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# Mechanical properties and Health monitoring of graphene nanoplatelet/epoxy composites

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## Abstract

Nanocomposites of epoxy with 3 and 5 wt% graphene nanoplatelets (GnPs) were fabricated with GnP sizes of \*5 and \1  $\mu\text{m}$  dispersed within an epoxy resin using a sonication process followed by three-roll milling. The morphology, mechanical, and thermal properties of the composites were investigated. Tensile and flexural properties measurements of these nanocomposites indicated higher modulus and strength with increasing concentration of small GnPs sizes (\1  $\mu\text{m}$ , GnP-C750). The incorporation of larger GnPs sizes (\*5  $\mu\text{m}$ , GnP-5) significantly improved the tensile and flexural modulus but reduced the strength of the resulting composites. At 35 C, the dynamic storage modulus of GnP-5/epoxy composites increased with increasing platelet concentration, and improved by 12 % at 3 wt% and 23 % at 5 wt%. The smaller GnP-C750 increased the storage modulus by 5 % at 3 wt% loading but only 2 % at 5 wt% loading. The glass transition temperatures of the composites increased with increasing platelet concentration regardless of the GnP particle size. A marked improvement in thermal conductivity was measured with the incorporation of the larger GnP size reaching 115 % at 5 wt% loading. The effects of different platelet sizes of the GnP reinforcement on the damage mechanisms of these nanocomposites were studied by scanning electron microscopy.

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## 1. Introduction

Epoxy resin is the most commonly used polymer matrix for advanced composite materials owing to its excellent mechanical properties [1]. There is an increasing demand for advanced materials with improved mechanical or multifunctional properties to meet new requirements for particular applications. Adding fillers to the matrix was found to be an effective and convenient way to achieve composite materials with multifunctional properties such as increased thermal conductivity, electrical conductivity, increased mechanical, and improved barrier properties [2]. In recent years, the availability of nano-scaled particles has offered a new type of filler to produce multifunctional properties. The most commonly used nanoparticles are nanoclay, carbon nanotubes, graphene, and nanocarbon fibers. Graphene discovered in 2004 [3] has been shown by nanoindentation measurements to be one of the stiffest and strongest materials available today with \*1 TPa in young's modulus and \*130 GPa in strength [4]. This 'wonder' material also possesses an outstanding thermal conductivity of around 5000 Wm<sup>-1</sup> K<sup>-1</sup> [5]. Graphene, few layer graphene, or graphene nanoplatelets (GnP) have emerged as one of the most attractive fillers for polymer matrices with an excellent balance between properties and cost.

## 2. Experimental

GnPs were kindly supplied by XG Sciences, Inc. (Lansing MI), and they are produced by thermal expansion of acid intercalated graphite compounds which result in 10–15 layers of graphene stacked into GnP particle with a thickness of \*5 to 10 nm. GnP-5 refers to the average diameter which is

around 5  $\mu\text{m}$  and surface area of 150  $\text{m}^2/\text{g}$ . GnPC750 particles have a similar thickness to GnP-5 and a

diameter smaller than 1  $\mu\text{m}$  with the surface area of 750  $\text{m}^2/\text{g}$ . GnP-5 has a higher aspect ratio than GnP-C750, and both the GnP-5 and GnP-C750 are not surface treated. Epon 828 was purchased from Miller-Stephenson Chemical Company Inc. USA and m-phenylene diamine (m-PDA, flakes C99 %, Aldrich) was used as curing agent.

### 3. Results and discussion

Dispersion of nanofillers is a challenging issue since GnPs have an inherent tendency to form agglomerates due to strong p-p interactions resulting from the large surface area of GnP. In this study, a sonication process was applied to assist in obtaining a uniform dispersion of GnP. Figure 1 shows the SEM images of both GnP-5 and GnPC750 before and after sonication in acetone. Before sonication, GnP-5 particles were stacked on each other and GnP-C750 was aggregated into ball like materials (Fig. 1c); after sonication, it can be seen that the GnP-5 and GnP-C750 were reduced in size.

