MECHANICAL PROPERTIES OF GRAPHITE FLAKE COMPOSITES

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Abstract

Graphite flake composites with high thermal conductivity were studied for their mechanical properties. The model composites were fabricated using three different flake sizes with mesophase pitch as a binder. Here we present flexural strength of as-prepared and carbonized (heat-treated to 1000°C) composites. Flexural strength of graphitised large flake composites is also presented. The flexural strength of small flake composites was found to be dominated by the binder whereas the flakes were found to dominate the flexural strength in composites with large flakes.

Introduction

Graphite exists in small size in nature (typically much less than 0.1 mm in thickness) and contains defects in the form of twinning planes and screw dislocations. Hence not much is known about the mechanical properties of natural graphite flakes (Fakuda M. et al, 1989). However, highly oriented pyrolytic graphite (HOPG) exhibits electronic, thermal and mechanical properties close to those of single-crystal graphite (Kelly B.T., 1981 and Dresselhaus M.E. and Endo M.S., 1988).

Kim D. P. et al (1992) prepared large graphite flakes via the decomposition of aromatic and heteroaromatic hydrocarbons. They showed that the strength of these synthetic flakes rapidly increases from ~0.3 GPa to ~20 GPa when the heat treatment temperature (HTT) is increased from 800 ºC to 2600 ºC which is explained to be due to the flexibility achieved (when annealed at high HTT) in bending an extended graphitic structure.

Extensive study is carried out on mechanical properties of carbon-carbon and carbon fiber-matrix composites. A study on pitch bonded carbon samples showed that the Young's modulus and electrical resistivity of these samples varied in a similar manner as the weight loss in the pitch binder with heat-treatment. The Young's modulus was found to decrease linearly as the HTT increased from ~200 ºC to 430 ºC. After a sharp decrease, at around 530 ºC the modulus was found to passes through a minimum value and increase with further increase in the HTT. This behavior of increase in modulus after initial decrease was assigned by the author to be due to the shrinkage of the binder semi-coke and an increase in the bulk density of the material (Huttinger K.J., 1971).

Here we present flexural strength of model composites fabricated using three different average flake sizes with mesophase pitch as a binder. The flexural strength of small flake composites was found to be dominated by the binder whereas the flakes were found to dominate the flexural strength in composites with large flakes.

Experimental Procedures

Preparation of composite

The composites were made using natural graphite flakes of different average flake sizes; small (avg. flake size: 180 µm), medium (avg. flake size: 300 µm) and large (avg. flake size: 600µm) with mesophase pitch as a binder.

Characterization and measurement

The flexural strength of the sample was determined by using a four-point loading system with test specifications as per ASTM C 651-90. However, the support span in this study was 2.24 times the load span due to the limitation of the sample length. The samples were cut to a precision of 5 micron using Loadpoint cutting machine. From the graph, the maximum load the sample can withstand before fracture was determined and using the formula (Eqn.1), the flexural strength of the sample was calculated.

$$S = \frac{PL}{bd^2}$$ **Equation 1**

Where:
S →flexural strength (MPa)
P → maximum applied load (N)
L→ support span (mm)
B → average width of the specimen (mm)
d → average thickness of specimen (mm)

Results

The flexural strength of the model composites as-prepared and heat-treated to different temperatures is summarized in table 1. The typical curves of load versus extension for as-prepared and carbonized samples of different flake sizes are shown in fig. 1 and fig.2 respectively. Flexural strength of large flake graphitized composites were also measured to check their application feasibility from strength point of view while using them as high thermal conducting (k= 655 W/m K) materials/ heat sinks to combat thermal management problems. Load versus extension curves of large flake composites heat-treated to different temperatures are shown in fig. 3.

