PREPARATION OF EXFOLIATED GRAPHITE USING NATURAL GRAPHITE OF SRI LANKA

N.W.B. BALASOORIYA\textsuperscript{1} and P.W.S.K. BANDARANAYAKE \textsuperscript{2}

\textsuperscript{1}Faculty of Applied Sciences, South Eastern University, Sammanthurai (E.P.), Sri Lanka
\textsuperscript{2}Department of Physics, University of Peradeniya, Peradeniya, Sri Lanka

\textsuperscript{*}E-mail: balasooriya@fas.seu.ac.lk

ABSTRACT

Graphite Intercalation Compounds (GIC) with HNO\textsubscript{3} were chemically obtained using different morphologies of natural graphite from Sri Lanka, Madagascar and Krömul. X-ray powder patterns of the products showed the formation of second stage GIC (HNO\textsubscript{3}). These compounds were used to produce exfoliated graphite at 550°C in air. The expansion ratio was measured and compared with different types of natural expanded graphite. Accordingly, the pellet type Madagascar graphite and Kahatagaha-Kolongaha shiny, slippery, fibrous graphite (KSSI; one of the morphology of natural graphite of Sri Lanka) showed the greatest volumes of expansion. The samples with the largest average particle sizes had the largest expansion volumes, which can be explained by the proportionality between structural, morphological and textural characteristics of expanded and raw graphite types.

Key Words: natural graphite, exfoliated graphite, Sri Lanka, Kahatagaha-Kolongaha, Bogala

INTRODUCTION

Graphite usually forms intercalation compounds; both electron donor and electron acceptor molecules can be inserted between the planes of the original graphite structure. During intercalation, the graphite layers tend to slide, and change their characteristics stacking from ABABAB...... to G,IG,IG,IG....., where I is the intercalated (guest) species and G is a graphite layer. These species are distributed along the c-axis with same periodicity, called I\textsubscript{s} (Hérold, 1978, Hérold \emph{et al.}, 1994). The number “s” of graphite layers between two intercalate layers is called intercalate stage. The exfoliated graphites produced using graphite intercalation compounds (GIC) are on Figure 1.

The common way to produce exfoliated graphite is to intercalate graphite using compounds, which either evaporate themselves or decompose with gaseous products formation under thermal shock or microwave irradiation (Anderson, 1984, Tryba \emph{et al.}, 2005). The interlayer disperse pressure is supposed to appear between layers during fast heating of Graphite Intercalation Compounds (GIC); the intercalated substances flow out of graphite matrix both parallel and perpendicular to the layers. Thereby results give a foam structure formation with a non-stoichiometric product.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{Figure1}
\caption{Formation of the exfoliated graphite}
\end{figure}
Preparation of exfoliated graphite was also conducted by different heating systems, including inductively coupled plasma, laser irradiation and flame heating (Chung, 1987; Kang et al., 2002; Zhao et al., 2012; Skaltas et al., 2012). The expansion behaviour of the natural graphite has been studied for industrial use of exfoliated graphite (Chung, 2002; Sorokina et al., 2006; Tryba et al., 2000; Li et al., 2011; Lin et al., 2009). The development of exfoliated graphite technology was initiated with a two pronged approach viz. to develop cheaper value added products from naturally occurring graphite after making it amenable to provide a substitute for asbestos which is hazardous and banned in many countries (Wang et al., 2011). Flexible graphite foils were also prepared by rolling and pressing technology for compressing the exfoliated graphite (Lee et al., 2010).

The purpose of this study is to understand the expansion behaviour of different types of natural vein graphite and compare them with natural flake graphite of Madagascar and Krömful, Germany. The different types of graphite are characterized in the following (Table 1).

