THERMAL CONDUCTIVITY OF GRAPHITE FLAKE COMPOSITES

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Abstract

Graphite based composites/products have been looked upon as potential substitutes for conventional heat sinks and heat spreaders. Here we present thermal properties of model composites fabricated using three different flake sizes (referred to as small, medium and large) following carbonization and graphitization and a study of graphitized composites with varying volume fraction of large flakes. Raman study of these model composites is also presented. Thermal conductivity of ~750 W/m K was achieved in the large flake graphitised composites with the highest estimated volume fraction of graphite used. The thermal conductivity was found to be strongly influenced by the flake size, heat-treatment temperature and the volume fraction of graphite. This material is particularly attractive to combat thermal management problems.

Introduction

In recent years, the number of applications requiring more efficient and light weight thermal management, e.g. advanced aircraft, car navigation systems, high density electronic equipments, etc., has considerably increased.

Conventional heat sinks are made from copper or aluminium. Although copper has high thermal conductivity (403 W/m K), its density is high (d_{Cu} = 8.82 g/cm^3) thus making it heavy, whereas aluminium has lower density (d_{Al} = 2.70 g/cm^3) but does not have high thermal conductivity (201 W/m K). Therefore, these industries are focusing attention on alternate materials to replace conventional heat sinks and heat spreaders.

Many materials have been examined for their properties in order to check their applicability in meeting the demands of new technology. Researchers at Oak Ridge National Laboratory (ORNL) were the first to identify the potential of carbon foams for enhancing heat transfer. With further study on pitch based foams, they developed a new foam having thermal conductivity equivalent to that of aluminium (in plane, k = 201 W/m K) with one fifth the weight (Klett J., ORNL).

Graf-tech International Ltd developed natural graphite/epoxy laminate material (eGrafTM) 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, 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).

Hence, graphite a highly anisotropic form of carbon, having low density (d ≈ 1.94 g/cm^3) and good thermal conductivity along two directions is therefore being looked upon as a potential candidate to combat thermal management problems.

Here we will discuss thermal conductivity of some model composites with various graphite flake sizes following carbonization and graphitization and that of composites with different estimated volume fraction of large flake graphitized composites. It will be shown that some composites developed in this work have thermal conductivity 190% of 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 (avg. flake size: 180 µm), medium (avg. flake size: 300 µm) and large (avg. flake size: 600 µm) have been prepared.

Preparation of composite

The composites were made using natural graphite flakes of different average flake sizes (small, medium and large) and mesophase pitch as a binder. The composites were heat-treated to different temperatures namely, 1000 °C, 1600 °C and graphitization temperature i.e. ~2900 °C and all the measurements were carried out at room temperature.

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 study the change in the structure of the binder with an increase in heat-treatment temperature (HTT) and its effect on thermal conductivity of the composites. Coherence length was calculated from the intensity ratios using empirical suggested by Knight D.S. and White W.B. (1989),

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

Where:
C = 44 Å 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)

Results

Composites of different flakes sizes and same starting volume fraction of graphite

The thermal diffusivity of as-prepared samples could not be measured as the pitch was found to soften with the heat of the laser.

The conductivities of medium and large flake composites heat-treated to 1000 °C were found to be 188 W/m K and 290 W/m K respectively. The small flake composites heat-treated to 1000 °C were presumed to have low thermal conductivity as no signal could be detected while measuring thermal diffusivity.

Fig.1 shows the thermal conductivities of the composites of different flake sizes heat-treated to 1600 °C and ~2900 °C. From Fig.1 it can be seen that the thermal conductivity of the composites increases with flake size and HTT. Fig. 2 shows Raman spectra acquired from the binder of large flake as-prepared composites and composites heat-treated to different temperatures. It can be seen that the intensity ratio (I_D/ I_G) i.e. intensity of D-peak (1350 cm^{-1}) to intensity of G-peak (1575 cm^{-1}) decreases with an increase in the HTT. This indicates an increase in the ordering of the basal planes in the binder with an increase in the HTT. Therefore, the coherence length of the binder in the composite increases with HTT, see Fig.3.

