

## Dimensional Behavior of Matrix Graphite Compacts during Heat Treatments for HTGR Fuel Element Fabrication

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### 1. Introduction

The fuel element for an HTGR is manufactured by mixing coated fuel particles with matrix graphite powder and forming them 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 the preparation of the graphite matrix powder, over-coating the fuel particles, mixing the fuel particles with a matrix powder, carbonizing the green compact, and a 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. This carbonization step is followed by the final high temperature heat treatment where the carbonized compacts are heat treated at 2073~2173 K in vacuum for a relatively short time (about 2 hrs).

In order to develop a fuel compact fabrication technology, and for fuel matrix graphite to meet the required material properties, it is essential to investigate the relationship among the process parameters of the matrix graphite powder preparation, the fabrication parameters of fuel element green compact and the heat treatments conditions, which has a strong influence on the further steps and the material properties of fuel element. In this work, the dimensional changes of green compacts during the carbonization and final heat treatment are evaluated when compacts have different densities from different pressing conditions and different final heat treatment temperatures are employed, keeping other process parameters constant, such as the binder content, carbonization time, temperature and atmosphere (two hours at 1073K and N<sub>2</sub> atmosphere). It should be noted that for the simplicity of the experiment coated particles were not used and only the matrix graphite powder was used in the compact sample preparation.

### 2. Experimental

#### 2.1 Materials used and methods of analysis in the experiment

Natural graphite powder and electro-graphite powder, supplied by Graphit Kropfmühl AG, and SGL, Germany, respectively, were used. A phenolic resin was used for a binder material. For the analysis of density, compact bulk density is measured with a geometrical method, i.e., measurements of the compact diameter and height to calculate compact volume and weight to derive the compact density. All the dimensions are measured using digimatic micrometers and weights of the compacts were measured using a micro-balance. Dimensional variations of the cylindrical compact samples were calculated using the following relationship;

$$\text{Dim. variation (\%)} = (X_a - X_b) / X_b \times 100, \quad (1)$$

where,  $X_a$  and  $X_b$  are the diameter, height or density after the heat treatment and  $X_b$  are those before heat treatment.

#### 2.2 Preparation of matrix graphite compact samples

The mixing of the natural graphite and electro-graphite powder was carried out 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 cakes and drying for a total of 15 hrs at 373K. Milling of the prepared matrix graphite cake was carried out using an attrition mill at 280 rpm for 8 hrs. Fig. 1 shows a schematic flow diagram of the matrix graphite powder preparation. To prepare the green cylindrical compact samples, the matrix powder prepared was compacted in a steel mold with about 3 grams of matrix powder using a uni-axial single-acting press. The compacting pressures applied were 1.0, 1.5, 2.0 and 2.5 tons/cm<sup>2</sup> and the compacting temperatures were room temperature (RT) and 373K. The pressure holding time was 2 min. 30 secs. Other preparation details can be found in other literature. [1] Table 1 shows the dimensions of as-fabricated matrix graphite compact samples with the variables described above. The heat treatment for carbonization was carried out for 2 hrs. at 1073K in a flowing N<sub>2</sub> atmosphere by use of an

electrical chamber furnace. The final heat treatment was carried out in vacuum for 2 hrs at 2073 and 2173 K.

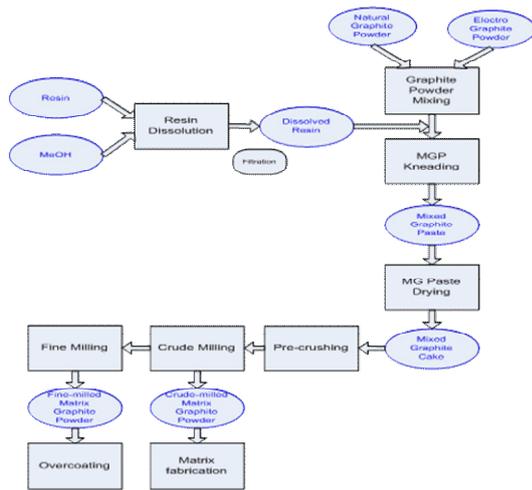


Fig. 2. Detailed process flow of graphite matrix powder preparation. [2]

Table I. Dimension of as-fabricated matrix graphite compact samples

sample ID	diameter	height	weight	density
#30-R-1	13.09	12.13	2.8145	1.726
#30-R-1.5	13.08	11.74	2.8276	1.793
#30-R-2	13.08	11.48	2.8207	1.830
#30-R-2.5	13.08	11.35	2.8257	1.854
#30-100-1	13.08	11.32	2.8412	1.869
#30-100-1.5	13.09	11.35	2.8240	1.851
#30-100-2	13.10	11.34	2.8280	1.852
#30-100-2.5	13.10	11.45	2.8235	1.833

\*Sample ID indicates the pressing conditions as follows: # batch no. – temperature (RT/100°C(373K)) – pressure (ton/cm<sup>2</sup>).