Table 1. Summary of flexural strength of the composites

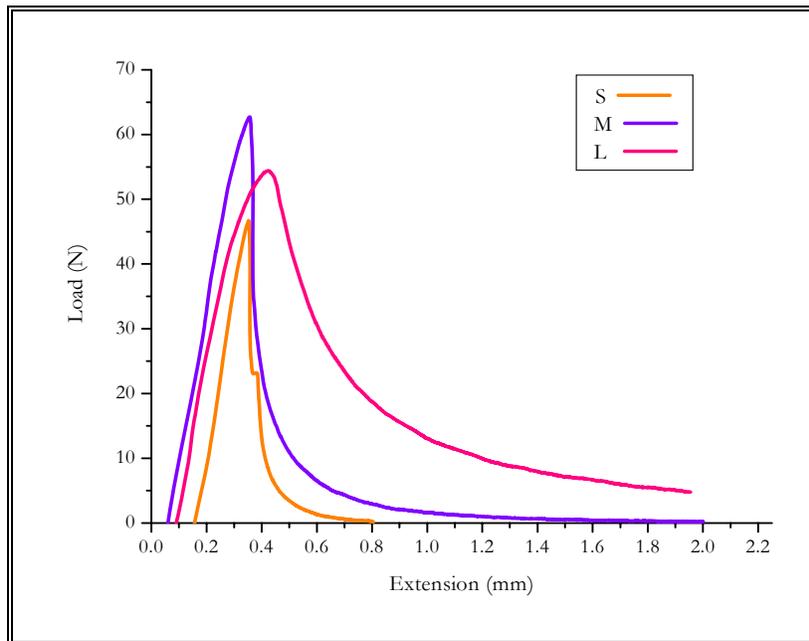| Flake size forming the composite	Average flexural strength of samples (MPa) as prepared and heat treated to different temperatures		
	Prepared at 450 °C	1000 °C	~2900 °C
S (180 μm)	12 ± 4	17 ± 5	-
M (300 μm)	20 ± 2	18 ± 3	-
L (600 μm)	16 ± 3	15 ± 4	9 ± 3

Figure 1. Load versus extension graph of as-prepared composites using different flake sizes
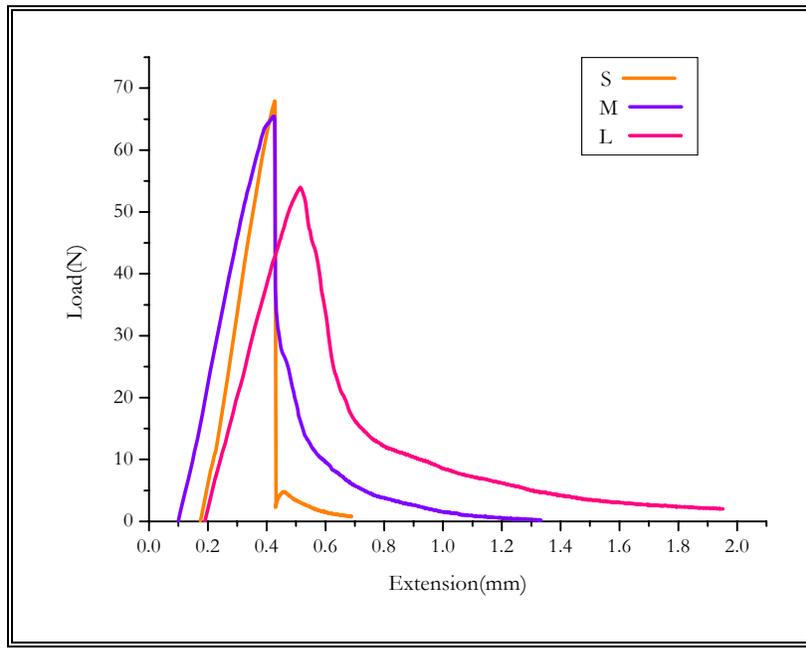
Figure 2. Load versus extension graph of carbonized composites (HTT= 1000 °C) prepared using different flake sizes
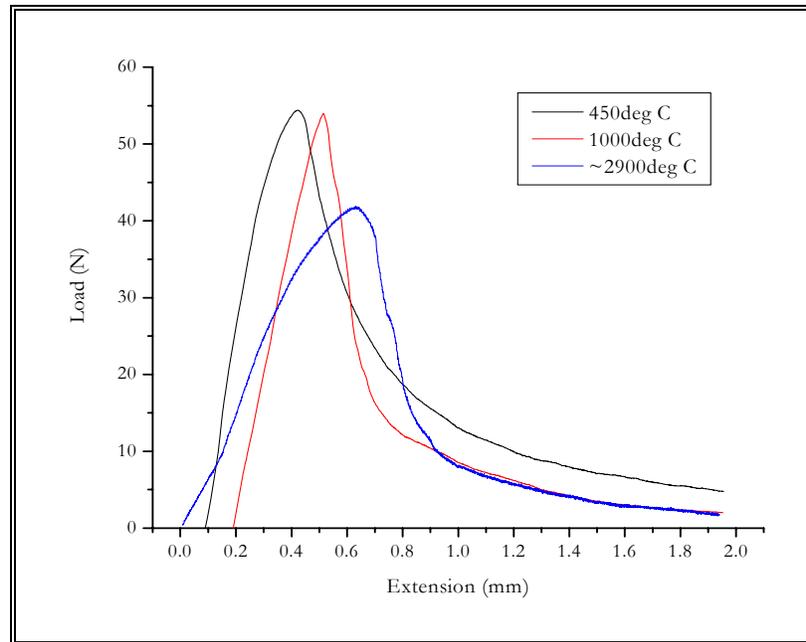

