



## MECHANICAL REINFORCEMENT OF CONDUCTIVE GRAPHITE/ BIOPOLYMER THIN FILM COMPOSITE

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### ABSTRACT

Mechanical properties such as tensile strength, elastic modulus and elongation at break of graphite/ biopolymer composites with different particulate fractions of graphite (5 wt.%, 10 wt.%, 15 wt.%, 20 wt.%, 25 wt.%, 30 wt.% by weight percent in the composites ) were investigated. The composites showed improved elastic modulus and tensile strength with increase treated graphite weight loading by ~300% and ~200% respectively at the percolation threshold, compared with those of its neat counterpart. Meanwhile, the functional group tends to decrease in the composites with increasing filler content in which contributes to the stiffness of the composites as the elongation at break of composites decline. The results implies that the mechanical properties of the composites mainly depend on dispersion condition of the treated graphite filler, crystallite structure and strong interfacial bonding between treated graphite in the biopolymer matrix.

**Keywords:** Conductive Composite • Graphite/ Biopolymer • Resistivity • Mechanical Properties • Thin Film

### INTRODUCTION

The assembly of functional fillers and biopolymer properties meets many possible needs of reducing greenhouse emissions, enhancing waste management, and improving sustainability in term of electrical and electronic applications. In order to improve the mechanical properties of polymeric materials, nano-carbons like carbon nanotubes (CNTs) (Pillai and Sinha Ray, 2011; Alessandro et al., 2011) carbon nanofibers (Senguptaa, 2011), and graphene (Sridhar, 2013; Naebe, 2014) has been reinforced both into thermosetting and thermoplastic polymers.

Graphite, which is naturally abundant, is well known as traditional carbon-based filler and is recognized as the best conductive filler for its excellent conductive properties and well dispersion in polymer matrix (Narimissa, 2012). Graphite reinforced polymer composites have exceptional mechanical behavior which are unequalled by other materials. The material is strong, stiff, and lightweight. Polymeric graphite composite is the material of choice for applications where lightweight and superior performance has raises eye brows. In recent years, Cai et al., (2013) found that with the incorporation of 4.4 wt% of graphite oxide nanoplatelets (GONPs), the Young's modulus and hardness of the polyurethane (PU) are significantly increased by ~900% and ~327%, respectively. Yasmin (2005) state that, an improvement of elastic modulus in expendable/ epoxy nanocomposite over pure epoxy can be attributed to the in situ formation of graphite nanosheets as well as uniform dispersion and exfoliation of graphite nanosheets in the former case.

Therefore, this work evaluate the influence of graphite filler on mechanical properties of graphite/ biopolymer thin film composites. The graphite flakes were treated first using sonication and then grafted into the biopolymer with varying graphite weight loading (wt.%). Then, the composites will undergo I-V test to determine the film conductivity. Meanwhile, the crystallite structure of

graphite/ biopolymer using X-ray diffraction (XRD) will be correlate with the mechanical properties in order to provide better understanding of the dispersion, structural and interfacial bonding between treated graphite into the biopolymer matrix.

### METHODOLOGY

#### Graphite preparation

Flake graphite mixture and ultrasonic solvent were placed into a flask in the ultrasonic cleaning bath at room temperature. After sonication, the treated graphite is washed to neutrality with water, dehydrated, and dried in an oven below 60 °C for 60 min. This method has been adopted from Li in 2007.

#### Biomonomer preparation

Biomonomer is prepared from renewable resources of virgin cooking oil (VCO). VCO is obtained and chemically manipulated at laboratory scale using less than 1L tan of waste cooking oil (Anika Zafiah, 2010). The biomonomer conversion begins with the catalyst preparation to generate the epoxies from the unsaturated fatty compound, and second reaction is the acid-catalyst ring opening of the epoxies to form polyols or bioepoxy (Anika Zafiah, 2009).

#### Composites preparation

Thin films are prepared by mixing the biomonomer with Methylene Diphenyl Diisocyanate (MDI) and acid treated graphite using mechanical stirrer and cast into square container which are then dried at ambient temperature for at least 6 hours. The resulting substrate films were peeled off and identified. Micrometer and optical microscope images are used to measure the thickness of the sample at particular point ranging ~0.1 mm.