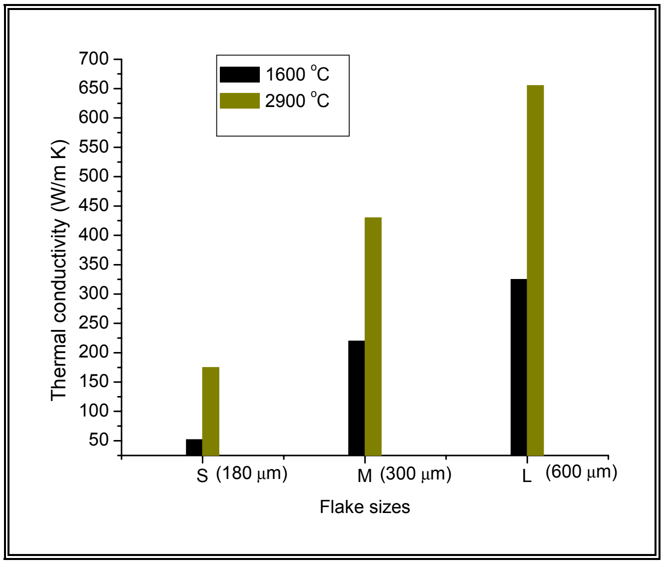

Figure 1. Thermal conductivity of composites of different flake sizes heat-treated to 1600 ºC and 2900 ºC

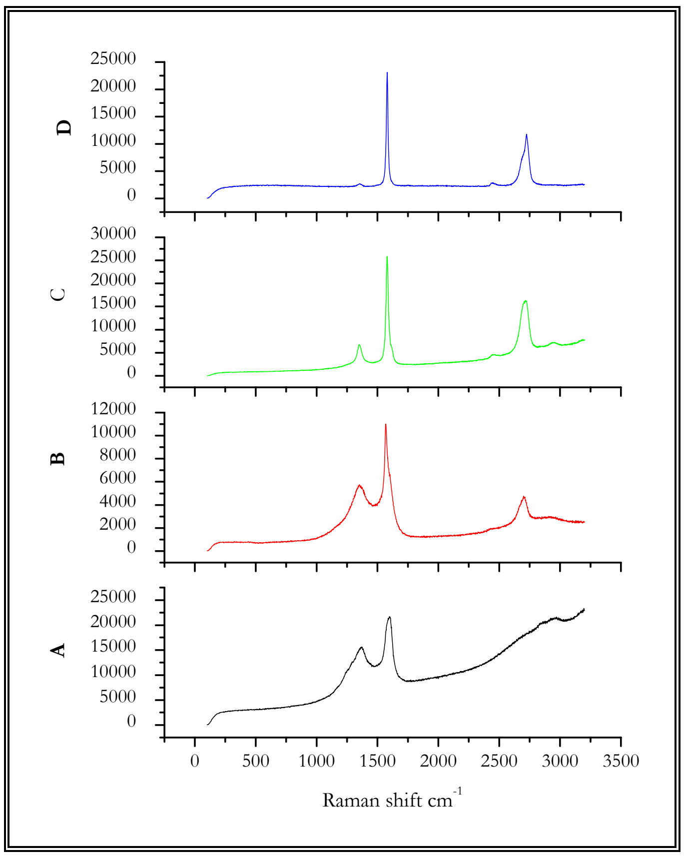

Figure 2. Raman spectra from the binder of samples prepared using large flake graphite at different HTT (A) as prepared, (B) carbonized to 1000 °C, (C) HTT 1600 °C and (D) graphitized (~2900 °C). The micrographs show the area of the sample at which spectra were acquired.

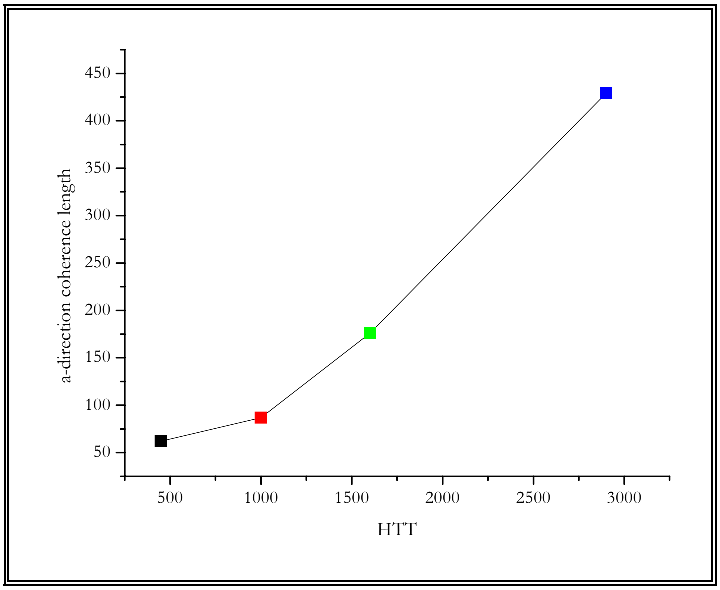

Figure 3. Graph showing increase in a-direction coherence length (calculated from the Raman spectrum from the binder of composites prepared from large flake graphite) with HTT

Composites of same flakes size (large flakes) and different estimated volume fractions of graphite

The effect of volume fraction of graphite on thermal conductivity of the composites was studied using large flake graphitized composites. Fig.4 shows a plot of thermal conductivity of the graphitized large flake composites (A) and a-direction coherence length (B) with an increase in the estimated volume fraction of graphite. It can be seen that the coherence length varies in a similar pattern as the thermal conductivity of the composites.

