Carbon Black Selection from Simulated Broth Solution for ADU Gel Spheres

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1. Introduction

The VHTR (Very High Temperature Gas Reactor) is one of the reactor concepts in the Gen IV International Collaboration. The nuclear fuel of a VHTR in the US is based on microspheres containing a mixture of UO_2 and UC_2 coated with multi carbon layers and a SiC layer. This mixture is called a "UCO (uranium oxicarbide)" kernel. The fabrication process of this kernel was based on the sol-gel method between an ADUN and HMTA and urea, a process referred to as internal gelation[1].

UCO kernel microspheres were first prepared at ORNL in the late 1970s. CB(Carbon Black) as a carbon source in the final UCO kernel is added during the broth solution preparation, in the processing of UCO kernel fabrication[2]. The preparation of a good quality UCO kernel is very difficult due to the homogeneous distribution of carbon in a UCO kernel.

The key requirement to obtain a good quality kernel is a uniform distribution of carbon in the ADU gel sphere forming process before the thermal treatment, i.e., during the gel formation step. The internal gelation concept was adapted in ADU gel sphere fabrication in the ORNL process of the US. Generally, UO_2 kernel microspheres are prepared by an internal gelation method (USA, India) or external gelation method (Germany, China, Japan). The UCO kernel microspheres prepared only in the US, use an internal gelation method.

A material flow chart on the preparation of the microsphere kernel is simply shown in Fig. 1.

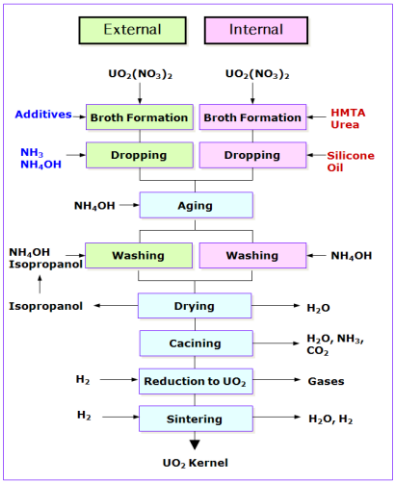

Fig.1. Material flow for microsphere fabrication.

The broth solution preparation, the raw material, additives, and thermal steps such as calcining and sintering processes were different to compared with the external gelation and internal gelation methods[3].

In this study, we first carried out the matching CB selection experiments among the various kinds of CBs in a broth solution, for UCO kernel preparation using an external gelation method.

2. Experiments

2.1 Simulated solution preparation

The UN(Uranyl nitrate) solution as a fertile material is generally used in the broth solution preparation in UCO kernel fabrication. In these experiments, we first prepared the simulated solution with $Ce(NO_3)_3$ in place of the UN solution. The metal ion concentration of this solution is 1.97 moles.

2.2 CB characteristics

Various kinds of CBs were purchased for the matching of CB selection, and the characteristics of the CBs were analyzed. CBs with nano-sized particles are more difficult to disperse. The CBs are selected for this study have a low surface area, and high purity CBs available commercially from Columbian Chemical Company (Raven series) and Cabot Corporation (Cabot series). Table 1 shows the properties of the CBs.

Table 1. CB properties.

	BET (total)	External surface area	Surface area in micro-pore	Cumulative pore volume	Micro-pore volume	Elements(%)		Volatile content
						O	N	
Black Pearls L	141	114	27	0.10	0.02	N/A	N/A	4.8
Raven 1000	106	106	0	0.13	0	3.1	0.1	4.1
Raven 1040	97	95	0	0.07	0	N/A	N/A	3.8
Raven M	81	79	0	0.06	0.01	2.4	0.2	2.3
Cabot A (pristine)	104	104	0	0.08	0	N/A	N/A	N/A
B (oxidized)	123	123	0	0.09	0	N/A	N/A	N/A
E (amine)	N/A	N/A	N/A	N/A	N/A	5.8	0.4	N/A
F (carboxyl)	80	80	0	0.06	0	5.8	0.9	N/A
G (sulfonate)	85	84	1	0.06	0	5.5	0.2	5.9

2.3 CB Dispersion

First, the mixed solution was prepared from the mixing of the simulated broth solution made by $Ce(NO_3)_3$, a trace of ammonia solution, organic alcohol, and organic polymer. The simulated solution was agitated with a mechanical stirrer or sonification force, and then various CBs were added to the above solution and mixed violently.