### Measurement and Characterization

In this research, XRD Bruker D8 Advance is used. The crystallite structure of the graphite can be obtained. Measurements of current-voltage characteristics of the prepared samples are carried out using Keithley 6517A electrometer as in Figure 1. The resistances  $R$  are determined from the slopes of the current-voltage characteristics. Electrical resistivity  $\rho$  and conductivity  $\sigma$  are calculated from the expression

$$\rho = \sigma^{-1} Rl/g \quad (1)$$

where  $l/g$  is the electrode geometrical factor ( $l$  is the total length of the electrode width and  $g$  is the electrode distance),  $L$  is the film thickness.

Then, the tensile test was conducted according to ASTM D883 (2012).



Figure 1: Two-point probe of I-V characteristic using Keithley 6517A electrometer

## RESULTS AND DISCUSSION

### X-ray study

The XRD analysis in Figure 2 verifies the intense peak at  $2\theta$  value of  $\sim 26.4^\circ$  presence in all composites (B 15wt.% - G 30wt.%) assigned to single graphite layers at a distance of 0.341 nm, in which similar to earlier studies (Murariu, 2010). The diffraction also shows scattered intensity distribution broad such peak ( $2\theta = 17 - 25^\circ$ ) in composites suggesting the semi crystalline structure of biopolymer. The intensity of the peaks differed in neat biopolymer and different ratios of graphite/ biopolymer composites. These pattern suggesting that a sufficient amount of graphite weight loading contributes to the crystallization properties of the thin film composites.

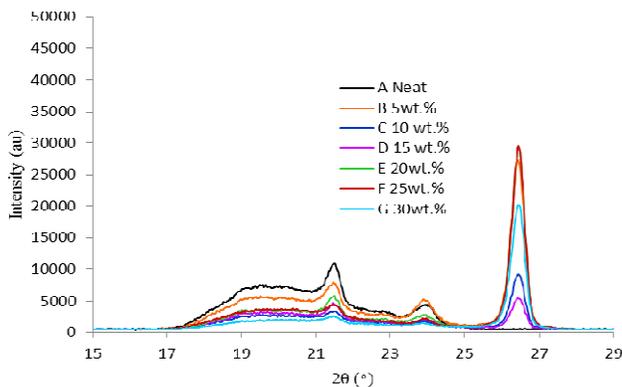


Figure 2: XRD traces of graphite/ biopolymer composites

### Electrical characteristic

Figure 3 represents the electrical conductivity ( $\sigma$ ) values is plotted as a capacity of graphite weight loading (wt. %) in biopolymer composites. This figure shows at the high weight loading of graphite in the polymeric materials concurrently amplifies electrical contacts between the particles and, as a result, the film resistivity decreases (Bachari, 2014). Moreover, it is apparently shown that the calculated electrical conductivities from the reciprocal of the resistivity were increased by numerous orders of magnitude from  $3-14 \times 10^3$  S/m upon 20, 25, and 30 wt.% of graphite/ biopolymer composites. Bare that slight differences in film thickness will imperil the film conductivity efficiency as thicker film will increase the resistivity.

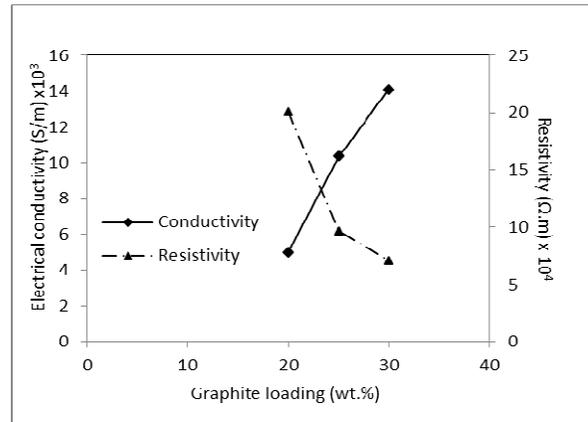


Figure 3: Electrical characteristic of graphite/ biopolymer composites

Low electron mobility in the composites might be the cause for lower graphite loading (5, 10, 15 wt. %) in graphite/ biopolymer composites having a low magnitude (lower than  $10^0$  S/m) of conductivity, where this characteristic cannot be justified by the model. It happens when the low loading of graphite are covered by biopolymer chains where the composite does not form a conductive interconnected network in the insulating bioepoxy matrix to reach the percolation limit. Obviously, the percolation threshold of the composites occurs at 20 wt.% of graphite loading.

At times, it is difficult to validate or invalidate the expected conduction mechanism by direct analytical measurements because of low currents implied. Hence, different measurement techniques should be occupied such as surface conductivity, DC conductivity, thermally stimulated DC current in understanding the relationship between their electrical properties (Bachari, 2014).

