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Spherical UO_2 Size and Microstructure on Broth Preparation Conditions

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1. Introduction

Without a doubt, the High Temperature Reactor will play a dominant role in the worldwide fleet of nuclear reactors of the next decade. It is being conducted by many countries, such as US, Japan, China, and so on, and mainly promoted for electricity production and high temperature heat[1,2]. There are two general designs for He gas cooled reactor; types of the modular high temperature gas cooled reactor and the pebble bed reactor.

Both reactor types use a basic fuel concept based on the dispersion of TRISO coated particles in graphite. The TRISO coated particle for these purposes prepared by pyro-carbon and silicone carbide coatings on a spherical UO_2 kernel surface as a fissile material[3].

Generally, UO_2 kernels are produced 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 processes[4]. Figure 1 depicts a flow diagram of the VTHR fuel preparation process.

After the formation of broth solution contained UN(Uranyl Nitrate) solution, PVA solution, and organic additives, spherical droplets are formed by a vibrating nozzle system. Spherical droplets are converted to liquid-ADU spheres in gelation column, and then finally aged-ADU gel particles in NH_4OH solution. Following the drying and calcining, reduction of the calcined kernels to UO_2 was carried out. This reduced UO_2 particles were converted to a dense structure UO_2 through the sintering process.

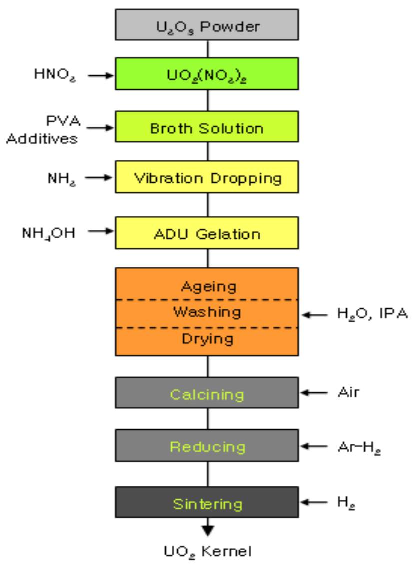

Fig.1. A flow diagram for UO_2 kernel fabrication.

2. UO_2 kernel preparation

The experimental apparatus for the spherical liquid droplets and ADU gel particles preparation mainly consisted of a broth solution storage tank, a flow-meter, vibrating dropping system, a gas supply system for pre-hardening of liquid droplets surface, and a gelation column, as shown in Figure 2. After ADU gelation, the AWD apparatus self manufactured in the lab was used for ageing, washing, and drying of liquid ADU droplets.

Also calcining and sintering furnaces were used for calcination of dried-ADU gel particles, and reduction of UO_3 particles and sintering for densification of UO_2 particles, respectively.

Fig.2. Experimental apparatus (left : gelation column right : sintering system).

3. Results

The sphericity of the final UO_2 kernel prepared by the change of uranium concentration of broth solution was measured by Micriscope observation. Sphericity was calculated to the ratio of maximum diameter and minimum diameter of ADU gel particle. Figure 3 showed the final sphericity variations of spherical ADU gel particle which were prepared according to uranium concentration changes in the broth solution of these experiments.

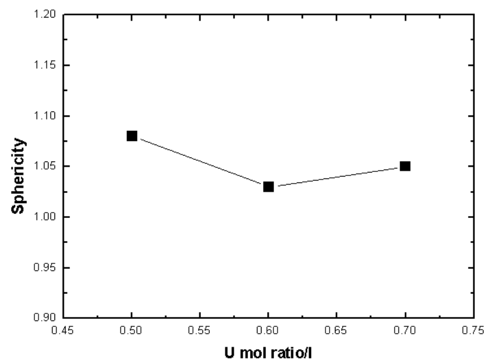

Fig.3. Sphericity variation of dried ADU spheres.

Sphericity had a good state at 0.6 mol-U/L condition in the broth solution. From this result on ADU gel particle preparation, the mole ratio on uranium and the organic additives in the broth solution were known very important factors.

Otherwise, the aged and dried ADU gel particles are converted to UO_3 and UO_2 particles through the calcination process at 450°C in air condition and sintering process of about 1600°C in hydrogen reduction atmosphere, respectively. The shapes of these spherical particles obtained in the laboratory are shown in Figure 4. The sphericity of the experiments was calculated within 1.0+0.15 from the ceramograph observation.

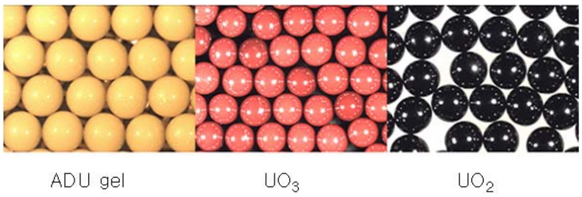

Fig.4. Photograhs of ADU, UO_3, and UO_2 spheres obtained in our experiments.

To check the thermal decomposition characteristics of ADU particles, dired-ADU gel particles were converted to UO_3 form at about 500°C in air condition. In this process, about 7~9% loss in total weight of ADU gel particle occurred due to the decomposition-off of organic additives and phase transformation. And the decomposition rate of ADU gel particle is divided into 3 steps as shown in Figure 5. Remaining water and impurities into dried ADU gel particles were decomposed in first step until about 100°C, and then the NH_4NO_3 as by-product into ADU gel particle formed by the reaction of UN solution and ammonia solution are slowly decomposed-off until 200°C.

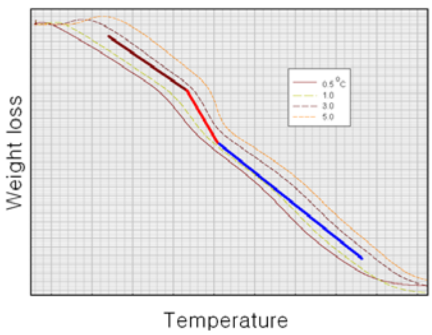

Fig.5. Thermal decomposition profiles of ADU gel particles.

Otherwise, the cross section shape of a final UO_2 sphere obtained from UO_3 calcined in air condition of 450°C showed in Figure 6. Most of the pores in the inner part of kernels showed closed pores structure, and its size were measured to 3~5 ㎛ within the closed structures.

Also crystallite sizes in the outer part of kernels appeared much larger than those of inner part, and its size were measured to about 30 ㎛. The reason suggested was that the ADU gel particles of the bulky state formed because of using the wet sol-gel method.

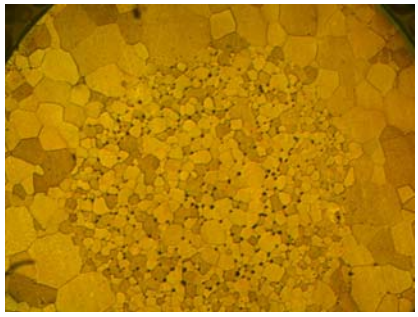

Fig. 6. Ceramograph of an UO_2 kernel cross section.

4. Conclusion

In this study for obtain a spherical UO_2 kernel, the most important factor in spherical ADU gel particles is the mole ratio between uranium and organic additives in broth solution preparation, an optimum uranium concentration in the broth solution is 0.6 mol/L. And the sphericity of ADU gel particle is 1.0+0.15 from the microscope observation. Also, weight loss in ADU decomposition process was measured at about 8% of total ADU weight. Decomposition process were divided into 3 steps, and the inner and outer pore sizes of UO_2 kernel were measured with 3~5 ㎛ and 30 ㎛ in cross section observation, respectively.

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