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Carbon Black Dispersion Experiments into Broth Solution for ADU Gel Preparation

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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 concept of fuel of a VHTR is based on a sphere kernel of fissile or fertile material, with multiple coatings 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. The size of the fuel kernel ranges between 100 and 500 ㎛. UCO kernel microspheres were first prepared through an internal gelation method at ORNL in the late 1970s.

The fabrication process of this kernel was based on the internal gelation between an ADUN and HMTA and urea[2]. The CB-ADU gel precipitation is based on the following chemical reaction,

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

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[3]. The preparation of a good quality UCO kernel is very difficult due to the homogeneous distribution of carbon in a UCO kernel.

The key technology to obtain a good quality sphere is a uniform distribution of carbon in the ADU gel sphere forming process before the thermal treatment, i.e., during the gel formation step. A material flow chart on the preparation of the microsphere kernel from the internal gelation process is simply shown in Fig. 1.

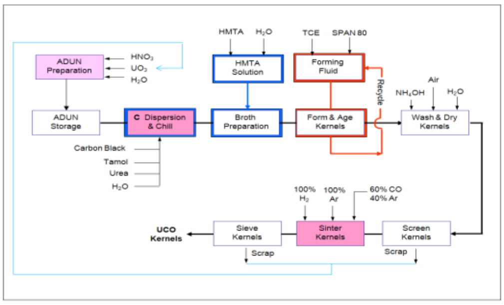

Fig.1. Material flow diagram for kernel fabrication.

The broth solution preparation, raw material, additives, and thermal steps such as calcining and sintering processes were different compared with the external gelation and internal gelation methods[4].

However, in this study, we first carried out the matching CB selection experiments among the various kinds of CBs and its dispersion characterisitic analysis in a broth solution, for UCO kernel preparation using an external gelation method which has a lot of experience in our UO_2 sphere preparation.

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 the 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.

Table 1. The compositions of simulate a broth solution.

$Ce(NO_3)_3$	A (ml)	1.97 mol/L
NH_4OH	B (ml)	minimum
THFA	C (ml)	original
CB	D (g)	10 samples
PVA	E (ml)	Mowiol
H_2O	trace	property control

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 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). SEM photographs of various CBs raw materials are shown in Fig. 2.

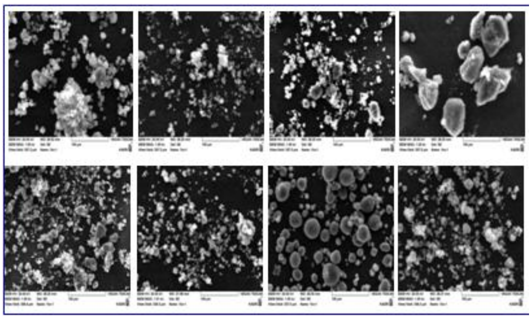

Fig. 2. The SEM photographs on various kind of CBs raw materials.

2.3 CB Dispersion experiments

First, the mixed solution was prepared from the mixing of the simulated broth solution made by 1.97 mole-$Ce(NO_3)_3$, THFA, and PVA. Here, the mixing method before adding the PVA used only a weakened sonification force, and CBs were added. The dispersion method of CBs used a little strong ultrasonic force dispersion, and finally high shear mechanical mixing for viscous PVA dispersion.

The dispersion degree was analyzed by a dispersion stability analyzer and Cryo-SEM.

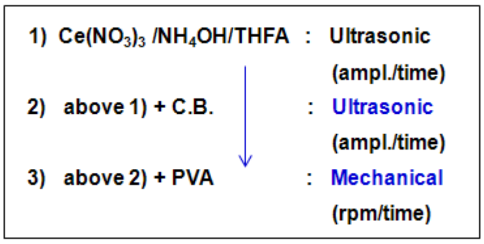

Fig. 3. Experimental procedure for CBs dispersion.

3. Results and discussions

Fig. 4 shows the results from the obtained cumulative velocity distribution of about 10 CBs dispersions using a LUMIsizer. CB10 shows that the relative cumulative velocity is the highest value. This is a Cabot Emperor 1800 CB. The basic properties of the Cabot 1800CB are as follows:

total BET : 85 cm^2/g, surface area : 84 cm^2/g
pore volume : 0.06 m^3/g, volatile : 5.9 %
functional group : sulfonate

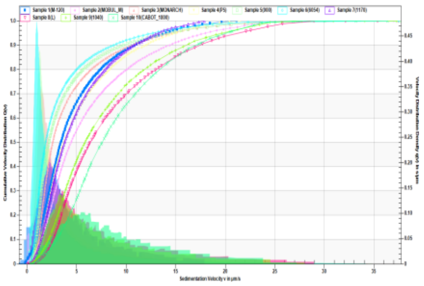

Fig. 4. The results of CB selection experiments.

Fig. 5 shows SEM photographs of raw particles of CB1-CB10, respectively. The particle size of the CB8 sample is much smaller than that of the others. 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.

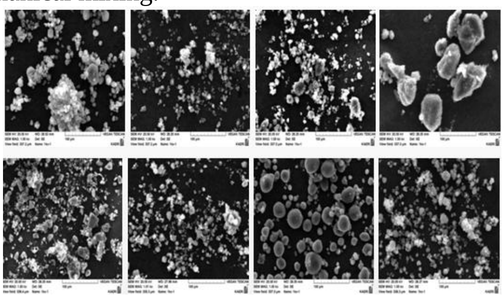

Fig. 5. SEM photographs of raw CBs.

Fig. 6 shows the results on final dispersion obtained from the above mixing procedure for raw material CB. Most aggregated-CB particles are well crushed and dispersed in a broth solution from a Cryo-SEM photograph. The following result was obtained from the CB 10 sample case.

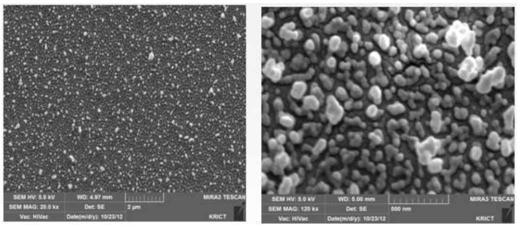

Fig.4. SEM photographs on the CB10 sample.

4. Summary

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

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