Preparation and Characteristics of ADU gel Particles

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

HTGR (High Temperature Gas Reactor) energy technology development has been spot-lighted for a clean hydrogen gas and electricity production for the countermeasures of supplying a massive energy production in the next decades because other energies such as solar heat, wind power, and tidal energy, can only produce a small amount of electricity or they are not as effective[1].

Generally, the production of spherical UO_2 kernels for a HTGR nuclear fuel can be carried out by wet chemical processes, a sol-gel process, based on a solidification of uranium liquid droplets as a raw material. Sol-gel process is a proper way to produce the UO_2 kernel because of its advantages in a high quality of products, low heat treatment temperature, and an easy control of the components.

However, there are only a few reports on the preparation of a spherical UO_2 kernel by a sol-gel method. In this study, spherical ADU gel particle and UO_3 particles *via* an UN(uranyl nitrate, $UO_2(NO_3)_2$) solution as a raw material were prepared. And the characteristics of these liquid droplets and the ADU gel particle prepared with the sol-gel method were analyzed, and the heat treatment characteristics from the ADU gel to UO_3 particle were analyzed by a Streoscope, TG/DTA, and X-ray.

2. Theory and Method

The UO_2 kernel preparation starts with dissolving uranium oxide powder in nitric acid[2].

$$3U_3O_8+20HNO_3 \rightarrow 9UO_2(NO_3)_2+10H_2O+2\ NO$$

As shown in the above chemical reaction equation, the obtained UN solution is rather acidic, therefore this UN solution was pre-neutralized with an ammonia solution, about a 7 mole solution, before a ADU gel precipitation. During the next step, the broth solution has to be prepared by mixing the pre-neutralized UN solution, THFA, PVA and demi-water. A preparation vessel is used for mixing all the ingredients until sufficiently homogenized. The kernel is prepared using a single nozzle system that is located at the top of the gelation column. The gelation column is filled up with ammonia solution. Broth solution prepared from above is fed through the nozzle system. And the vibrating frequency of the nozzle system cuts the broth solution flow at the nozzle outlet and forms a broth solution droplet.

The droplets fall through the NH_3 gas layer which hardens the surface of the droplet by a reaction of the UN solution and NH_3 gas. These falling-droplets react with the ammonia solution in a gelation column, and then they are changed the ADU gel particles.

Final, the liquid-ADU gel particle obtained from the above reaction is transferred to the ageing, washing, and drying steps. First, after an ageing with an ammonia solution with a concentration of 7 mole-NH_4OH/ℓ, aged ADU gel particles are washed first with a diluted ammonia solution, and then with pure water, azeotropic IPA, and pure IPA. The experimental apparatus for the spherical liquid droplets preparation mainly consist of a PVA storage tank, a nozzle and vibrating system, a gas supply system, and a gelation column as shown in Figure 1.

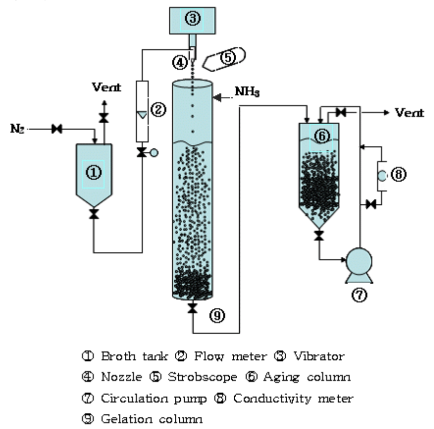

Fig.1. Experimental apparatus for an ADU gel particle production.

3. Results

3.1 Spheical Droplets Preparation

Figure 2 shows the results of the droplets photographs obtained from our droplet preparation experiments. The droplet size was calculated with the flow rate and the used nozzle diameter, and the natural laminar jet length which was observed to be about 5~10mm.

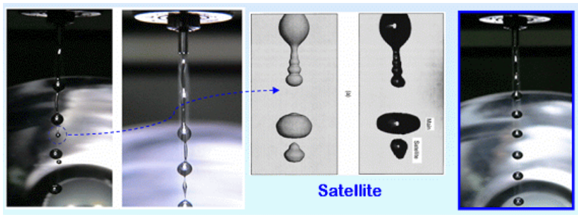

Fig.2. Droplets preparation by using a vibrating nozzle system.

