

## Anisotropic Material Behavior of Uni-axially Compacted Graphite Matrix for HTGR Fuel Compact 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 generally 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 partially 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 developing the fuel compact fabrication technology, and fuel graphite material 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 and the material properties of fuel element.

It was observed, during this development, that the pressing technique employed for the compaction fabrication prior to the two successive heat treatments (carbonization and final high temperature heat treatment) was of extreme importance in determining the material properties of the final compact product. In this work, the material behavior of the uni-axially pressed graphite matrix during the carbonization and final heat treatment are evaluated and summarized along the different directions, viz., perpendicular and parallel directions to pressing direction. 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 preparation of matrix graphite compact samples

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. The mixing ratio of the raw materials is; natural graphite powder : artificial graphite powder : phenolic resin = 64 : 16 : 20 (in wt%).

In order to prepare the graphite powder mixture incorporating the phenolic resin, mixing of the natural graphite and artificial graphite powders was first carried out using a V-mixer for 1 hr at 100 rpm. And then, 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. Finally, milling of the prepared matrix graphite cake was carried out using an attrition mill at 280 rpm for 8 hrs to obtain the matrix graphite powder. 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 laboratory hand press. The compacting pressures applied were 1.0, 1.5 and 2.0 tons/cm<sup>2</sup> and the compacting temperatures were Room Temperature (RT) and 373K. The pressure holding time was 2 min. 30 secs. Compact samples were carbonized at 1173K for 38 hrs in a nitrogen atmosphere and finally heat-treated at 2073K and 2173K for 2 hrs in vacuum.

#### 2.2. Methods of material property analyses

Dimensions of compact samples were measured using digimatic micrometers to obtain the bulk volume and dimensional variations of the compact samples. The sample bulk density was calculated from the measured volume and the weight measured by a microbalance. Dimensional variation of the compact sample during carbonization heat treatment and linear thermal expansion co-efficient was measured using Thermo-Mechanical Analyzer (TMA) SETARAM, France. Thermal diffusivity of the compact sample after

final heat treatment was measured by the laser-flash method using Netzsch LFA 427, Germany, and the thermal conductivity ( $\lambda$ ) of the compact samples was calculated using the following equation:

$$\lambda = \rho \times C_p \times \alpha,$$

where,  $\rho$  is density of compact sample,  $C_p$  heat capacity and  $\alpha$  thermal diffusivity of compact sample. Compact samples were also tested on the Vickers micro-hardness using a Mitutoyo micro-hardness tester, MVK-G1, Japan.

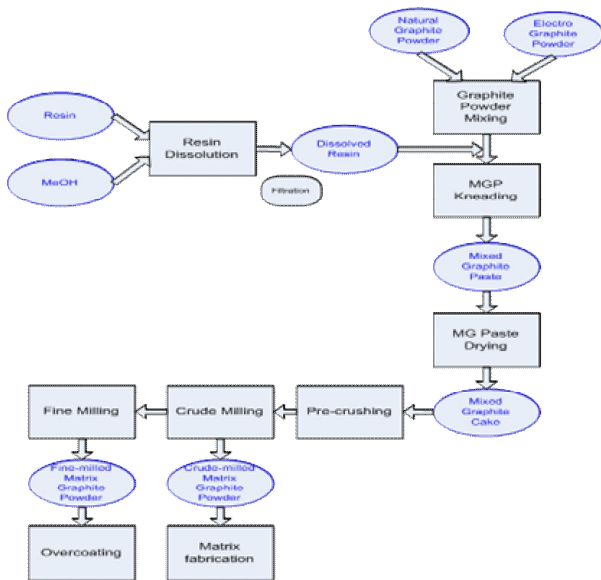


Fig. 1. Detailed process flow of graphite matrix powder preparation. [1]

### 3. Results and Discussions

#### 3.1 Dimensional changes of matrix graphite green compacts during carbonization and final heat treatment

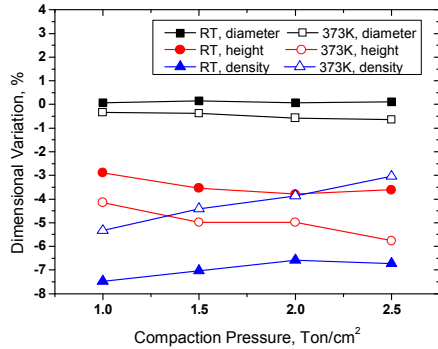
Fig. 2 (a) shows the variations of diameter, height and density of green graphite matrix compacts during the carbonization as functions of compaction pressure at two different compaction temperatures, i.e., room temperature and 373K. The diametral variation is quite small compared with those of height 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.