Table 1: Different Types of Natural Graphite

<table>
<thead>
<tr>
<th>Country &amp; Location</th>
<th>Occurrence</th>
<th>Graphite morphology</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sri Lanka:</td>
<td>Vein</td>
<td>Shiny, slippery, fibrous graphite (BSSI &amp; KSSI)</td>
</tr>
<tr>
<td>Bogala mine &amp; Kahatagaha-Kolongaha mine</td>
<td></td>
<td>Coarse flakes of radial graphite (BCFR &amp; KCFR)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Coarse striated, flaky graphite (BSCF &amp; KSCF)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Platy, needle graphite (BNPG &amp; KNPG)</td>
</tr>
<tr>
<td>Madagascar:</td>
<td>Flake</td>
<td>Graphite powder (E1099)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Graphite pellets (MGP)</td>
</tr>
<tr>
<td>Germany: Krömful</td>
<td>Flake</td>
<td>Graphite powder (UF4)</td>
</tr>
</tbody>
</table>

METHODS OF STUDY

Material: Sri Lanka vein graphite was collected from graphite mines at Bogala and Kahatagaha-Kolongaha. The selected graphite samples were crushed for a few minutes by using an agate mortar and separated to the particle size of 63 μm and approximately separated in to 1-2 mm particle size by using a mechanical sieve-shaker. Madagascar and Krömful graphite obtained from Carbone-Lorraine Ltd were used for this study.

Preparation of GIC (HNO₃): HNO₃ was used for chemical intercalation with natural graphite which has particle maximum sizes of 63μm and 1-2 mm. The graphite powder (1g) was mixed separately with 10 ml of fuming HNO₃ (MERCK) in 25 ml conical flask. The graphite intercalation compounds (GIC) were protected from air with a plastic cap and kept to intercalate for a few days or sometime more than a week under ambient conditions.

Characterization of the GIC (HNO₃): Characterization of the synthesized GICs was done by X-ray powder diffractometry (Cu Kα, Philips X-Ray generator). Thickness of the intercalate layers, interplanar distances and stages of the GIC (HNO₃) were studied using 001 diffraction pattern. Initially, the raw graphite samples were analysed by XRD with internal silicon standard. The thicknesses of the c-axis, Lc, were calculated using the Scherrer equation and the interlayer spacing (d₀₀₂) was determined from the Bragg equation (Iwashita et al., 2004). The SEM studies were done using a JSM 6400 scanning electron microscope.
Stage parameters of Graphite Intercalation Compounds (GIC): During the intercalation of acceptor compounds into graphite, it is sometimes not possible to obtain a pure stage compound and the GIC may have one of the following compositions.

(i) mixture of two, three or more pure stage compounds or
(ii) a solid solution compound with an average stage number.

The average stage number can be calculated and be given as a non-integer number.

Mixture of pure stage compounds: In GIC containing a mixture of two or three stages, the relative intensities of strongest reflections give an indication of the GIC stages present. Since intercalate density in galleries is approximately stage independent for GICs, it is useful to derive a single parameter that approximately scaled with intercalate content. Then the average staging parameter, \( n^\prime \), is calculated using the following equation (Zhang et al., 2000).

\[
n^\prime = \sum x_i n_i
\]

where \( x_i \) = Molar fraction of each stage of \( n_i \) compound

\( n_i \) = Stage number

Solid solution compounds: In some cases, the GIC is not constituted of a mixture of pure stage compounds, and non- integral X-ray reflections can be observed. Then the GIC can be considered as a solid solution compound consisting a disordered mixture of graphite planes and intercalated planes along the c-axis (Metz and Hohlwein, 1975). Then, there are two different calculation methods to obtain the average stage number as follows. First calculation made by Zhang et al., (2000) is based on these classical equations:

\[
I_c = d_c + (n-1)3.354 \, \text{Å}
\]

Where,

\( I_c \) = Cell parameter along the c-direction

\( d_c \) = Distance between two carbon planes adjacent to the intercalant species

\( d_e = d_c + 3.354 \, \text{Å} \) with \( e \) = the intercalant thickness

\( n \) = The stage number and \( I_c = (n+1) d_{\text{max}} \)

where, \( d_{\text{max}} \) is the strongest reticular distance (0 0 \( l \)) of the X-ray diffractogram which corresponds to the (0 0 \( n+1 \)) reflection in the case of a largest intercalate (\( e > 3.354 \, \text{Å} \)) or \( I_c = nd_{\text{max}} \), where \( d_{\text{max}} \) corresponds to the (0 0 \( n \)) reflection in the case of a smaller intercalate (\( e < 3.354 \, \text{Å} \)).