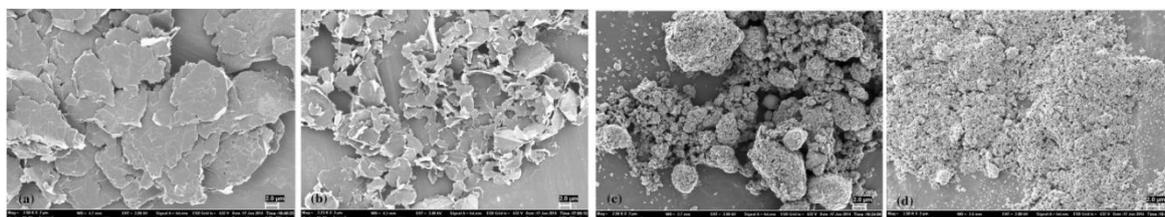


Fig. 1 SEM images of GnP-5 before sonication (a) and after sonication (b); GnP-C750 before sonication (c) and after sonication (d)

A three-roll mill was used to further mix the GnP/epoxy mixture, and the dispersion of GnP-5 and GnP-C750 of the final composites was analyzed with SEM. As is shown in Fig. 2a, b a very uniform dispersion of the GnP-5 could be obtained for both 3 and 5 wt% GnP-5 loading by using the combined sonication and shear mixing process. As for GnP-C750/epoxy composites, the 3 wt% loading appears to be well dispersed (Fig. 2c), but agglomerates (Fig. 2d) can be detected at 5 wt% loading due to the high concentration and surface area of GnP-C750 (750  $\text{m}^2/\text{g}$ ).

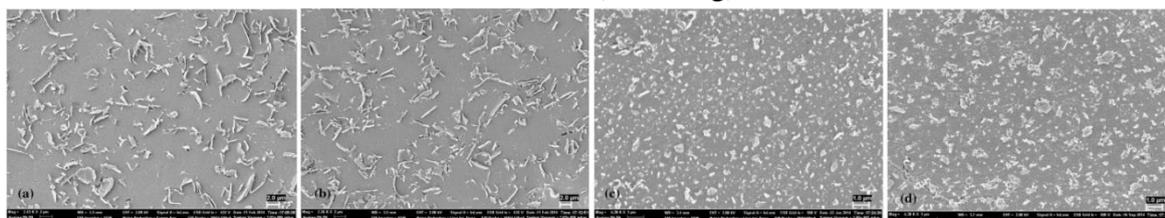


Fig. 2 SEM images of GnP-5 dispersion in epoxy matrix 3 wt% (a) and 5 wt% (b); GnP-C750 dispersion in epoxy matrix 3 wt% (c) and 5 wt% (d)

XPS analysis of GnP-5 and GnP-C750 was performed to determine their chemical structure and the XPS spectra are shown in Fig. 3. XPS can quantify the types of atoms on the surface of the samples and can also identify the types of chemical bonds. The results were collected and are presented in Table 1. It shows that the atomic concentration of oxygen for GnP-C750 was 8.79 % which is more than two times higher than the oxygen concentration (4.01 %) of GnP-5, and the ratio of O:C also confirmed the abundant oxygen element of GnP-C750. The XPS analysis indicates that functional groups containing oxygen atoms (e.g. epoxide, carboxyl and hydroxy groups) on GnP-C750 are higher than on GnP-5, and GnP-C750 could be more compatible with polymer matrix than GnP-5. The higher concentration of oxygen on the smaller GnP-C-750 is due to the larger proportion of edges in that sample because of the smaller particle size. The Raman spectra of the GnP-5 and GnP-C750 are shown in Fig. 4, the D peak ( $\sim 1350 \text{ cm}^{-1}$ ) intensity is associated with disordered  $\text{Sp}^3$ -hybridized carbon present as impurities and defects in the graphene structure of GnP, and the 2D peak ( $\sim 2700 \text{ cm}^{-1}$ ) is more sensitive to the number of graphene layers in a platelet.

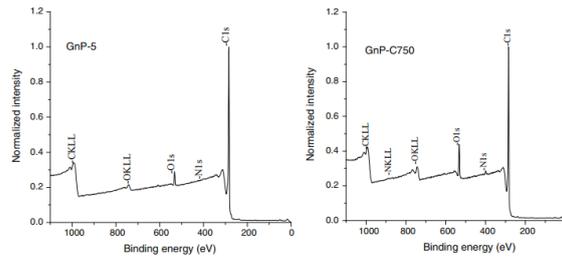


Fig. 3 XPS spectra of GnP-5 and GnP-C750

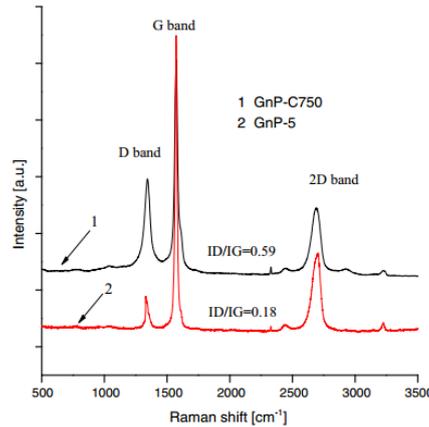


Fig. 4 Raman spectra of GnP-5 and GnP-C750

Figure 5 shows the tensile and flexural properties of GnP-5- and GnP-C750-based epoxy composites. It is well known that the modulus of a composite is dependent on the moduli and volume fraction of the composite constituents. As we can see from Fig. 5, the elastic and flexural moduli increase with increasing GnP loading regardless of the particle size.