Figure 3. Load versus extension curves of composites prepared using large flake graphite heat-treated to different temperatures.

Discussion and Conclusions

In the above plots (fig. 1 and fig. 2), S, M and L refer to curves corresponding to samples fabricated using small, medium and large flakes respectively. Comparing the curves, it can be seen that the as-prepared composites prepared using small flakes are brittle and fail catastrophically. This behavior is expected as the flakes size is small and hence, when the matrix fails, the composite fails. However, when these composites are carbonized i.e. heat-

treated to 1000 °C, they still fail catastrophically but at a higher load. This indicates that the matrix gets stronger after heat-treatment and hence the composite can withstand a higher load before failure. TGA of the mesophase pitch in an inert atmosphere showed that the major weight loss in the binder takes place between 350 °C to 550 °C and the density of the mesophase phase pitch is found to increases from 1.38 g/cm^3 to 1.8 g/cm^3 following carbonization (Desai S. 07). This is because the low molecular weight species volatilize during carbonization (1000 °C) leading to solid state polymerization involving condensation of aromatic molecules (Lewis I. C., 1997, Huttinger K. J , 1971). This results in stronger C-C bonding which can be assumed to be the reason for the increase in the flexural strength of the composite after carbonization.

From the curves for larger flakes (fig. 1 and fig. 2), it can be seen that there is no significant difference in the maximum load the composite can withstand before and after carbonization. Also, in both cases, the load does not go to zero and the composites do not break after failure for measured extension (2mm). This can be assigned to the large size of the flakes that allow larger length for shear thus giving rise to ductile nature to the composites.

The composites with medium flakes were found to show in-between behavior *i.e.* the curve after the failure starts off falling sharply indicating the composite is brittle but later on the tailing off takes place showing ductile nature. This implies that the composite fails when the matrix fails but after the failure of the matrix, the load is transferred to the flakes which give rise to the ductile nature to the fracture after initial brittle fracture.

If the load versus extension curve of large flake graphitised composite (fig. 3) is compared to those of the as-prepared and carbonized composites prepared using the same flakes size, it can be seen that the maximum load the composite can withstand decreases on graphitization. Also, the curve for the graphitised composite does not show an initial steep rise as in the case of the as-prepared and carbonized samples. This may be due to the huge stress developed during the graphitization process due to the typical thermal expansion behavior of the graphite. Although some of this stress is accommodated by the cracks and porosity obtained during carbonization, the residual stresses within the layers of the flakes obtained during graphitization and cooling of the flakes down to room temperature tend to develop cracks within the flakes. This may be the result of the low strength of the graphitised composite.

Acknowledgment

References

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