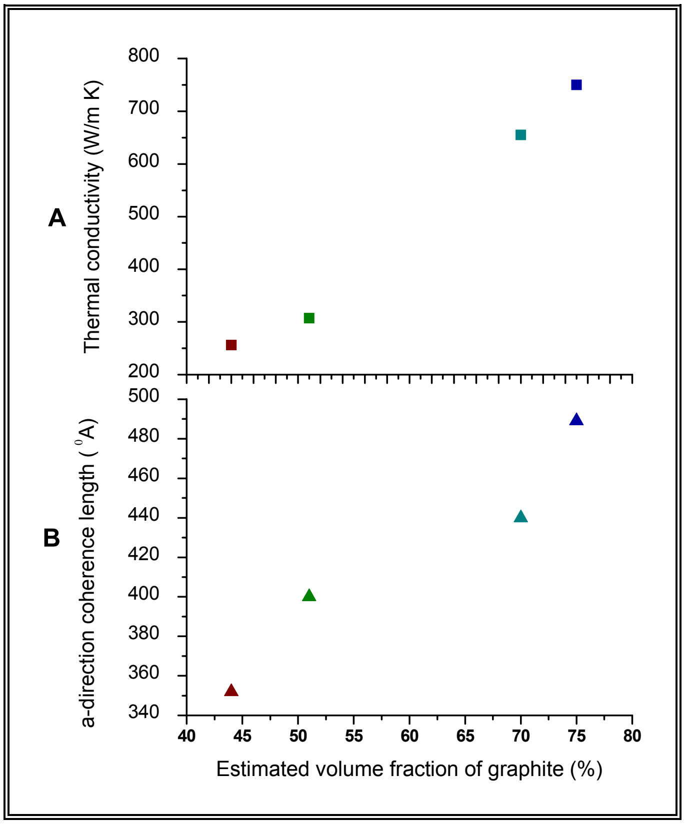

Figure 4. Plot of thermal conductivity of graphitized large flake composites (A) and a-direction coherence length of the binder (B) with varying estimated volume fraction of graphite

Discussion and Conclusions

Thermal conductivity of the composites is found to increase with an increase in the flake size and with an increase in the heat-treatment temperature. As the flake size increases, the number of grain boundaries within a length of a composite decreases. Hence, the phonon loss due to boundary scattering decreases thus giving rise to an increase in thermal transport with an increase in the flake size.

With an increase in the HHT, the pitch binder loses its low molecular species and begins to develop a graphitic structure which can be concluded from the decrease in the D-peak of Raman spectra with an increase in the HTT. It can be seen that there is a huge increase in the coherence length (from 178 Å to 440 Å) when the heat-treatment temperature is increased from 1600 ºC to 2900 ºC which is due to graphitization of the binder (Fig. 3). This increase in the coherence length decreases phonon loss due to an increase in the phonon path length thus giving rise to high thermal conductivity.

When the volume fraction is varied, conductivity as high as 750 W/m K was achieved in graphitized large flake composites with highest estimated volume fraction of graphite. It may be noted (Fig. 4 A), when the estimated volume fraction changes from 44% - 51% to 70% - 75% there is a huge increase in the thermal conductivity. The thermal conductivity of graphitized composites with 44% and 51% estimated volume fraction of graphite is less than that of even carbonized composites (HTT= 1600 ºC) with 75% starting volume fraction of graphite (325 W/m K, see fig. 1). The low conductivity in these composites (composites with 44% and 51% graphite flakes) can be assigned to the high volume fraction of porosity which disrupts the connectivity between the flakes and hence affects the phonon transfer. This is confirmed from the microstructure study carried out using the optical microscopy which showed higher porosity and the pole figure study showed higher disorder in the alignments of the flakes in these composites.

The increase in thermal conductivity from 655 W/m K to 750 W/m K (~100 W/m K or 14.5%) with an increase in estimated volume fraction of graphite from 70% to 75% can be attributed to the higher estimated volume fraction of graphite in the composite (~5% higher volume of flakes) and an increase in the a-direction coherence length of the binder (Fig. 4B) which aids thermal transport. The thermal conductivity values obtained in this study are reproducible and hence, these composites may be looked upon as attractive materials to combat thermal management problem.

Acknowledgment

The authors are thankful to the Morgan Carbon Europe, Swansea for sponsoring this work.

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