Calcination, Reduction and Sintering of ADU Spheres for HTGR Fuel

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1. Introduction

The international oil market is again in turmoil in accordance with the increasing of human needs and energy consumption. Soaring oil prices, fears of supply security, and climate change are concerned becoming more concrete make for an uncertain energy future. In this view point, nuclear energy is an important, yet controversial option for energy supply.

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 [1]. HTGR have two reactor types which use the basic fuel concept based on the dispersion of TRISO coated particles in graphite in shown Fig.1.

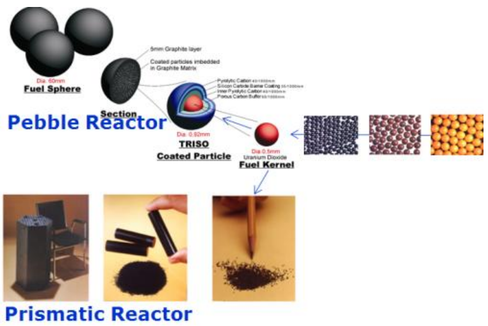

Figure 1. General concepts of VHTR fuel.

The TRISO coated particle for these purposes is prepared with pyro-carbon and silicone carbide coatings on a spherical UO_2 kernel surface as fissile material. The TRISO fuel particle consists of a microsphere (i.e., UO_2 kernel) of nuclear material; encapsulated by multiple layers of pyro-carbon and a SiC layer [2,3].

This multiple coating layers system has been engineered to retain the fission products generated by fission of the nuclear material in the kernel during normal operation and all licensing basis events over the design lifetime of the fuel.

UO_2 kernels are produced by using the modified sol-gel process, a wet process, generally known as the GSP method. Wet chemical processes are flexible in producing kernels of different size and chemical composition with high throughout and yield, good spherical shape, and narrow size distribution. This chemical processing route is well-known to the potential kernel fabrication processes [4].

The principle, as set out in Fig.2, involves first of all preparing a pseudo-sol(also known as a "broth") from an initial uranyl nitrate solution . This broth solution is obtained through addition of a number of additives, as determined by process know-how, including a soluble organic polymer, that are subsequently gels into droplets and are dispersed for ADU precipitation.

The formation of spheres is then formed by the dispersed in air and ammonia gases atmosphere.

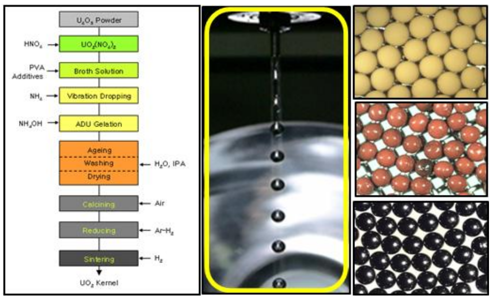

Figure 2. Schematic material flow diagram (left) and particle shapes at each step.

The gelation of polymer was induced at the surface of the droplets in this step. The surfaces of these liquid droplets are slightly solidified by reaction of ammonia gas and uranyl ion into the droplets. Spherical droplets are converted to aged-ADU gels in NH_4OH solution. Then, many steps, such as the ageing and drying of ADU gel, the calcining to UO_3, the sintering to UO_2, were progressed continuously.

2. UO_2 kernel preparation

The experimental apparatus for the spherical UO_2 kernel mainly consisted of a broth solution preparation system, vibrating dropping system, AWD(Ageing-Washing-Drying) system, and thermal treatment system. The shape and size of the ADU gel and UO_3 particles, and inside microstructure of a final UO_2 kernel were observed by means of a SEM.

3. Results

UO_2 kernel prepared with the optimum experimental conditions using the above explained procedures. Here, the sintered-UO_2 spheres were obtained after sintering of calcined-UO_3 spheres. The specimen, shown in Figure 3, was obtained from the UO_2 spheres made by resin treating procedure for the observation of the inside microstructure UO_2 sphere. Figure 3 showed the SEM photos at a horizontal section of a final sintered-UO_2 kernel. In calcining process of dried-ADU gel particles, the heating rates were changed from 0.5℃/min to 5℃/min. with an interval of 1~2℃/min.

Microstructures at the cross section of UO_2 kernel were obtained at low heating rate, from 0.5℃/min. to 3℃/min., in cacination process were comparatively good states, as shown in at (a) and (b) of Figure 3, but in the case of (c), the inside microstructure was not better than (a) or (b). Small space containing the macro pore and micro pore existed in center of the UO_2 kernel.

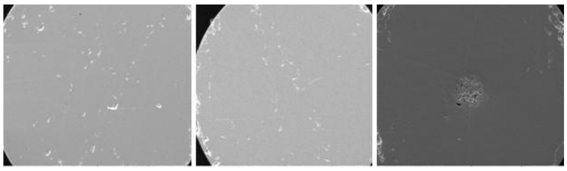

Figure 3. UO_2 inside microstructure according to the change of heating rate in calcining step, (a) 1℃/min, (b) 2℃/min, and (c) 5℃/min.

If the heating rate is faster, it means that is a high heating rate, is critical value in calcining step, the thermal treated-UO_3 particles were cracked or broken, or the shrinkage to reach the center of kernel was not occurred as shown in Figure 4.

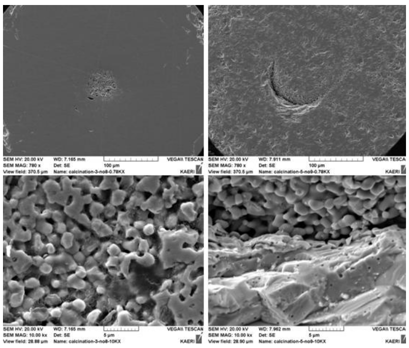

Figure 4. The microstructures of UO_2 kernel center cross section, in the case of a fast heating rate in the calcining step.

Otherwise, the cross section shape of a final sintered UO_2 sphere was obtained from calcined-UO_3 in air condition of 450^{o}C. Figure 5 shows the sample specimen obtained from polishing of a spherical UO_2 particle with height. Inside microstructures of three samples were well state at near top, middle, and center of kernel.

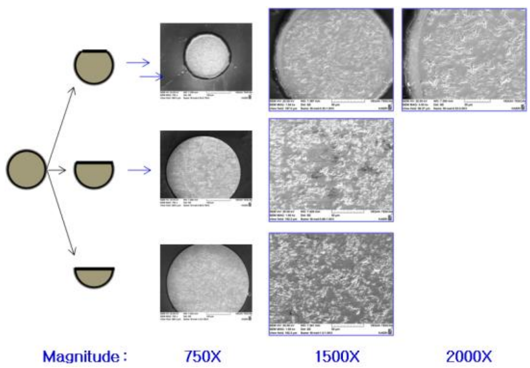

Figure 5. SEM photos on microstructure at cross section of an UO_2 kernel.

The state of microstructure appeared same shapes and crystallite sizes. Therefore, the heating rate at the calcining step should be kept to below 3℃/min. in preparing of spherical UO_2 kernel.

4. Conclusion

In this study, the technical reviews and laboratory scale experiments were carried out about the preparation of ADU gel particles and the thermal treatments on UO_3 cacination and UO_2 sintering processes to obtain a spherical UO_2 kernel. The most important factors are the mole ratio of uranium concentration and organic additives and the heating rate profiles in the calcining and the sintering processes.

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