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Thermal properties of natural graphite flake composites

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Abstract

Graphite flake composites are important thermal management materials with strong potential in applications such as electronic cooling devices and aerospace materials. Here we present thermal properties of some model composites with various graphite flake sizes following carbonisation and graphitisation. The thermal diffusivity is measured by standard laser flash and an in-build line heat-source method and a ratio of graphitic peak to disorder peak from Raman spectrum is used to calculate a-direction coherent length using empirical equation. Thermal conductivities significantly higher than that of copper have been calculated for certain compositions.

Keyword: Natural graphite, thermal conductivity, graphite composites.

INTRODUCTION

There is a continuing need for light weight, high performance heat sinks and heat spreaders to meet the needs of growing technology (Norley J. and Chen G., 2002). Amongst the different classes of materials nowadays being considered in electronics, carbon/metal composites have two main competitors Al/SiC composites (thermal conductivity below 250 W/m K) and diamond/metal composite family (thermal conductivity ranging from 350 W/m K to 780 W/m K) (Prieto R. et al, 2008). Graphite is considered to be one of the potential material because of its high in-plane thermal conductivity.

Researchers at Oak Ridge National Laboratory (ORNL) developed a graphite foam having thermal conductivity equivalent to that of Aluminum (In plane, k=201 W/m/K) at $1/5^{th}$ the weight (Klett J, 2001).

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Graf-tech International Ltd developed natural graphite/epoxy laminate material (eGraf^{TM}) for plasma display panels (PDP). The in-plane thermal conductivity of this laminate was reported to be ~370 W/m/K, which is 77% higher than aluminum and comparable to that of copper. Following advances in the material development, an in-plane thermal conductivity of approximately 400 W/m/K (i.e. approximately equal to that of copper) was achieved in this composite (Norley J., 2003).

eGraf's product 'Spreadershield^{TM}' are made from chemical treatment on natural graphite (intercalation) and used as heat-spreader in for example, notebooks and digital cameras. eGraf 1200 is reported to have an in-plane thermal conductivity of 120 W/m/K (Technical Bulletin 268M).

Mariner et al (1999) invented electrically conductive composites by combining thermally treated graphite flakes with a polymeric binder, giving thermal conductivity greater than 100 W/m/K.

Prieto et al (2008) prepared different composites with different reinforcements for Al-Si and Ag-Si alloys, that is, range of different proportions of graphite flakes and carbon fibers, graphite flakes and SiC, diamond, graphite particles, carbon fibers and graphite foam. They found the graphite flake/ metal composites exhibited superior thermal properties (in-plane thermal conductivity ranging from 350 W/m K to 548 W/m K) representing a clear alternative to commercialized Al/SiC material as they are cheaper and machinable.

Here, we will discuss thermal properties of some model composites with various graphite flake sizes following carbonization and graphitization with 75% starting volume fraction of graphite. It will be shown that some composites developed in this work have thermal conductivity higher than that of copper with a simple low cost fabrication process compared to that of synthetic graphite/carbon fiber composites.

EXPERIMENTAL PROCEDURES

Composites with three different average flake sizes; small (S) with avg. flake size: 180 μm, medium (M) with avg. flake size: 300 μm and large (L) with avg. flake size: 600 μm have

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been prepared. The large flake composites were specially supplied by Branwell graphite Ltd for experimental purpose.

Preparation of composite

The composites were made using precursors (a) graphite flakes of known average flakes size supplied by Branwell graphite ltd (b) Mesophase pitch supplied by Mitsubishi and (c) Ammonium ligno-sulfonate (ALS). The mesophase pitch supplied in the form of pellets was ground into fine powder and passed through a sieve of 35 micron.

The precursor mixture consisting of natural graphite flakes, ammonium lingo-sulfonate and mesophase pitch binder was degassed at 450 ºC and hot-pressed to obtain a composite.

Characterization and measurement

Thermal conductivity of the samples was calculated using the relation between thermal diffusivity, bulk density and specific heat. The specific heat of the samples was estimated using Spencer's formula (Hust J.G., 1984), the density was calculated from the weight and dimensions of the sample and the thermal diffusivity was measured using the conventional laser flash method and a line heat source method developed at Tyndall Institute, Cork.