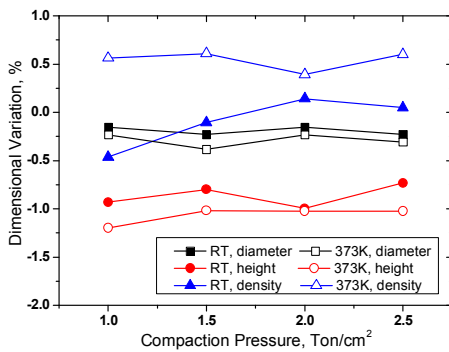
There are two observations to be noted: 1) the diametral variation of compact compacted at room temperature differs to those compacted at 373K. While the compacts swell during carbonization they shrink at 373K. 2) The diametral and axial variations of compacts shows a large anisotropic behavior in shrinkage, 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, as mentioned earlier in section 2.1, 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 (b) shows 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 2173K. The differences between diametral and axial variations are reduced compared with that after carbonization. The variations range only -0.15 ~ 0.38% for diametral variation and -0.73 ~ -1.12% for axial variation for the compacts heat-treated at 2173K. However, it is to be noted that the variation in density increases and remains above zero in the case of compacts compacted at 373K when samples were finally heat treated at 2173K. The heat treatment at 2073K showed also similar behaviors. This implies that these compacts were densified, regardless of the compaction pressure applied, and thus, regardless of as-fabricated density. The variation in weight showed -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.

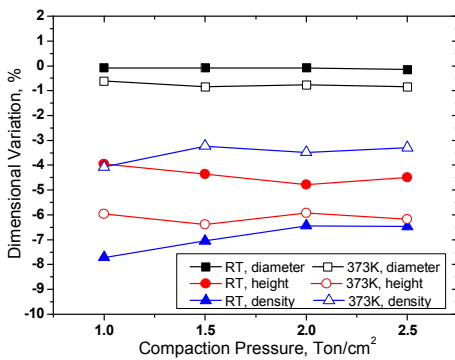
Fig. 2 (c) shows the total variations of diameter, height and density of the green graphite matrix compacts during carbonization and final heat treatment as functions of compaction pressure at room temperature and 373K and heat-treated at 2173K. It shows a further considerable decrease in height whereas there is practically no further variation in diameter. The variation in density shows positive for both samples compacted at RT and 373K, to a different extent of densification.



(a)



(b)

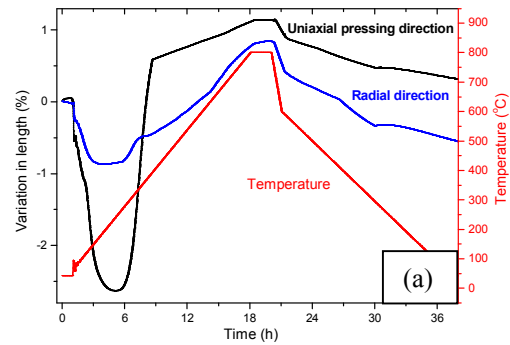


(c)

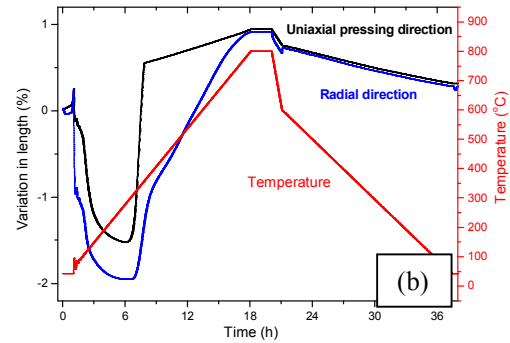
Fig. 2. Variations of diameter, height and density of green graphite matrix compact (a) after carbonization, (b) after final heat treatment (compared with carbonized compact samples) and (c) after final heat treatment (compared with green compact samples) as functions of compaction pressure and compaction temperature (final heat treatment temperature: 2173K)