### 3. Results and Discussions

#### 3.1 Dimensional changes of matrix graphite green compacts during carbonization

Fig. 2 shows the variations of diameter, height and density of green graphite matrix compacts as functions of compaction pressure at two different compaction temperatures, i.e., room temperature and 373K. The diametral variation is quite small compared with those of height and density. It ranges between 0.08 and 0.15% for the compacts compacted at room temperature and -0.34 and -0.65% for those compacted at 373K. It remains essentially constant with compaction pressure increase at room temperature while it decreases slightly at 373K. The variation in height ranges from -2.89 to -3.79% along with compaction pressure increase at room temperature and from -4.15 to 5.76% at 373K, showing decreases at both temperatures. The variation in density increases

from -7.47 to -6.58% with increasing compaction pressure at room temperature and from -5.33 to -3.04% at 373K. The variation in weight is about -9.9% in average ranging from -9.81 to -10.04%, implying that the admixed phenolic resin vaporized during carbonization at a level of about half of the as-admixed amount (20 wt%) in the compacts.

There are two observations to be noted: 1) The diametral variation of compact compacted at room temperature differs to those compacted at 373K. While the compacts swell during carbonization they shrink at 373K. 2) The diametral and axial variations of compacts shows a large anisotropic behavior in shrinkage, as mentioned above, i.e., the diametral variation is small (ranging 0.08 ~ 0.15% at room temperature and -0.34 ~ -0.65% at 373K) while the axial variation is large (ranging -2.89 ~ -3.79% at room temperature and -4.15 ~ -5.76% at 373K). The first observation may be attributed to the different responding behaviors of phenolic resin during compaction at different temperatures. The second observation may be due to the compaction method. In this experiment, compaction was performed by a uni-axial single-acting press, which applies the compacting force to the axial direction only. During carbonization, the stresses caused by the force would be released in to the axial direction together with the phenolic resin vapor.

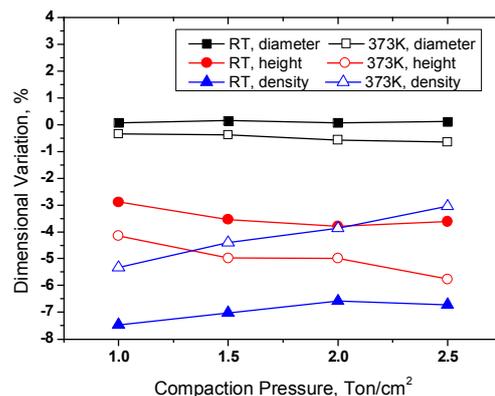
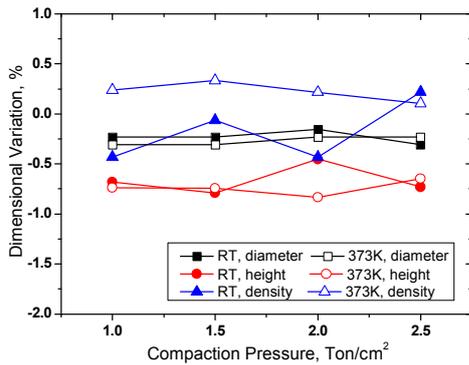


Fig. 2. Variations of diameter, height and density of green graphite matrix compact during carbonization as functions of compaction pressure and compaction temperature

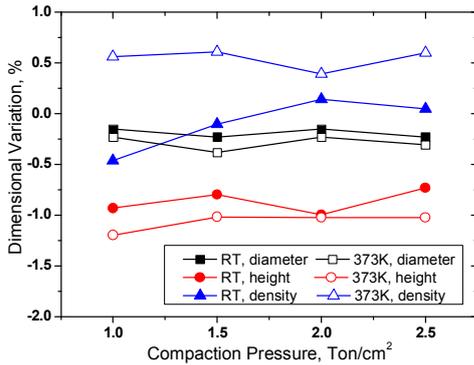
#### 3.2 Dimensional changes of carbonized matrix graphite compacts during final heat treatment

Fig. 3 (a) and (b) show the variations of diameter, height and density of the carbonized graphite matrix compacts as functions of compaction pressure at room temperature and 373K, heat-treated at 2073K and heat treated at 2173K, respectively. The differences between diametral and axial variations are reduced compared

with that after carbonization. The variations range only  $-0.15 \sim -0.30\%$  for diametral variation and  $-0.45 \sim -0.83\%$  for axial variation for the compacts heat-treated at 2073K, and  $-0.15 \sim 0.38\%$  for diametral variation and  $-0.73 \sim -1.12\%$  for axial variation for the compacts heat-treated at 2173K, respectively. However, it is to be noted that the variation in density increases and remains above zero in the case of compacts compacted at 373K at both heat treatment temperatures (2073 and 2173K), implying that these compacts were densified, regardless of the compaction pressure applied, and thus, regardless of as-fabricated density. The variation in weight showed  $-1.17\%$  for the compacts heat-treated at 2073K and  $-1.22\%$  for those heat-treated at 2173K, which imply that the final heat treatment vaporizes slightly further. However, there is no difference in weight decrease caused by the difference in temperature for final heat treatment.