In the case of an acid intercalation, when \( e > 3.354 \, \text{Å} \), the combination of the two equations gives the average stage number:

\[
n^\prime = \frac{(d_{\text{max}} - d_1 + 3.354)}{(3.354 - d_{\text{max}})} \quad \text{with}
\]

\( d_1 = 7.96 \, \text{Å} \) for GIC(HSO\(_4\)) (Fuzellier, 1974)

\( d_1 = 7.79 \, \text{Å} \) for GIC(HNO\(_4\)) (Rudorff, 1954)

\( d_1 = 7.70 \, \text{Å} \) for GIC(HClO\(_4\)) (Bottomley et al., 1964)

The other calculation of average stage number proposed by Buscarlet et al., (1976) is more accurate. For that, the value of the two important reticular distances \( d_1 \) and \( d_2 \) (with \( d_1 < d_2 \)) on both side of the \( d_{(002)} \) line of the graphite (3.35 Å) are taken. The advantage of this calculation is the determination of

1. the intercalate size, \( e \), and
2. the average stage (or pseudo-stage), \( s \), without taking into account the value of the cell parameter \( d_c \).

\[
\frac{1}{d_1} - \frac{1}{d_2} = \frac{1}{3.354}
\]

For \( e < 3.35, \quad e = \left( \frac{1}{d_2} - \frac{1}{d_1} \right) \) \( (\text{where } d_1 < d_2) \)

\( d_1 = d \) for (0 0 \( n+1 \))

\( d_2 = d_{\text{max}} \) for (0 0 \( n \))
For \( e > 3.35 \),

\[
\frac{\frac{3.354}{d_1} - \frac{2 \times 3.354}{d_2}}{1 - \frac{1}{d_1}} = \frac{1}{d_2}
\]

(where \( d_1 < d_2 \) )

\[
d_1 = d \text{ for } (00 \ n+2)
\]

\[
d_2 = d_{max} \text{ for } (00 \ n+1) \text{ and }
\]

\[
x = \frac{e}{3.354 + n} + 1 - 3.354 \left( \frac{e}{3.354 + n} \left( \frac{1}{d_1} - \frac{1}{d_2} \right) \right)
\]

where \( n \) is the value of the lower stage number of the solid solution (integer number).

If \( 0 < x < 1 \), the pseudo stage \( s = n + x \)

**Preparation of exfoliated graphite (EG):**

The nitric acid was filtered off from the GIC (HNO\(_3\)) and the compound was washed with distilled water and then carefully dried at a maximum temperature of 105°C in order to prevent expansion. For measuring expansion behaviour, the dried compound was filled into a measuring cylinder with an internal diameter of 35 mm and a capacity of 100 ml. The cylinder was well-shaken until the level of the compound was constant. The compound was carefully put into a pre-heated (550°C) quartz crucible in a chamber furnace and kept for a few minutes to exfoliate the compound. The expansion factor (degree of expansion) was determined as the volume ratio of exfoliated graphite to its precursor; \( V_{EG} / V_{GIC} \) was calculated to measure the change in sample volume before and after exfoliation.