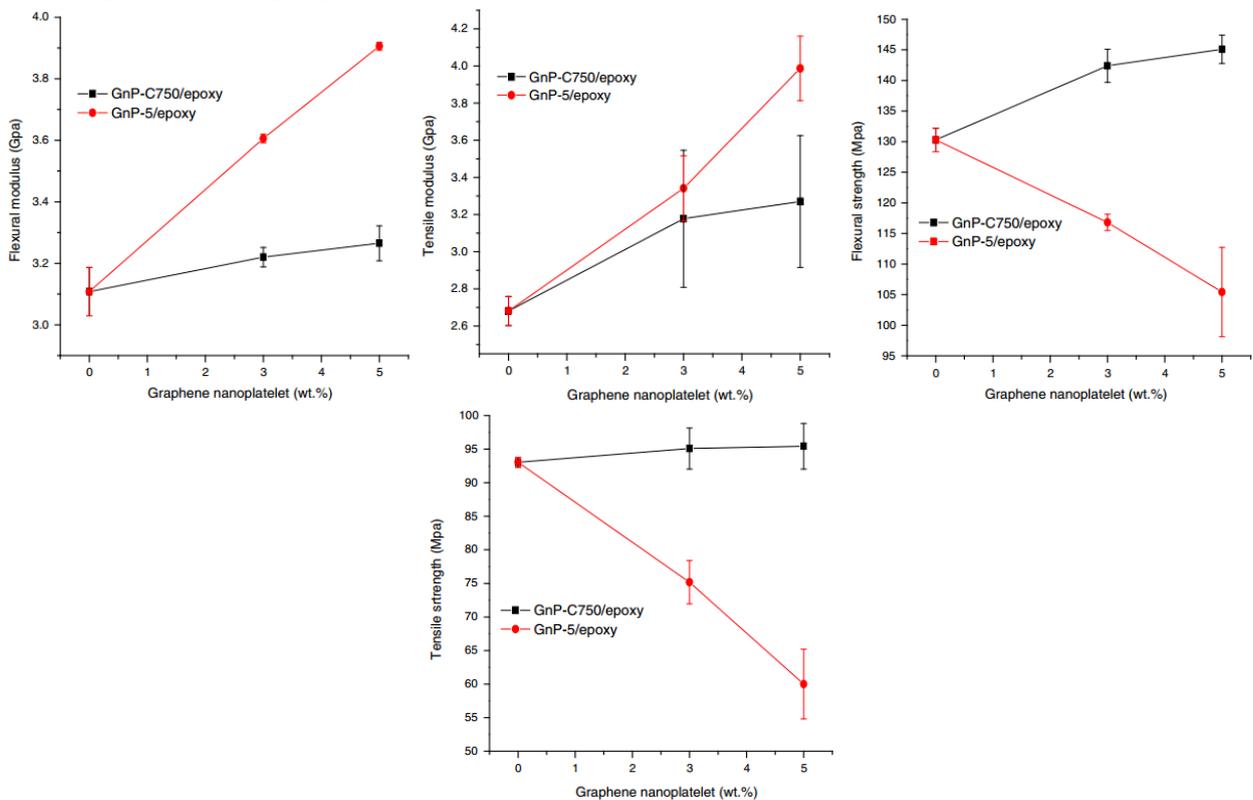


Fig. 5 Tensile and flexural properties of the neat epoxy matrix and the GnP/epoxy nanocomposites. Figure 6 illustrates the variation of storage modulus and tan delta with temperature for pure epoxy and its composites. For clarity, only one representative DMA curve for each case was plotted. It can

be seen that the incorporation of GnP-5 and GnP-C750, both result in an increase of the storage modulus compared to that of the neat epoxy. It is believed that this is caused by the increased stiffness due to the reinforcement of the GnP and confinement of the epoxy chains between the GnP particles. It is clearly shown that the effect of GnP-5 reinforcement was larger than that of GnP-C750. For GnP-5 reinforced epoxy nanocomposites, the composites with 3 and 5 wt% GnP-5 showed about 12 % ( $\pm 1.1$  %) and 23 % ( $\pm 1.5$  %) higher storage modulus than the pure epoxy matrix at 35 C.

