C-ADU Gel Particle Preparation by Modified-External Gelation Method

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1. Introduction

VHTR (Very High Temperature Gas Reactor) fuel technology is being developed in the US for a Next Generation Nuclear Plant(NGNP). The concept of fuel of a VHTR is based on a sphere kernel of fissile or fertile material, with multiple coating layers to create a gas-tight particle [1]. The fuel particle of a VHTR in the US is based on microspheres containing a mixture of UO_2 and UC_2 coated particles with multi carbon layers and a SiC layer. This mixture is called a "UCO" kernel. This was first prepared through an internal gelation process at ORNL in the late 1970s. The size of the fuel kernel ranges between about 200 and 500 ㎛.

The fabrication process of this kernel was based on the sol-gel reaction between ADUN(acid deficient uranyl nitrate) and additives such as HMTA and urea[2]. The UCO kernel preparation procedures from this process are as follow:

- ADUN solution preparation
- broth solution preparation containing carbon source
- spherical liquid droplets formation by vibration
- gelation by chemical reaction of uranyl ion in ADUN solution and ammonium ion in additive
- ageing , washing, and drying
- thermal treatment 1, 2, 3
- carbonization
- sintering

The C-ADU(carbon contained ADU) gel precipitation is based on the following chemical reaction :

$$[2UO_2(NO_3)_2 + HMTA + Urea + C] + 3NH_4OH \rightarrow C\text{-}(NH_4)_2U_2O_7 + 2NH_4NO_3 + H_2O$$

Carbon black powder as a carbon source in the final UCO kernel is added during the broth solution preparation, in the processing of UCO kernel fabrication [3]. The preparation of a good quality UCO kernel is very difficult owing to the homogeneous distribution of carbon in a UCO kernel.

The key technology used to obtain a good quality sphere (sphericity, density, C/U, O/U ratios) is a uniform distribution of carbon particles into the C-ADU gel sphere, i.e., during the gelation step of liquid droplets formation before the thermal treatment.

We first carried out carbon source selection experiments on the various kinds of carbon black powder and a dispersion test in a simulated broth solution. Second, the C-ADU gel particles were prepared using a depleted UN solution through a **modified-external gelation method**. The C-ADU gel particles formation is based on the following chemical reaction by a modified- external gelation process :

$$[2UO_2(NO_3)_2 + PVA + THFA + C] + 3NH_4OH \rightarrow C\text{-}(NH_4)_2U_2O_7 + 2NH_4NO_3 + H_2O$$

The initial step for UCO fabrication is converting UO_3(or U_3O_8) power into an UNH solution, with nitric acid. The UNH solution is mixed with carbon particles (with carbon black) using a well dispersing method, and a THFA (Tetrahydrofurfuryl alcohol) solution was added. This mixture again is added to a PVA (Polyvinyl alcohol) dispersion in water to form a broth solution.

The broth solution produced was transferred to a gelation column attached with a vibrating nozzle system, and spherical ADU liquid droplets were produced with a proper frequency, amplitude, and flow-rate.

The overall material flow on the UCO sphere fabrication from our **modified-external gelation process** is simply shown in Fig. 1.

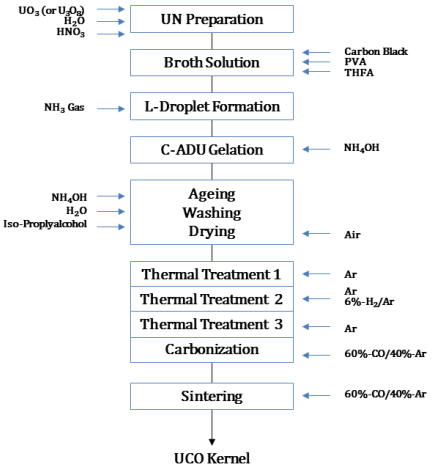

Fig.1. Overall material flow for UCO kernel fabrication.