Here, the mixing method before and after feeding of the organic polymer used only sonification force, and then CBs were added. The dispersion method of CBs was divided into ultrasonic force dispersion and high shear mechanical dispersion (Fig.2).

After the final mixing of CBs, the samples for the dispersion effect were extracted from the broth solution.

The dispersion degree was analyzed by a dispersion stability analyzer.

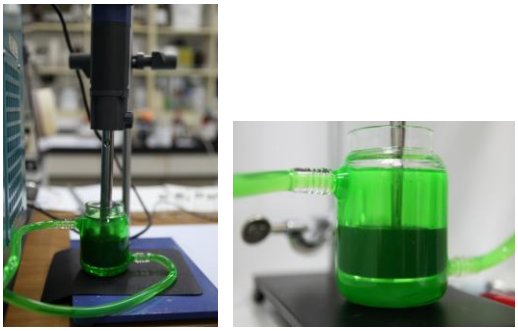

Fig. 2. Experimental apparatus for CBs dispersion.

3. Results and discussions

Matching CBs in our broth system were selected through the various dispersion experiments. Samples of 10 CBs were obtained after the dispersions of ultrasonic and mechanical mixings, and these samples were analyzed by a dispersion stability analyzer.

Fig.3 shows the results of CB dispersions. The CB10 sample shows that the relative cumulative velocity is the highest value. This CB is Cabot Emperor 1800 CB in table 1. Otherwise, if the ultrasonic force is kept at the beaker during the mixing of the solution and CB dispersion, the inside temperature of the beaker increased by the continuous sonification force. Thus, the beaker was cooled by a chilling system as shown in Fig 2.

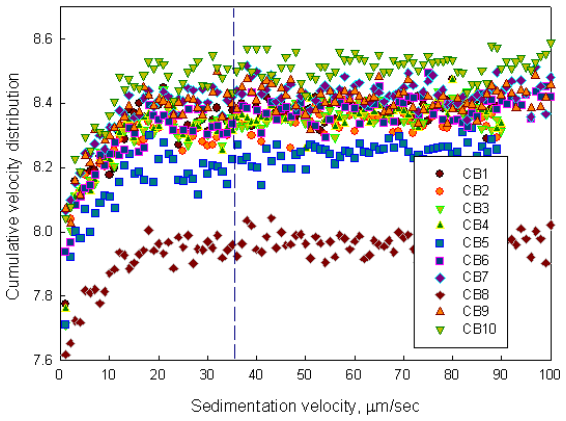

Fig. 3. The results of CB selection experiments.

Fig. 4 shows SEM photographs of samples CB8 and CB5, respectively. The particle size of the CB8 sample is much smaller than that of CB5. The size is nearly 50~80nm, and a few amounts of CB were aggregated. These aggregates are crushed by sonification force but not crushed by high shear mechanical mixing.

Otherwise, we considered the CB dispersion order in the simulated solution. The CB feeding chance is double at the broth solution preparation, which is before and after organic polymer feeding in the simulated solution. The CB feeding after mixing with a simulated solution and organic alcohol showed a better result than the other method. The heat occurs by many times the treating ultransonic force used in this system. If these forces are added continuously after an organic polymer feeding, the physical property of the final broth solution can be changed. In our case, the ultrasonic dispersion method considered after CB feeding in the simulated solution that is mixed solution with simulated solution and organic alcohol, and then the mechanical shear mixing method selected in case organic polymer mixing.

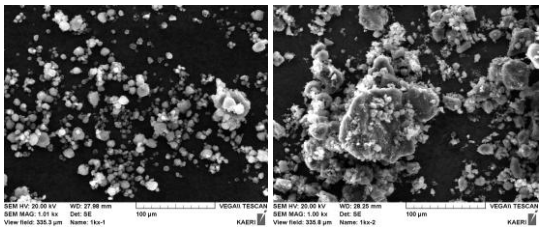

Fig.4. SEM photographs on the CB8 and CB5 samples.

4. Summary

Matching CBs in our broth system were selected through the various dispersion experiments for apply to the broth preparation of an external gelation. The CB10 sample showed that the relative cumulative velocity has the highest value in our tests.

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