Raman spectroscopy was used to measure the intensity ratio of graphitic peak (I_G) to disorder peak (I_D) and coherent length was calculated using empirical equation suggested by Knight D.S. and White W.B. (1989)

$$L_a = C\frac{I_G}{I_D} \times 10^{-10}\ \mathrm{m}$$

Where:

C = 44Å for exciting wavelength of the laser, λ_L=514.5 nm, $I_G \rightarrow$ Intensity of the G-peak (λ~1575 Å) & $I_D \rightarrow$ Intensity of the D-peak (λ ~1350 Å)

THERMAL DIFFUSIVITY AND ESTIMATION OF THERMAL CONDUCTIVITY OF COMPOSITES WITH 75% STARTING VOLUME FRACTION OF GRAPHITE

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All the measurements were carried out at room temperature on samples heat treated (HT) to 1000 °C, 1600 °C and graphitised to ~3000 °C. It was not possible to measure thermal diffusivity of as-prepared samples as the pitch was seen to soften with the heat of the laser beam.

Samples heat-treated to 1000 °C prepared from medium and large flakes were found to have diffusivities in the range between 14×10^{-5} m^2/s to 15×10^{-5} m^2/s and 21×10^{-5} m^2/s to 25×10^{-5} m^2/s respectively. The samples prepared from small flakes and heat-treated to 1000 °C are assumed to have poor conductivity since no signal could be detected while measuring thermal diffusivity.

Thermal diffusivities and the estimated thermal conductivities of the samples heat-treated to 1600 °C are summarized in table 1 below

TABLE 1 Diffusivity and estimated thermal conductivity of samples HT to 1600 °C of different flake sizes

Flakes forming the composite	Average of diffusivity obtained by laser flash and line heat source method ($\times10^{-5}$ m^2/s)	Thermal conductivity of the sample (W/m K)
S	4.54±01	52
M	15.47±01	220
L	24.98±01	325

From the above table it can be again seen that the thermal diffusivity and hence the thermal conductivity increases significantly with the flake size. The results of the graphitised samples are shown in the figure 1.

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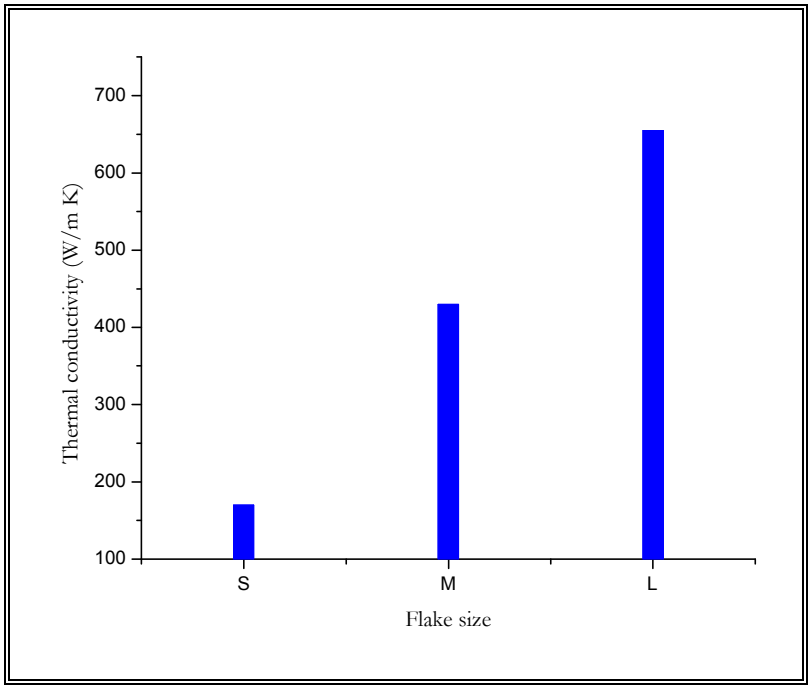


Figure 1 Thermal conductivity of graphitised composites (75% starting volume fraction of graphite) versus flake sizes

Again it was found that that when the samples are graphitised, the thermal conductivity increases as the flake size increases. The graphitised samples prepared using large flakes are found to have thermal conductivity of ~655 W/m K which is more than one and half times that of copper and with density only one fourth of that of copper.

a-direction coherent length measurement using Raman Studies on the binder of composites

Figure 2 shows Raman spectra of large flake composites heat-treated to different temperatures. It can be seen that ratio of the disorder peak (~1360 cm^{-1}) to graphitic peak (1575 cm^{-1}) decreases as the heat-treatment temperature (HTT) increases. Also, the graphitic peak (1575 cm^{-1}) and the second order peaks are seen to get more defined with an increase in HTT.