2. Experiments

In these experiments, we first prepared the simulated solution using $Ce(NO_3)_3$ in place of the UN solution. The metal ion concentration of this solution is 1.97 moles. Various kinds of carbon black powder were purchased for carbon source selection, and the characteristics of the carbon black powder were analyzed. Nano-sized carbon black powder is more difficult to disperse. We obtained high purity carbon black powder available commercially from Columbian Chemical Company (Raven series) and Cabot Corporation (Cabot series).

After the above experiments, uranyl ions inside the spherical ADU liquid droplets and ammonia ions in the gelation column were gelled through a chemical reaction (above chemical reaction on external gelation

method), which produces C-ADU gel particles. These particles were completely aged through a maturing process which is unreacted uranyl ions inside the C-ADU gel particles perfectly reacted with ammonia ions in the gelation column.

Finally, the dried C-ADU gel particles were obtained through a washing processes with distilled-water and an IPA solution, and then underwent a drying process. We prepared a broth solution with depleted UN solution and other additives following table 1.

Table 1. Broth solution and liquid C-ADU droplets forming conditions by depleted UN solution.

Broth	$UO_2(NO_3)_2$	1.97 M
	$NH_4(OH)$	-
	Carbon Black	Cabot
	THFA(원액)	99.9%
	PVA(20%)	Mowiol
Vibration	Frequency	90 -100Hz
	Amplitude	4-5G
Mixing	CB dispersion	Ultrasonic
	Add THFA	Mechanical
	Add PVA	Mechanical

3. Results and discussions

Fig. 2 shows the results from the obtained cumulative velocity distribution of about 10 CBs particle dispersions using a LUMIsizer. The CB10 sample shows that the relative cumulative velocity and the velocity distribution density have the highest value. This is a Cabot Emperor 1800 CB particle.

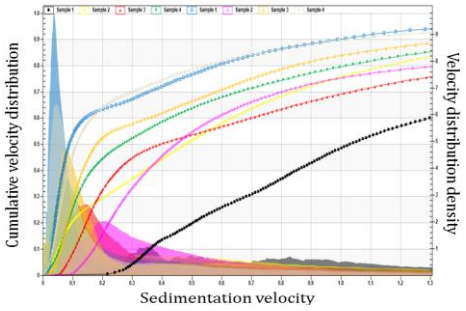

Fig. 2. The experimental results from the CBs' selection experiment.

Otherwise, the broth solution containing carbon particles selected above was prepared with a mixing of THFA and PVA solutions in a depleted-UN solution with adjusted viscosity by adding distilled-water. Fig. 3 shows a SEM photograph of ADU and C-ADU gel particles obtained from our depleted UN solution.

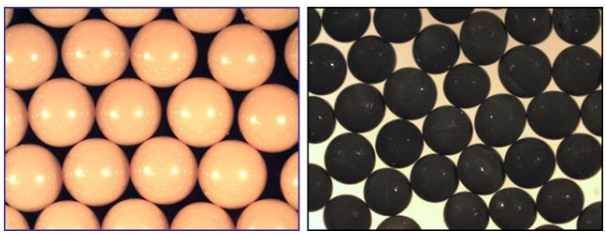

Fig.3. ADU gel particles (left : without carbon, right : with carbon)

Fig. 4 shows that the C-ADU gel particles based on a FT-IR curve have strong absorption peaks at about 2450 cm^{-1}. When comparing compounds without carbon, they show rather complicated peaks at 800-1800cm^{-1} because the C-ADU gel particles are composed of more complicated compounds.

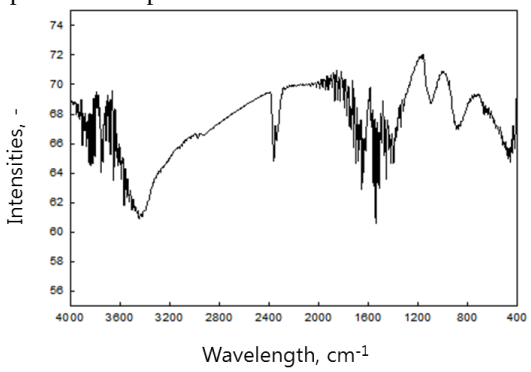

Fig.4. FT-IR curve of C-ADU gel particle.

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