**RESULTS AND DISCUSSION**

**Chemical intercalation with HNO\(_3\):**

Intercalation of acceptor type acids creates positive holes in the graphite layers. The intercalation between carbon atoms and intercalated ions is supposed to be strictly ionic and the delocalised charge gives a metallic behaviour along the graphene layers (Métrot and Fisher, 1981). Chemical intercalation of the graphite with the acceptor type acids can be shown as

\[
nC (\text{graphite}) + x\text{HNO}_3 \Leftrightarrow C_n^+ (\text{HNO}_3)_{x1} \text{ NO}_3^- + H^+ + e^{-}
\]

(Hérold, 1978)

\[
\text{HNO}_3 + e^- + H^+ \Leftrightarrow \text{NO}_2 + H_2O
\]

**Characterization of the GIC:** 

Interlayer nitric acid can be intercalated without the intervention of reagents because of their own oxidizing nature; only the second stage GIC (HNO\(_3\)) was observed in X-ray diffractogram with and without the use of an oxidant (Figure 2). The reflections of the second stage compound were predominant with an average \( d_{max} = 7.80 \) Å but additional reflexes of third stage GIC, near the position of graphite (020) were also observed without using the oxidant. The second stage compound was predominant when P\(_2\)O\(_5\) was used as an additive. Table 2 shows the variation of interplanar distances of the GIC (HNO\(_3\)) compared with some reference values.

**Average stage number calculation using XRD peak intensities:**

The example from Figure 2 for intercalation of nitric acid without oxidant indicates that the mixture of pure 2\(^{nd}\) and 3\(^{rd}\) stage compounds are obtained. The intensity \( I \) for the strongest peaks (in the same 2\(\theta\) zone) of X-ray diffractograms is measured. Graphite nitrate obtained in fuming HNO\(_3\) acid has a \( d_{max} = 7.80 \) Å which is reported correspond to a composition C\(_{5n}\)HNO\(_3\) (n is the stage). Figure 3 shows that NO\(_3^-\) plane is perpendicular to the graphene planes and all the oxygen atoms are in contact with carbon atoms (Touzain, 1979-80).
### Table 2: The variation of interplanar distance of the GIC (HNO$_3$) and comparison with some reference values

<table>
<thead>
<tr>
<th>Graphite morphology</th>
<th>Particle size</th>
<th>$d_i$ (Å)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Natural (Krömful and Ceylon)</td>
<td>Fine</td>
<td>7.84</td>
<td>7.79</td>
</tr>
<tr>
<td>Natural (Madagascar)</td>
<td>1-2mm, flakes</td>
<td>7.80</td>
<td></td>
</tr>
<tr>
<td>BSSI</td>
<td>63µm</td>
<td>7.82</td>
<td>7.72</td>
</tr>
<tr>
<td>BCFR</td>
<td></td>
<td>7.76</td>
<td>7.82</td>
</tr>
<tr>
<td>BSCF</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>BNPG</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>KSSI</td>
<td>63µm</td>
<td>7.80</td>
<td>7.80</td>
</tr>
<tr>
<td>KCFR</td>
<td></td>
<td>7.79</td>
<td>7.79</td>
</tr>
<tr>
<td>KSCF</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>KNPG</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>E1099</td>
<td>40µm</td>
<td>7.80</td>
<td></td>
</tr>
<tr>
<td>UF4</td>
<td>4µm</td>
<td>7.78</td>
<td></td>
</tr>
<tr>
<td>BSSI</td>
<td>63-250µm</td>
<td>7.75</td>
<td>7.74</td>
</tr>
<tr>
<td>BCFR</td>
<td></td>
<td>7.81</td>
<td>7.79</td>
</tr>
<tr>
<td>BSCF</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>BNPG</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>KSSI</td>
<td>63-250µm</td>
<td>7.79</td>
<td>7.74</td>
</tr>
<tr>
<td>KCFR</td>
<td></td>
<td>7.79</td>
<td>7.79</td>
</tr>
<tr>
<td>KSCF</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>KNPG</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*Figures and text from the document:*

- *Figure 2: X-ray diffractograms for the GIC (HNO$_3$), prepared from BSSI graphite with and without a chemical additive.*

- Avg. stage number, $n' = 1.955$ (95.5% of 2$^{nd}$ stage GIC and 4.5% of 3$^{rd}$ stage GIC)
- Avg. stage number, $n' = 2.803$ (80.3% of 3$^{rd}$ stage GIC and 19.7% of 2$^{nd}$ stage GIC)
Figure 3: Structure of GIC(HNO$_3$) along graphite C-axis ($I_c$ = The repeat distance along the c-axis, for the first stage compound $I_c=d_i$ (Interplanar distance) and $e$ = thickness of the intercalated stage)