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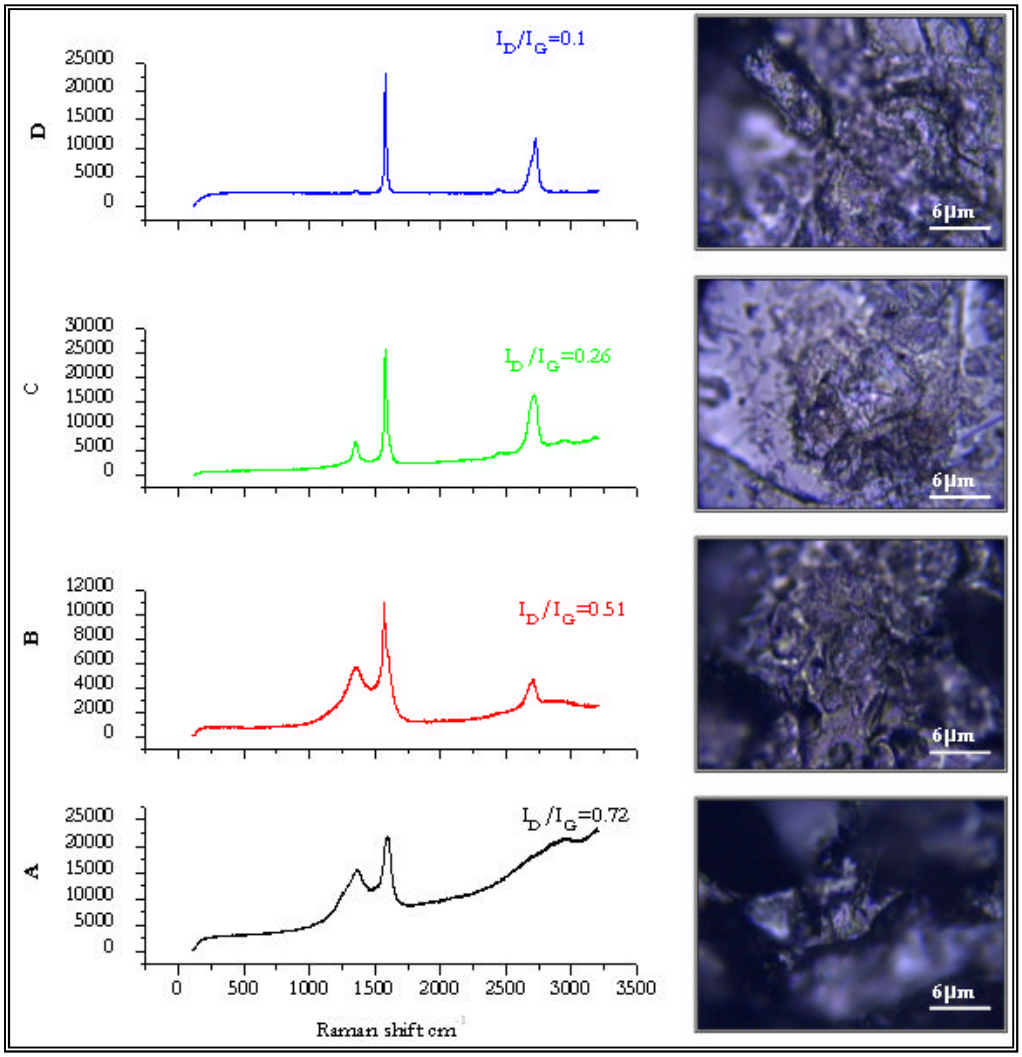


Figure 2 Raman spectra from the pitch of samples prepared using large flake graphite at different HTT (A) as prepared, (B) carbonised to 1000°C, (C) HTT 1600°C and (D) graphitised (~2900°C). The micrographs show the area of the sample at which spectra were acquired.

Using the empirical relation between coherence length and the intensity ratios, the a-direction coherence length was calculated and it was found to increase with HTT (fig.3). It can be seen that there is a huge increase in the coherence length (from 169Å to 440Å) when the heat-treatment temperature is increased from 1600ºC to 2900ºC which is attributed to the graphitisation of the binder.

