azeo-IPA(2) → pure-IPA(2)
- drying : slightly vacuum state and ~60℃
- calcining : 300~450℃ in air atmosphere
- reduction : H_2 atmosphere
- sintering : H_2, 1700℃

On the other hand, the preparing characteristics were evaluated in the UO_2 spheres prepared with the optimum experimental conditions using the above explained procedures. Fig. 4 shows SEM photos of the specimen, which were obtained from the UO_2 spheres made by a resin treating procedure for the observation the inside microstructure of the UO_2 sphere. Also, the analysis profile of the pore volume and surface area of the UO_2 spheres measured with BET and porosity analyses are, in right side of Fig. 4.

Microstructure of the cross section of the UO_2 sphere obtained at heating rate of 0.5℃/min., in the calining process, showed comparatively good states, as shown in the middle photo of Fig.4. In the case of higher heating rate during calcining, the macro pores within the narrow part and micro pores concentrated at the center part existed together in the final UO_2 sphere. But if heating rate is lower than 1℃ in calcining process of ADU gels, the pores appeared uniform in all cross sections.

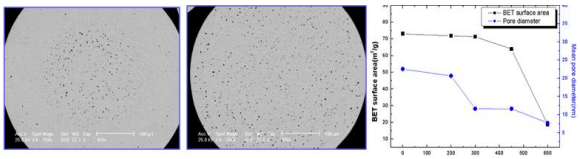

Fig. 4. SEM photos and pore characteristics of UO_2 sphere.

The average pore diameter and surface area of this sphere measured at around 20 nm and 70m^2/g, in case of 200℃ calcination of ADU gels, respectively. These values are a good trend to increase the calcining temperature in the thermal treatment of ADU gels. But, if the heating rate is faster than 5℃/min., the thermal treated-UO_3 particles were cracked or broken, and/or the shrinkage of pores was occurred to reach the center of the sintered -UO_2 sphere.

Otherwise, the SEM photo on the cross section of a final sintered-UO_2 sphere obtained from calcined-UO_3 in an air atmosphere of 450°C. Figure 5 shows the improved-sample specimen obtained from the polishing of a UO_2 sphere. The typical conditions in our experiments were a 0.6 mol-U/L uranium concentration in a broth solution, a 450℃ calcining temperature, and ~1700℃ sintering temperature in a 100%-H_2 atmosphere. The state of the internal microstructure showed the same shapes and same crystallite sizes, as shown in the middle SEM of Fig. 5. Here, the heating rate at the calcining step should be kept to below 3℃/min. for good UO_2 spheres.

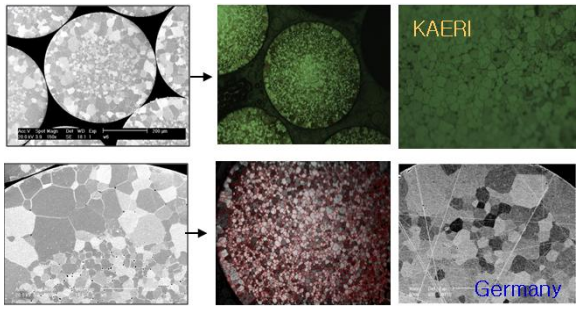

Fig. 5. SEM photos of the internal microstructure at the cross section of an improved-UO_2 sphere.

4. Conclusion

Technical reviews and various lab.-scale experiments on the preparation of ADU gels, and the calcining, reducing, and sintering processes to obtain good UO_2 spheres were carried out in this study.

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