

An Experiment on the Carbonization of Fuel Compact Matrix Graphite for HTGR

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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- μm 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 properly prepared matrix graphite powder, pressed into a spherical shape or a cylindrical compact, and finally heat-treated at about 1800°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. The carbonization is a process step where the binder that is incorporated during the matrix graphite powder preparation step is evaporated and the residue of the binder is carbonized during the heat treatment at about 1073 K. In order to develop a fuel compact fabrication technology, and for fuel matrix graphite to meet the required material properties, it is of extreme importance to investigate the relationship among the process parameters of the matrix graphite powder preparation, fabrication parameters of fuel element green compact and the carbonization condition, which has a strong influence on further steps and the material properties of fuel element. In this work, the carbonization behavior of green compact samples prepared from the matrix graphite powder mixtures with different binder materials was investigated in order to elucidate the behavior of binders during the carbonization heat treatment by analyzing the change in weight, density and its change rate of the compact samples.

2. Experimental

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 (PR) and a PVB (poly vinyl butyral), were used to observe the difference in behaviors of binder during carbonization. For the analysis of weight change of compact samples thermogravimetric apparatus was used, where the temperature of carbonization was held at 1073K and flowing N_2 and Ar gas atmospheres were separately employed. Densities of compact samples were measured geometrically.

2.2 Preparation of binder solutions and matrix graphite powder samples

Two different binder solutions were prepared, having the same viscosity of binder solutions (6.45Cp) by dissolving 40g of phenol resin in 73.6g of methanol and 4.24g of PVB in 109.3g of ethanol by use of laboratory magnetic stirrer. Consequently, two different matrix graphite powder samples were prepared, whose compositions are given in Table 1.

Table 1. Preparation of matrix graphite powder samples with different binder solutions

material	Phenol resin	PVB
N. graphite wt., g	128	128
E. graphite wt., g	32	32
Binder wt., g	40	4.24
Solvent wt., g	MeOH (73.6)	EtOH (109.3)
Binder fraction, wt%	20	2.58

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 as described in 2.2 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 373K. Milling of the prepared matrix graphite cake for each sample was carried out using an attrition mill at 280 rpm for 8 hrs. To prepare the green cylindrical compact samples, the matrix powder prepared are uni-axially compacted in a steel mold with about 3 grams of matrix powder sample. The compacting pressures applied were 1.0, 1.5, 2.0 and 2.5 tons/cm² and the compacting temperatures were room temperature, 373K, 393K and 423K. The heat treatment for carbonization was carried out for 2 hrs. at 1073K

and 1173K, respectively, in a flowing N₂ atmosphere by use of an electrical chamber furnace.

3. Results

3.1 Thermo-gravimetric analysis

Fig. 1 shows the evaporation behaviors during carbonization heat treatment of the compact samples mixed with 20 wt% phenol resin (blue line) and 2.58 wt% PVB (black line), respectively. The compact sample was prepared at room temperature with a compacting pressure of 2.5 tons/cm², having density of 1.83 and 2.02 g/cm³ for the compact with PR binder and that with PVB binder, respectively. The heat treatment was carried out following the temperature profile given in the figure (red line) in a flowing N₂ atmosphere. As shown in the figure, the total amount of the PVB binder admixed in the compact sample evaporated (calculated value: 2.55 wt%), while the evaporated amount of the PR binder admixed in the compact sample reached only to 11.7 wt%, which is 58.3% of the total amount of PR binder admixed in the compact sample. For other compact samples prepared at the compaction temperatures at 373K and 393K, the behaviors of binder evaporation were similar.

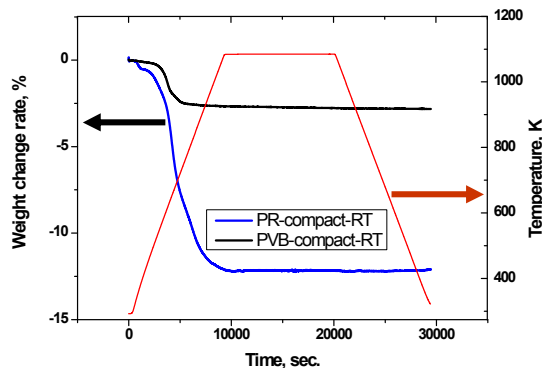


Fig. 1. Weight change of PR- and PVB-containing compact samples during the carbonization heat treatment

3.2 Analysis of carbonization heat treatment

Table 2 summarizes the total weight change fraction of compact samples prepared at different compaction temperatures and heat-treated in different atmospheres. It is noticeable that, for the compact samples with both PR and PVB binders, the weight change fraction is higher in N₂ atmosphere than in Ar. Also, with the compaction temperature increase, the weight change fraction tends to increase for the compact samples with PR binder, which implies that, even during the compaction at the temperature higher than 373K, the binder materials evaporate to some extent.

3.3 Density and its change of compact samples

As shown in Table 3, the density change rate is much larger for the compact samples with PVB binder than for those with PR binder. Also, the density after heat treatment increases as the compacting pressure increases and the compacting temperature does not influence strongly the density at the temperature higher than 373K. Generally, the density change decreases as the compacting temperature increases.

Table 2. Weight change fraction of compact samples prepared at different compaction temperatures and heat-treated in different atmospheres

Atmosphere	Ar atmosphere		N ₂ atmosphere	
	phenol resin	PVB	phenol resin	PVB
Room temperature	0.551	0.812	0.583	0.989
373K	0.557	0.813	0.592	0.991
393K	0.581	0.842	0.605	0.987
423K	0.583	0.845	0.602	0.988

Table 3. Density and its change rate of compact samples after carbonization heat treatment depending on compaction temperature and pressure

Compaction temperature (K)	Compaction pressure (ton/cm ²)	Density (g/cm ³) and density change rate (%)*	
		PR binder	PVB binder
Room temperature	1.0	1.61 (8.31)	1.61 (14.91)
	1.5	1.67 (7.86)	1.69 (13.80)
	2.0	1.72 (8.18)	1.74 (12.94)
	2.5	1.74 (8.10)	1.77 (12.27)
373	1.0	1.74 (6.74)	1.65 (13.21)
	1.5	1.76 (6.69)	1.72 (12.19)
	2.0	1.76 (6.50)	1.77 (11.28)
	2.5	1.77 (9.38)	1.80 (10.73)
393	1.0	1.70 (3.43)	1.66 (12.99)
	1.5	1.72 (3.69)	1.73 (12.06)
	2.0	1.70 (3.92)	1.76 (11.83)
	2.5	1.69 (1.30)	1.79 (10.74)
423	1.0	1.64 (1.90)	1.67 (12.74)
	1.5	1.65 (2.12)	1.72 (12.27)
	2.0	1.64 (3.87)	1.76 (10.91)
	2.5	1.67 (2.28)	1.79 (10.44)

*((green density - density after heat treatment)/green density) x 100 (%) and given in the parenthesis

4. Summary

- 1) The total amount of the PVB binder admixed in the compact sample evaporated during carbonization while the evaporated amount of the PR binder admixed in the compact sample reached only to 58.3% of the total amount.
- 2) The density change rate is much larger for the compact samples with PVB binder than for those with PR binder and the density after heat treatment increases as the compacting pressure increases.
- 3) Generally, the density change rate decreases as the compacting temperature increases.