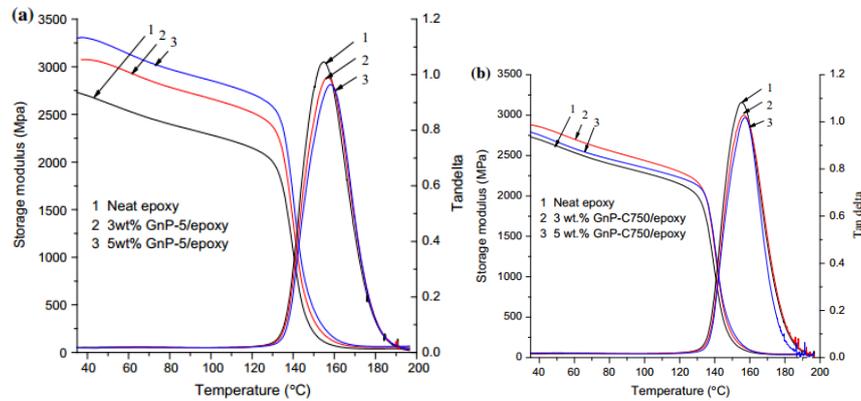


Fig. 6 Dynamic mechanical properties of pure epoxy and its composites

Thermal conductivities of epoxy and GnP/epoxy nanocomposites are shown in Fig. 7. The thermal conductivity of epoxy with 3 wt% GnP-750 and 5 wt% GnP-C750 shows almost the same value and increases only slightly compared with the neat epoxy.

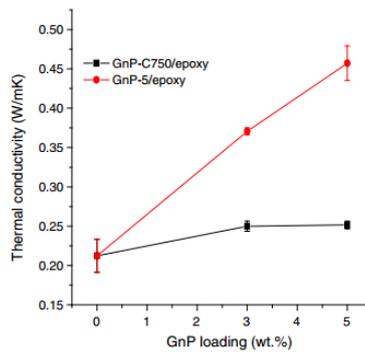


Fig. 7 Thermal conductivities of neat epoxy and its composites

The interfacial interaction between the epoxy resins and GnP can be revealed through examination of the morphologies of fracture surfaces of GnP nanocomposites. Composites with 5 wt% GnP-5 and 5 wt% GnP-C750 are shown in Fig. 8.

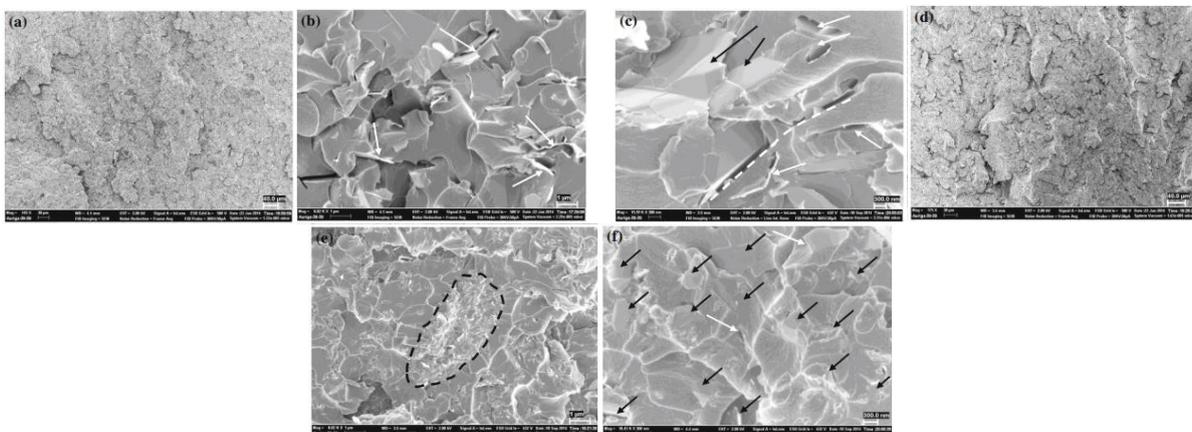


Fig. 8 SEM micrograph of fracture surface of 5 wt% GnP-5/epoxy composite (a), (b), and (c); and 5 wt% GnP-C750/epoxy composite (d), (e), and (f)

#### 4. Conclusion

This study investigated the influence of GnP sizes and dispersion on the mechanical and thermal properties of epoxy nanocomposites. Analysis of the GnP with XPS shows that there are more oxygen functional groups present on the GnP-C750 than on the GnP-5. A uniform distribution of GnP-5 in epoxy matrix was obtained after using the combined tip sonication and high shear mixing process, while agglomerations are detected for GnP-C750-based composites at high filler loading. The mechanical property measurements showed that larger nanoplatelets (GnP-5) exhibited greater reinforcement in improving the modulus of the composites compared to GnP-C750.

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