### 3.2. Dimensional variation of compact samples in perpendicular and parallel directions during carbonization

Fig. 3 shows the dimensional variations of compact samples (density of  $1.74\text{g/cm}^3$ ) prepared (a) by uniaxially pressing (UP) and (b) by cold isostatic pressing (CIP) technique, in perpendicular (radial) and parallel (axial) directions to the pressing direction during carbonization heat treatment. Fig. 3 (a) shows a large difference of the dimensional variations along the two different directions: the variation along the axial direction (parallel to the compaction direction) is larger than that along the radial direction (perpendicular to the compaction direction). However, Fig. 3 (b) shows no practical difference in variations between the two different directions.



(a)



(b)

Fig. 3. Dimensional variations of compact samples in perpendicular and parallel directions to the pressing direction during the carbonization heat treatment: (a) sample prepared by uniaxially pressing and (b) sample prepared by cold isostatic pressing

### 3.3. Thermal expansion behavior of compact samples prepared by uni-axial pressing

Fig. 4 shows the thermal expansion behavior (variation of length) of the compact samples along the two different (axial and radial directions) when heated up to 1473K in He atmosphere. It shows a large difference in expansion behavior between perpendicular and parallel directions. Thermal expansion in perpendicular directions is more than

twice larger than that in parallel direction. It shows also that the sample with higher density varies less than the sample with lower density.

Fig. 5 shows the variation of the average linear thermal expansion coefficients between 673K and 1473K as a function of the compact bulk density along the axial and radial directions to the compaction direction. It can be observed that the coefficient is higher in radial direction than that in axial direction and it increases linearly with the density increase, though showing a large fluctuation.

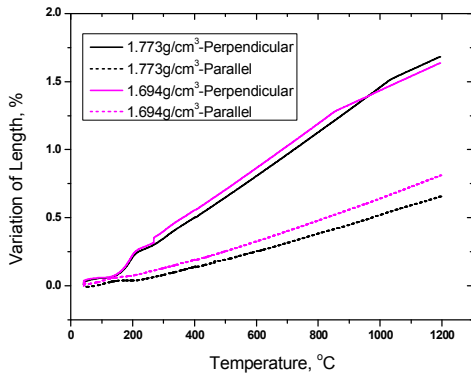


Fig. 4. Thermal expansion behavior of compact samples along the axial and radial directions when heated up to 1473K in He atmosphere

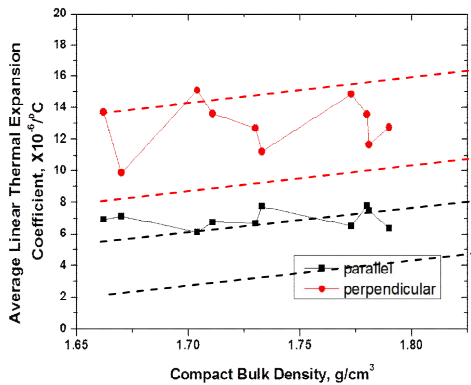
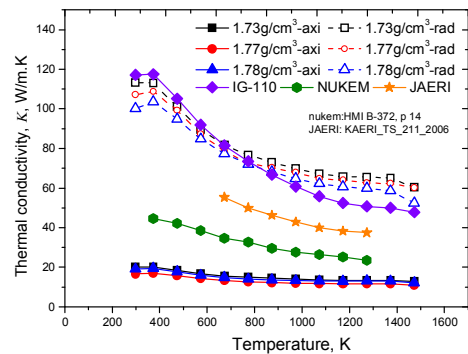


Fig. 5. Variations of average linear thermal expansion coefficients between 673K and 1473K as a function of the compact bulk density along the axial and radial directions to the compaction direction

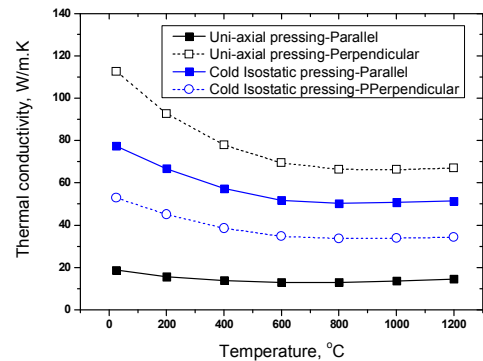
### 3.4. Variations of thermal conductivity of compact samples prepared by uni-axial pressing