(a)

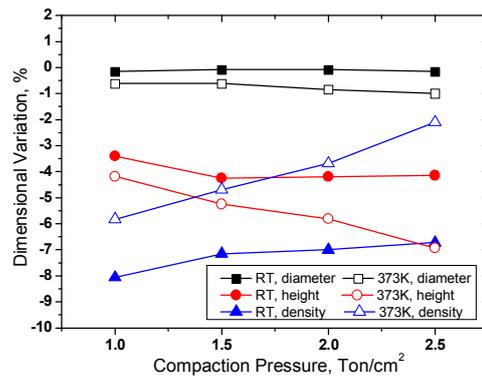


(b)

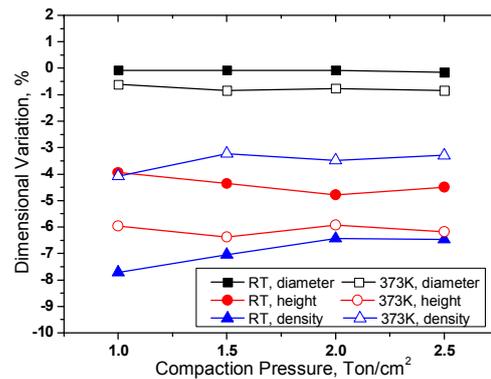
Fig. 3. Variations of diameter, height and density of carbonized graphite matrix compacts during final heat treatment as functions of compaction pressure at room temperature and 373K. (a) heat-treated at 2073K and (b) heat treated at 2173K

Fig. 4 (a) and (b) show the total variations of diameter, height and density of the green graphite matrix compacts during carbonization and final heat treatment as functions of compaction pressure at room temperature and 373K, heat-treated at 2073K and heat

treated at 2173K, respectively. Comparison of these two results reveals that the total variations of the diameter, height and density have similar behaviors at both heat treatment temperatures, implying that the heat treatment at the two temperatures give the same results in dimensional variations. However, small difference can be observed in the variation in density that for the compacts heat treated at 2073K shows the variation increases continuously as the compaction pressure increases, hence as-fabricated density increases when compacted at 373K, whereas it remains almost constant at higher pressure of 1.5 ton/cm<sup>2</sup> affected by the density decrease in as-fabricated compacts with pressure (see Table I).



(a)



(b)

Fig. 3. Variations of diameter, height and density of green graphite matrix compacts during carbonization and final heat treatment as functions of compaction pressure at room temperature and 373K. (a) heat-treated at 2073K and (b) heat treated at 2173K

#### 4. Summary

In this work, the dimensional variations of green compacts during the carbonization and final heat treatment are evaluated when compacts have different densities from different pressing conditions and different final heat treatment temperatures are employed. From this work, the followings are observed.

- 1) The diametral variation is quite small compared with those of height and density. It remains essentially constant with compaction pressure increase at room temperature while it decreases slightly at 373K.
- 2) The variation in height shows decreases with compaction pressure increase at both temperatures.
- 3) The variation in density increases increasing compaction pressure at room temperature and 373K. The variation in weight is about -9.9% in average ranging from -9.81 to -10.04%, implying that the admixed phenolic resin vaporized during carbonization at a level of about half of the as-admixed amount (20 wt%) in the compacts. The diametral variation of compact compacted at room temperature differs to those compacted at 373K. While the compacts swell during carbonization they shrink at 373K.
- 4) The diametral variation of compact compacted at room temperature differs to those compacted at 373K. While the compacts swell during carbonization they shrink at 373K. This observation may be attributed to the different responding behaviors of phenolic resin during compaction at different temperatures.
- 5) The diametral and axial variations of compacts shows a large anisotropic behavior in shrinkage, as mentioned above, i.e., the diametral variation is very small while the axial variation is large. This observation may be due to the compaction method. In this experiment, compaction was performed by a uni-axial single-acting press, which applies the compacting force to the axial direction only. During carbonization, the stresses caused by the force would be released in to the axial direction together with the phenolic resin vapor.
- 6) The total variations of the diameter, height and density have similar behaviors at both heat treatment temperatures, implying that the heat treatment at the two temperatures give the same results in dimensional variations. However, small difference can be observed in the variation in density that for the compacts heat treated at 2073K shows the variation increases continuously as the compaction pressure increases, hence as-fabricated density increases when compacted at 373K, whereas it remains almost constant at higher pressure of 1.5 ton/cm<sup>2</sup> affected by the density decrease in as-fabricated compacts with pressure.

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