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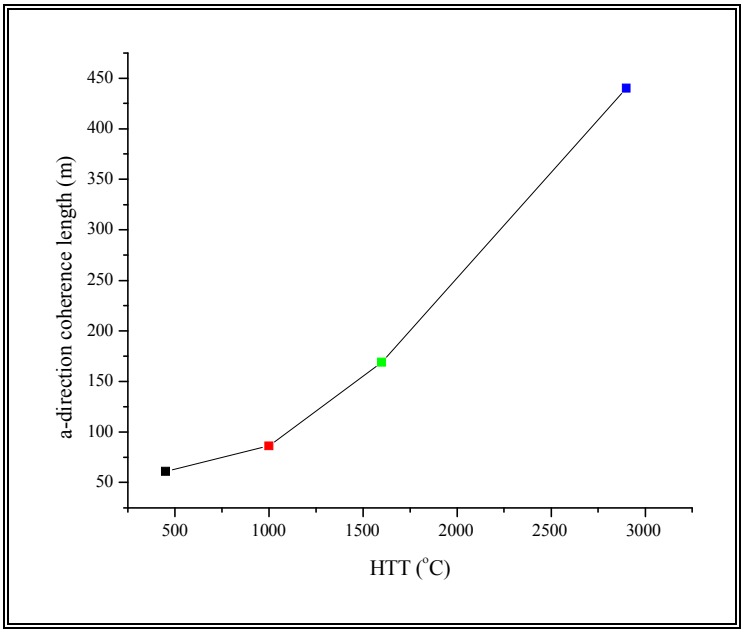


Figure 3 Graph showing increase in a-direction coherence length (calculated from the Raman spectrum from the binder of composites prepared from large flake graphite) with HTT

Thermal conductivity of samples heat-treated to different temperatures

When the conductivity of the sample prepared from same average flake size and heat-treated to different temperatures is compared, it is found that the conductivity increases as the heat-treatment temperature increases, see fig 4. There is a huge increase in conductivity when the heat-treatment temperature is increased from 1600 °C to graphitization temperature. This can be directly related to the graphitization of the binder. Issi J-P et al (1991) showed that is a relationship between a-direction coherence length and thermal conductivity. Hence, the significant increase in thermal conductivity is as expected because a significant increase in a-direction coherence length is observed when the samples were heat treated from 1600 °C to graphitization temperature.

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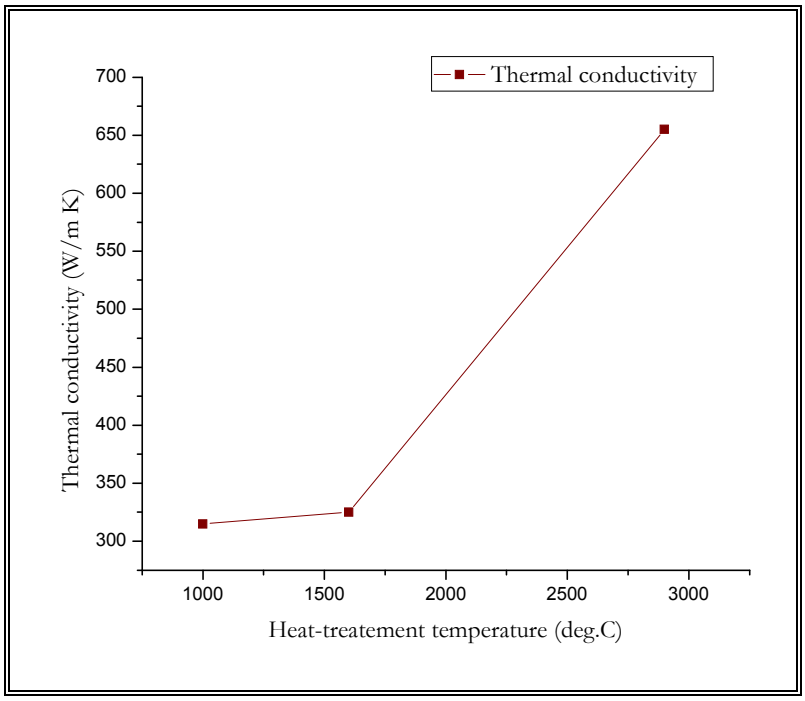


Figure 4 Graph of thermal conductivity versus heat-treatment temperature of samples fabricated using large flakes

DISCUSSION AND CONCLUSIONS

The present march towards miniaturization had set a challenge to cooling industries. Although it was possible to have highly dense circuit boards, the hot spots reduce the life span of the devices due to the limitations of traditional heat sinks. The composites of this study are shown to have high thermal conductivity and hence will find potential application as heat-sinks/heat spreaders in high density electronic devices.

The thermal conductivity of composites of this study was found to be dominated by the flake sizes. For composites of a same flake size, when the heat-treatment temperature was increased, the conductivity was found to increase which was in agreement with the correlation between thermal conductivity and a-direction coherence length shown by Issi P. (1991). A large difference in conductivity was seen following graphitization as compared to different carbonisation stages which was attributed to the graphitization of the binder.

With thermal conductivity of ~655 W/m K, relatively cheap fabrication process compared to carbon fiber based composites, this material is particularly attractive for thermal management problems.

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