Milling Behavior of Matrix Graphite Powders with Different Binder Materials in HTGR Fuel Element Fabrication: I. Variation in Particle Size Distribution

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1. Introduction

The fuel element for HTGR is manufactured by mixing coated fuel particles with matrix graphite powder and forming into either pebble type or cylindrical type compacts depending on their use in different HTGR cores. The coated fuel particle, the so-called TRISO particle, consists of 500-㎛ spherical UO_2 particles coated with the low density buffer Pyrolytic Carbon (PyC) layer, the inner and outer high density PyC layer and SiC layer sandwiched between the two inner and outer PyC layers. The coated TRISO particles are mixed with a matrix graphite powder properly prepared and pressed into a spherical shape or a cylindrical compact finally heat-treated at about 1900°C. These fuel elements can have different sizes and forms of compact.

The basic steps for manufacturing a fuel element include preparation of graphite matrix powder, over-coating the fuel particles, mixing the fuel particles with a matrix powder, carbonizing green compact, and the final high-temperature heat treatment of the carbonized fuel compact. In order to develop a fuel compact fabrication technology, it is important to develop a technology to prepare the matrix graphite powder (MGP) with proper characteristics, which has a strong influence on further steps and the material properties of fuel element. In this work, the milling behavior of matrix graphite powder mixture with different binder materials and their contents was investigated by analyzing the change in particle size distribution with different milling time.

2. Experimental

In the experiment, natural graphite powder, electro (artificial) graphite powder and binders were mixed with different ratios to produce the matrix powder samples. The experimental procedure consists largely of 4 steps as follows. 1) resin dissolution, 2) MGP mixing, 3) MGP kneading and drying 4) MGP milling In order to find the MGP with intended particle sizes suitable for compact fabrication, a total of 6 kinds of graphite powder samples with two kinds of resins and their 3 different contents were prepared.

2.1 Materials used and methods of analysis in the experiment

A natural graphite powder and an electro-graphite powder, supplied by Graphit Kropfmühl AG, Germany and SGL, Germany, respectively, were used. Two binder materials, a phenolic resin and a PVB (poly vinyl butyral), were used to observe the difference in behaviors of binder during matrix graphite cake preparation and milling. For the analysis of MGP milling behavior, particle size distribution was measured by sieve test.

2.2 Preparation of binder solutions with different contents

In order to examine any change in the quality of MGP depending on binders, PVB (Poly Vinyl Butyral) and phenolic resin with 3 different contents which give viscosities of binder solution of 3.63, 6.47 and 10.60 centipoise(Cp) were selected and prepared as shown in Table 1. Different amounts of two different binder materials were dissolved to have the same viscosity in binder solution in each content category. These 3 viscosities correspond to 30, 35.2 and 39.0 wt.%, respectively, for phenol resin binder and 2.54, 3.74 and 4.95 wt.%, respectively, for PVB binder.

Table 1. Preparation of samples with different contents of binder

binder type	material	content 1	content 2	content 3
Phenol resin	N. graphite wt., g	128	128	128
	E. graphite wt., g	32	32	32
	binder wt., g	31.5	40	47.1
	solvent (wt., g)	MeOH (73.6)	MeOH (73.6)	MeOH (73.6)
PVB	N. graphite wt., g	128	128	128
	E. graphite wt., g	32	32	32
	binder wt., g	2.9	4.24	5.62
	solvent (wt,.g)	EtOH (113.5)	EtOH (109.3)	EtOH (107.89)
binder solution viscosity, Cp		3.63	6,45	10.6

2.3 Experimental conditions

Mixing of natural graphite powder and electro-graphite powder was carried out by using a V-mixer for 1 hr at 100 rpm. Kneading of the powder mixture and each binder solution prepared following the conditions as summarized in Table 1 was conducted for 30 min. using a laboratory kneader, followed by the forced sieving of

the kneaded bulk to make matrix graphite cake and drying for a total of 15 hrs at 100°C. Milling of the prepared matrix graphite cake for each sample was carried out using an attrition mill at 280 rpm for 2, 4, 6 and 8 hrs.

3. Results

The variations in particle size distribution of MGP samples prepared with phenol resin (PR) of different contents (Table 1) and milled for 2 hrs and 8 hrs are shown in Fig. 1 and 2, respectively..

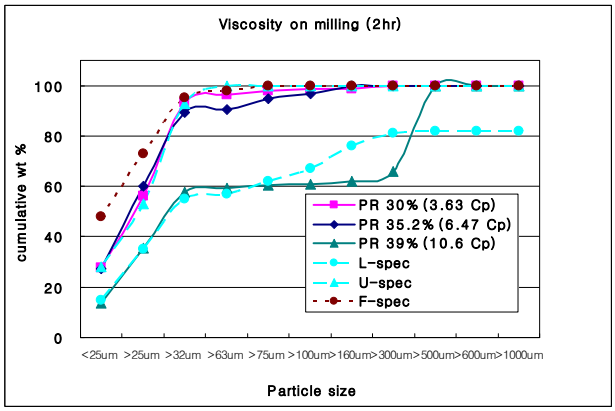

Fig. 1. Variation in particle size distribution of MGP samples with different contents of phenol resin binder milled for 2 hrs.