**Average stage number calculation using interplanar distances:**
The GIC(HNO$_3$) obtained from KSCF Sri Lanka graphite coarse sample (from Table 3) was considered as an example. Two reticular distances which are in both sides of the $d$ (002) graphite line has been measured:

$$d_i = 2.7993 \, \text{Å}$$
$$d_s = d_{\text{max}} = 3.7341 \, \text{Å}$$

The average stage number $n'$ can be calculated with the value of the $d_i$ parameter ($d_i = 7.79 \, \text{Å}$).

$$n' = \frac{3.7341 - 7.79 + 3.354}{3.354 - 3.7341} = 1.847$$

This value corresponds to 85% of 2$^\text{nd}$ stage GIC and 15% of 1$^\text{st}$ stage GIC.

In fact, above calculation is not accurate because it considers that the average stage $n'$ corresponds to a theoretical compound with a hypothetical value $I_c$ of cell parameter which is not realistic.

<table>
<thead>
<tr>
<th>Graphite type</th>
<th>Particle size (µm)</th>
<th>$d_{\text{max}}$ (Å)</th>
<th>$s$ (Pseudo stage)</th>
<th>$n'$ (Avg. stage number)</th>
<th>Thickness of the interlayer, $e$ (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BSSI</td>
<td>63</td>
<td>3.6385</td>
<td>2.956</td>
<td>2.803</td>
<td>4.485</td>
</tr>
<tr>
<td></td>
<td>63-250</td>
<td>3.6989</td>
<td>1.996</td>
<td>2.137</td>
<td>4.394</td>
</tr>
<tr>
<td>BSSI (with additive)</td>
<td>63</td>
<td>3.7259</td>
<td>2.017</td>
<td>1.955</td>
<td>4.486</td>
</tr>
<tr>
<td>KSSI</td>
<td>63</td>
<td>3.7434</td>
<td>2.012</td>
<td>1.779</td>
<td>4.534</td>
</tr>
<tr>
<td></td>
<td>63-250</td>
<td>3.7118</td>
<td>1.996</td>
<td>2.024</td>
<td>4.430</td>
</tr>
<tr>
<td>BSCF</td>
<td>63</td>
<td>3.6958</td>
<td>2.028</td>
<td>2.168</td>
<td>4.398</td>
</tr>
<tr>
<td></td>
<td>63-250</td>
<td>3.7141</td>
<td>2.020</td>
<td>2.005</td>
<td>4.448</td>
</tr>
<tr>
<td>KSCF</td>
<td>63</td>
<td>3.7527</td>
<td>2.019</td>
<td>1.714</td>
<td>4.560</td>
</tr>
<tr>
<td></td>
<td>63-250</td>
<td>3.7341</td>
<td>1.996</td>
<td>1.847</td>
<td>4.492</td>
</tr>
<tr>
<td>BCFR</td>
<td>63</td>
<td>3.7118</td>
<td>2.035</td>
<td>2.024</td>
<td>4.446</td>
</tr>
<tr>
<td></td>
<td>63-250</td>
<td>3.6815</td>
<td>2.071</td>
<td>2.305</td>
<td>4.364</td>
</tr>
<tr>
<td>KCFR</td>
<td>63</td>
<td>3.7027</td>
<td>2.054</td>
<td>2.103</td>
<td>4.422</td>
</tr>
<tr>
<td></td>
<td>63-250</td>
<td>3.6868</td>
<td>2.036</td>
<td>2.254</td>
<td>4.370</td>
</tr>
<tr>
<td>BNPG</td>
<td>63</td>
<td>3.7225</td>
<td>2.065</td>
<td>1.936</td>
<td>4.490</td>
</tr>
<tr>
<td></td>
<td>63-250</td>
<td>3.6853</td>
<td>2.125</td>
<td>2.266</td>
<td>4.390</td>
</tr>
<tr>
<td>KNPG</td>
<td>63</td>
<td>3.7141</td>
<td>2.030</td>
<td>2.005</td>
<td>4.450</td>
</tr>
<tr>
<td></td>
<td>63-250</td>
<td>3.7279</td>
<td>1.991</td>
<td>1.894</td>
<td>4.479</td>
</tr>
<tr>
<td>E1099</td>
<td>40</td>
<td>3.7072</td>
<td>2.038</td>
<td>2.064</td>
<td>4.570</td>
</tr>
<tr>
<td>UF4</td>
<td>4</td>
<td>3.7241</td>
<td>2.025</td>
<td>1.924</td>
<td>4.470</td>
</tr>
<tr>
<td>HOPG</td>
<td>250</td>
<td>3.6951</td>
<td>2.007</td>
<td>2.172</td>
<td>4.350</td>
</tr>
</tbody>
</table>
Therefore, the thickness of the interlayer, \( e \) and pseudo-stage, \( s \) was calculated as below:

\[
e = \left[ 1 + \frac{3.354}{2.7993} \times 3.354 \right] \left[ 1 - \frac{1}{2.7993} - \frac{1}{3.7341} \right] = 4.492 \text{ Å and with } n=1
\]

\[
s = 1 + \left( \frac{e}{3.354} + 1 + 1 \right) \left[ 1 - 3.354 ( \frac{e}{3.354} + 1 ) \left( \frac{1}{2.7993} - \frac{1}{3.7341} \right) \right]
\]

\( s = 1.996 \) corresponds to 99.6% of 2\textsuperscript{nd} stage GIC and 0.4% of 1\textsuperscript{st} stage GIC, i.e., practically pure second stage.

\[
I_c = 99.6\% \left( e + n \times (2^n) \times 3.354 \right) + 0.4\% \left( e + 1 \times 3.354 \right)
\]

\[
= 11.19 \text{ Å,}
\]

The \( d_1 \) and \( d_2 \) values were recalculated as:

\[
d_1 = I_c/(s+2) = 10.63/(1.996+2) = 2.799 \text{ Å}
\]

\[
d_2 = I_c/(s+1) = 10.63/(1.996+1) = 3.734 \text{ Å close to the original values.}
\]

This calculation is more accurate because it considers that the obtained pseudo-stage’s corresponds to a hypothetical average cell parameter.

**GIC (HNO\textsubscript{3}-H\textsubscript{2}SO\textsubscript{4}) system:** The second stage GIC (HNO\textsubscript{3}) is obtained when graphite powder reacts with the mixture HNO\textsubscript{3}:H\textsubscript{2}SO\textsubscript{4} in the ratio of 1:1 & 5:1. H\textsubscript{2}SO\textsubscript{4} acts as the additional oxidizer. It can be explained by using Figure 4, the value of the pseudo stage \( (s = 2) \) is constant achieving the second stage GIC (HNO\textsubscript{3}). The predominant first stage GIC (H\textsubscript{2}SO\textsubscript{4}) is obtained mixing graphite with the mixture in the ratio of 1:5. But some peaks of second stage GIC (HNO\textsubscript{3}) also appeared in Figure 4. Finally, no evidence for co-intercalation has been observed with the mixture of nitric acid and sulphuric acid, but only parallel intercalations have formed. Graphite co-intercalation compounds do not represent a new class of substances. In this case the co-intercalation may not form mixed nuclei or mixed intercalated layers.

In the system of GIC (HNO\textsubscript{3} –R), (where R=H\textsubscript{2}SO\textsubscript{4}, HClO\textsubscript{4}) R acts as the oxidant and increases intercalation. The formation of the intercalation compounds with mixing solutions mainly depends on the concentration of the strongest acid.

