Evolution of Matrix Density and Open Porosity of Graphite Matrix for HTGR Fuel Element

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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 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 evolution of matrix density and related open porosity is evaluated for different carbonization temperature profiles are employed, i.e., with rapid and slow increase rate of temperature, keeping other process parameters constant, such as binder content, carbonization time, temperature and atmosphere (two hours ant 1073K and N_2 atmosphere).

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. A phenolic resin was used for binder material. For the analysis of density and open porosity of compact samples, compact bulk density is measured with the geometrical method, i.e., measurement of compact dimension and weight and calculation of volume. For the immersion density and open porosity, a water immersion method is employed similar to one described in ASTM B 311-08.

2.2 Preparation of matrix graphite compact samples

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 binder solution, pre-prepared by dissolving phenolic resin in methanol to get an appropriate viscosity of the binder solution, 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/cm^2 and the compacting temperatures were room temperature (RT) and 373K. The heat treatment for carbonization was carried out for 2 hrs. at 1073K in a flowing N_2 atmosphere by use of an electrical chamber furnace. To vary the temperature profile with rapid and slow increase between RT and 1073K, the increase time of 4 hours and 18 hours and the cooling time of 10 hours and 18 hours were used, respectively, and referred as profile A and profile B.

3. Results and discussions

3.1 Compact bulk density and immersion density

Fig. 1 compares the bulk density and immersion density of compact samples compacted at RT and 373K with different compacting pressures and carbonized following the profile A. Both the bulk density and immersion density of green compacts and the carbonized compacts increases as the compacting pressure increases, except for the compact sample compacted at 373K and carbonized. The difference between the bulk density of green compact and carbonized compact compacted at RT is larger than that of immersion density. However, this observation is not really true for the compact samples compacted at 373K. This reveals that the evolution of density, open porosity in particular, of the compact samples compacted at RT and 373K is different in view of the microstructural modification during the carbonization.

Fig. 2 shows the bulk density and immersion density of compact samples compacted at RT and 373K with different compacting pressures and carbonized following the profile B. It can be seen that in general, the density variation behavior of the compact samples is quite different from that with profile A as shown in Fig.

1. It is observed that the density obtained with profile B is a little higher than that with the profile A for the compact samples under the sample condition. This implies that slow increasing temperature with profile B gives more evaporation of binder material embedded in the compact samples the with the profile A, hence, less binder material would be carbonized and left in the compact samples.
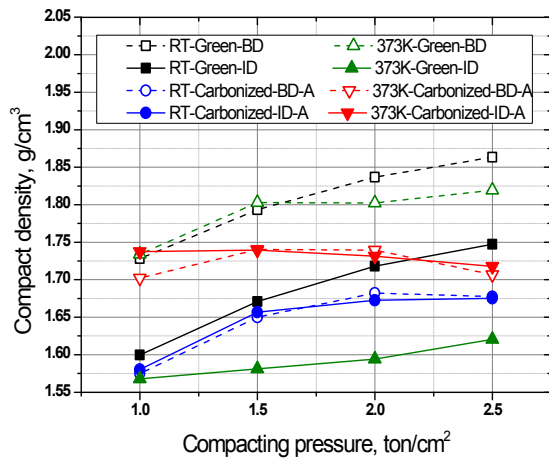

Fig. 1. Variation of bulk and immersion density of the compact samples compacted at RT and 373K with different compacting pressures and carbonized following the profile A as a function of compacting pressure. (BD: bulk density; ID: immersion density)
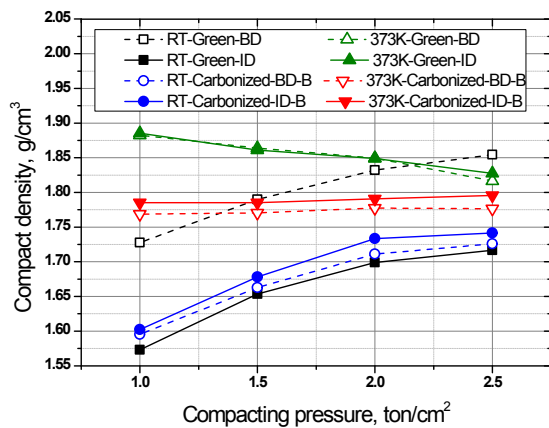

Fig. 2. Variation of bulk and immersion density of the compact samples compacted at RT and 373K with different compacting pressures and carbonized following the profile B as a function of compacting pressure. (BD: bulk density; ID: immersion density)

3.2 Porosity evolution of compact

Fig. 3 compares the open porosity of the compact samples compacted at RT and 373K with different compacting pressures and carbonized following the profile A and B. This figure clearly shows that the open

porosity increases after the carbonization, which makes density decrease of the compact samples. One can note that the increase in open porosity after carbonization is larger with the profile B than with the profile A.
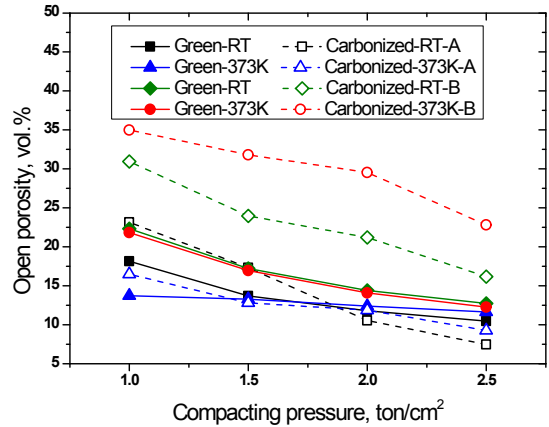

Fig. 3. Variation of open porosity of the compact samples compacted at RT and 373K with different compacting pressures and carbonized following the profile A and B. (BD: bulk density; ID: immersion density)

4. Summary

1) The difference between the bulk density of green compact and carbonized compact compacted at RT is larger than that of immersion density. However, this observation is not really true for the compact samples compacted at 373K. This reveals that the evolution of density, open porosity in particular, of the compact samples compacted at RT and 373K is different in view of the microstructural modification during the carbonization.

2) The density obtained with profile B is a little higher than that with the profile A for the compact samples under the sample condition. This implies that slow increasing temperature with profile B gives more evaporation of binder material embedded in the compact samples the with the profile A, hence, less binder material would be carbonized and left in the compact samples.

3) The open porosity increases after the carbonization, which makes density decrease of the compact samples. Also, the increase in open porosity after carbonization is larger with the profile B than with the profile A.