Fig. 6 (a) shows the variations of thermal conductivities of uni-axially pressed compact samples of different densities along the axial and radial directions to the pressing direction and compared with

other published data from NUKEM [2] and JAERI [3]. The NUKEM data were obtained with their A3-27 matrix graphite pebble samples heat treated at 2223K (the method of thermal conductivity measurement not given). The JAERI data were obtained from the measurement of their fuel compact including coated fuel particles by the HTL method. In this figure, the thermal conductivity of uni-axially pressed compact samples in the radial (perpendicular) direction shows fairly high (about 6 to 3 times depending on temperature) compared with those in the axial (parallel) direction to the pressing direction, and the NUKEM and JAERI data show between these two variations. Fig. 6 (b) shows the difference in variations of thermal conductivities along the axial and radial directions for the compact samples prepared by uni-axial pressing and cold isostatic pressing.



(a)



(b)

Fig. 6. Variations of thermal conductivities matrix graphite : (a) uni-axially pressed compact in the axial and radial directions and comparison with other published data, (b) comparison of variations in two different directions between samples prepared by uni-axial pressing and cold isostatic pressing

### 3.5. Vickers Hardness measurement on the compact samples prepared by uni-axial pressing

Fig. 7 shows variations of Vickers hardness (Hv) of the compact samples on the perpendicular and parallel cross sections to the pressing direction. The samples used for the measurement had an average bulk density of about  $1.70\text{g/cm}^3$ , comprising between  $1.60$  and  $1.76\text{g/cm}^3$ . The hardness measured on the parallel section shows higher values than that measured on the perpendicular section. The average values of Vickers hardness on parallel and perpendicular section give  $176.6\pm 10.5$  MPa and  $125.6\pm 14.2$  MPa, respectively.

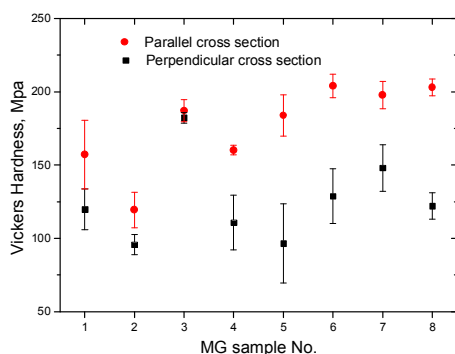


Fig. 7. Variations of Vickers hardness (Hv) of compact samples prepared by uni-axial pressing

#### 4. Summary

In this work, the dimensional variations and variations in thermal expansion, thermal conductivity and Vickers hardness of the graphite matrix compact samples in the axial and radial directions prepared by uni-axial pressing are evaluated, and compared with those of samples prepared by cold isostatic pressing with the available data. From this work, the followings are observed.

- 1) Dimensional changes of matrix graphite green compacts during carbonization show that the difference in radial and axial variations shows a large anisotropic behavior in shrinkage. The radial 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.
- 2) Dimensional variation of compact samples in perpendicular and parallel directions during carbonization shows a large difference in behavior when compact sample is prepared by uni-axial pressing. However, when compact sample is prepared by cold isostatic pressing, there is practically no difference in variation between axial and radial directions.

- 3) Thermal expansion behavior of compact samples prepared by uni-axial pressing show also an anisotropic behavior: Thermal expansion in perpendicular directions is more than twice larger than that in parallel direction.
- 4) The thermal conductivity of uni-axially pressed compact samples in the radial (perpendicular) direction shows fairly high (about 6 to 3 times depending on temperature) compared with those in the axial (parallel) direction to the pressing direction. However, the thermal conductivity of cold isostatic pressed compact samples show a much reduced difference between those in axial and radial directions.
- 5) The variations of Vickers hardness measured on the parallel section shows about twice higher values than that measured on the perpendicular section, the difference of which shows an anisotropic behavior.
- 6) The results obtained up to now imply anisotropic behaviors in material properties of the graphite matrix compact primarily due to the uni-axial pressing method.

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