![Figure 4: X-ray diffractograms for the GIC’s in HNO\textsubscript{3}-H\textsubscript{2}SO\textsubscript{4} system, prepared using BSSI graphite.](image-url)
Exfoliated graphite: SEM images show the natural and acid treaded graphite before the exfoliation and exfoliated graphite (Figure 5).

![SEM images of graphite before and after exfoliation](image)

Figure 5: SEM images of: (a) natural BSSI graphite (NG), (b) acid-treated BSSI graphite, (c) and (d) exfoliated graphite (EG).

The degree of expansion during the exfoliation: Figures 6a-6h show the behaviour of graphite exfoliation with time. These photographs were taken using a camera with the speed of 3000 images per second.

The better degree of crystallization of the graphite and the more regular structure and shape of the flakes increase the degree of expansion (Inagaki et al., 1983). Accordingly, Table 4 shows that the pellets-types of graphite (MGP and KSSI coarse particles) have the greatest volume of expansion. The MGP graphite is very different from the others. This graphite has mechanically sorted pellets that have uniform diameter. The same conditions were used to exfoliate the MGP graphite.

The other types of natural graphite show lower degree of expansion (2.5). In this case, gentle preparation, especially grinding to preserve the pellet-shape, is very important and will be worthwhile. Fine particles of different types of natural graphite and synthetic graphite showed lower expansion. The samples with the largest average particle sizes had the largest expansion volumes, which can be explained by the proportionality between the structural, morphological and textural characteristics of the expanded and the raw graphites.
Figure 6a: Before start to exfoliate (0.00 Second)  
Figure 6b: After 0.072 Seconds

Figure 6c: After 0.125 Seconds  
Figure 6d: After 0.193 Seconds

Figure 6e: After 0.411 Seconds  
Figure 6f: After 0.674 Seconds

Figure 6g: After 0.864 Seconds  
Figure 6h: Exfoliated graphite 1.265 Seconds

Figure 6: The behaviour of graphite exfoliation
Table 4: Show the degree of expansion of coarse and fine particles of natural graphite

<table>
<thead>
<tr>
<th>Type of graphite</th>
<th>Particle size</th>
<th>Degree of expansion</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Fine (µm)</td>
<td>Coarse (mm)</td>
</tr>
<tr>
<td>BSSI</td>
<td>63</td>
<td>≈1-2</td>
</tr>
<tr>
<td>BCFR</td>
<td>63</td>
<td>≈1-2</td>
</tr>
<tr>
<td>BSCF</td>
<td>63</td>
<td>≈1-2</td>
</tr>
<tr>
<td>BNPG</td>
<td>63</td>
<td>≈1-2</td>
</tr>
<tr>
<td>KSSI</td>
<td>63</td>
<td>≈1-2</td>
</tr>
<tr>
<td>KCFR</td>
<td>63</td>
<td>≈1-2</td>
</tr>
<tr>
<td>KSCF</td>
<td>63</td>
<td>≈1-2</td>
</tr>
<tr>
<td>KNPG</td>
<td>63</td>
<td>≈1-2</td>
</tr>
<tr>
<td>E1099</td>
<td>10</td>
<td></td>
</tr>
<tr>
<td>MGP</td>
<td></td>
<td>≈1-2</td>
</tr>
<tr>
<td>UF4</td>
<td></td>
<td>4</td>
</tr>
<tr>
<td>KS25</td>
<td>25</td>
<td></td>
</tr>
</tbody>
</table>

CONCLUSIONS

The exfoliated graphite has been prepared using different morphologies of natural graphite from Sri Lanka, Madagascar and Krömful. The exfoliation behaviour of the natural graphite mainly depends on the method of synthesis of the exfoliated graphite, in particular, on GIC stage number and temperature of thermal treatment. In addition, the graphite particle size, the edge dimension and the textural characteristics are parameters that influence the expansion behaviour of the EG as well as the intercalation and the kind of intercalate used. Therefore, further studies have to be carried out for utilizing Sri Lankan natural graphite for preparation of exfoliated graphite as a cheaper value-added product.

REFERENCES