If the relation between the feeding rate of the broth solution and the frequency/amplitude of the vibrating system is not discordant, small satellite drops are formed as shown in the center photograph of Figure 2.

As a result, the harmony among the flow rate of the feed solution, the frequency and the amplitude of the vibrating system are important factors to obtain the same size spherical droplets.

3.2 ADU gel Preparation and Characteristics

From technical reviews on a HTGR fuel preparation in advanced nations, in our case the basic flow diagram for a UO_2 kernel production was accomplished. Figure 3 shows the ADU gel and UO_3 particles obtained from our UO_2 kernel production experiments. Broth solution is made from a mixture of UN solution (an about 1.8 mole-U), ammonia solution, concentrated THFA, and the PVA solution. And the frequency and amplitude of the used vibrating nozzle system were ~100 Hz and ~150V respectively, and the nozzle size was 1ϕ mm in inner diameter.

Fig.3. ADU gel and UO_3 particles obtained in our experiment.

The ADU gel particle size was obtained at about 1900~2000 μm, at nearly the same size and spherical type, but not an exact sphere. The reason is that we could not harmonize the feed rate of the broth solution and the frequency and the amplitude of the vibrating nozzle system.The XRD pattern of the ADU gel particle obtained from our experiment and a similar-ADU compound particle from a JCPDS XRD source were compared as shown in Figure 4[3].

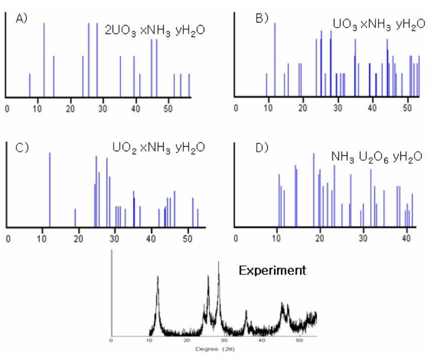

Fig.4. X-ray diffraction profiles for similar ADU.

ADU particle from our experiment and a similar-ADU particle from a JCPDS XRD source do not correspond perfectly, but the C or D peak in Figure 4 appeared to be alike at a 2θ angle. Therefore we assumed that the ADU gel particle obtained from our experiment has a similar $UO_3 \cdot xNH_3 \cdot yH_2O$ or $2UO_3 \cdot xNH_3 \cdot yH_2O$ phase. On the other hand, the thermal behavior of the dried ADU gel particle was studied by TG-DTA analysis. Figure 5 is the TG-DTA curves of the dried-ADU gel particle which was heated at a rate of 1~5 ^oC/min. in an air atmosphere and cooled down to room temperature when the desired temperature was reached. From the DTA profile in Figure 5, the exothermic peaks showed at around 190 ^oC and 390 ^oC. These peaks are due to the thermal decomposition of the THFA and PVA contained in the ADU dried-gel particle.

The THFA and PVA decomposition temperature is a very important factor for the thermal treatment of a dried-ADU gel particle. If the dried-ADU gel particle receives a thermal shock by a rapid heating, the dried-ADU gel particle would crack. Therefore the thermal treatment of dried-ADU gel particle must be progressed very carefully in the region of 150~450 ^oC.

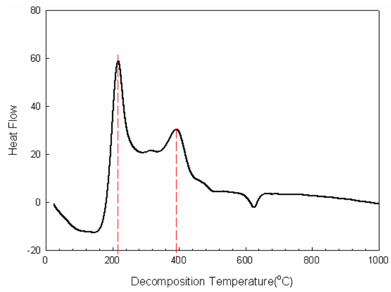

Fig.5. Thermal treatment curve of dried ADU gel particle.

4. Conclusion

In this study, for a spherical droplets formation by using a vibrating nozzle system, the most important factor is a harmony of the feeding rate of the broth solution and the frequency and the amplitude of the vibrating nozzle system. From various analyses, the ADU gel particle was judged to be a $UO_3 \cdot xNH_3 \cdot yH_2O_{(s)}$ form, and the dried ADU gel needs to avoid a rapid heating rate in the range of 150~450 ^oC during a thermal treatment.

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