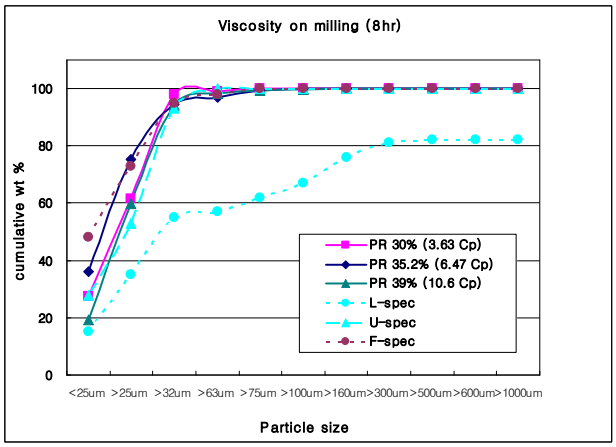

Fig. 2. Variation in particle size distribution of MGP samples with different contents of phenol resin binder milled for 8 hrs.

As shown in Fig. 1, the sample with the largest amount of binder (39 wt.%) shows large fraction of coarse particle size compared with smaller amounts of binder. This reveals that the amount of binder influences the hardness of MG cake, and the milling for 2 hrs was not sufficient for the sample with 39 wt.% binder to be milled appropriately for further step, when compared with lower and upper limits of the specified range. When this sample was milled for 8 hrs, its particle size distribution became similar to those with smaller amounts of binder, as shown in Fig. 2.

The variation in particle size distribution of MGP samples prepared with PVB of different contents (Table 1) and milled for 2 hrs and 4 hrs are shown in Fig. 3 and 4, respectively. As shown in Fig. 1, contrary to the behavior of the largest amount of PR binder, the largest amount of the PVB binder shows similar particle size distribution to those with other smaller amounts of binder. Increasing milling time did not change the variation of particle size distribution, as shown in Fig. 4.

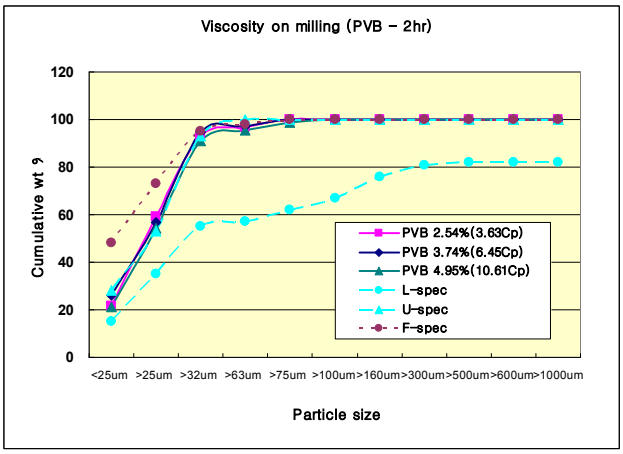

Fig. 3. Variation in particle size distribution of MGP samples with different contents of PVB binder milled for 2 hrs.

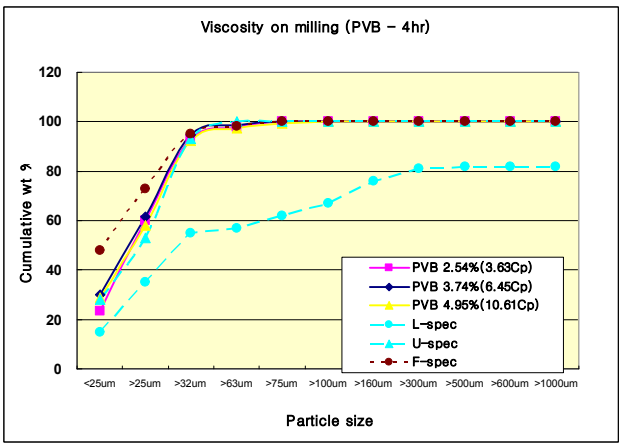

Fig. 4. Variation in particle size distribution of MGP samples with different contents of PVB binder milled for 4 hrs.

4. Conclusions

1) The amount of phenol resin binder influences the hardness of the MGP, i.e., increasing binder content make MGP powder difficult to mill to finer particles, whereas the PVB binder does not.
2) Two lower viscosities (3.63 and 6.45 Cp) of binder solution show similar milling behavior, i.e., similar particle size distribution of the two different contents of binder can be obtained in both PR-containing and PVB-containing matrix powder graphite samples.