### Mechanical behaviour

Certain properties of samples such as tensile strength, elastic modulus and elongation at break are expected to be improved with addition of treated graphite in the biopolymer matrix. Figure 4 until 6 demonstrate the mechanical properties of composites with neat biopolymer thin film as reference, wherein increased of graphite weight loading (5 wt%, 10 wt, 15 wt%, 20 wt%, 25 wt%, 30 wt%) respectively.

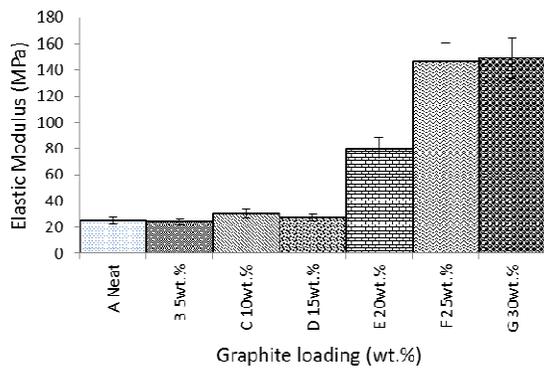


Figure 4: Elastic modulus (MPa) of graphite/ biopolymer composites

It can be clearly seen that the elastic modulus of the graphite/ biopolymer composites in Figure 4 is increased than that of the neat biopolymer, which can be attributed to efficient load transfer between the treated graphite and the biopolymer matrix resulting from the chemical bonding and physical bonding. The study also revealed that as the elastic modulus increases, the tensile strength increases (Figure 5) with increases of graphite weight loading in the biopolymer matrix. Noted that both modulus and strength of the thin film composites increased dramatically by about ~300% and ~200% respectively at the percolation threshold.

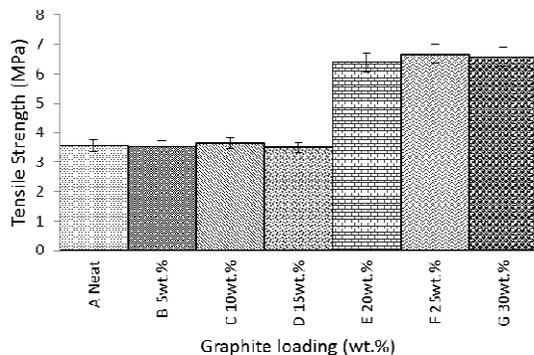


Figure 5: Tensile strength of graphite/ biopolymer composites

Figure 6 indicates that the addition of treated graphite weight loading caused a decrease in elongation at break or break displacement of biopolymer composites. These mechanical behavior shows that the thin film composites having strong interfacial bonding between the treated graphite and matrix interfaces and aggregation of treated graphite in the biopolymer composites that increases the embrittlement and increases of graphite weight loading contribute to the stiffening effect. Hence, results proved that graphite flakes have the synergistic effect on improving mechanical properties of biopolymer. A similar behaviour has also been observed for nanographite platelets (NGP) based polylactide (PLA) (Narimissa, 2012) and expanded graphite reinforced epoxy resin matrix (Yasmin, 2005).

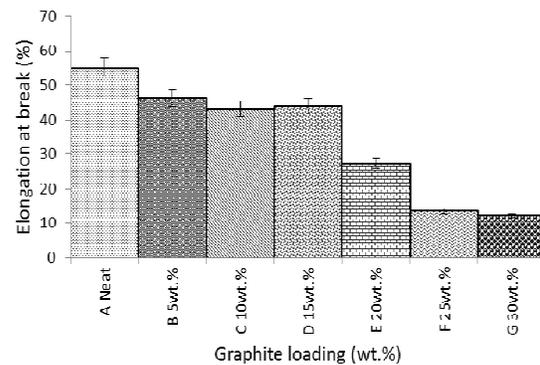


Figure 6: Elongation at break (%) of graphite/ biopolymer composites

## CONCLUSION

Mechanical reinforcement showed significant improvement with increased of treated graphite weight loading over neat biopolymer which attributed to strong interfacial bonding, which allows effective load transfer of the composites. Furthermore, functional properties of the composite greatly depend on the structure of treated graphite in which can contribute to the crystallite structure of the composites. Noted that the elongation at break of composites decreases as the functional group tends to decrease in composites with increasing filler content. Finally, it can be conclude that the development of such multifunctional composites has potential for tailorability of desired mechanical properties for desired conductive thin film applications.

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## REFERENCES

Alessandro, C., Micaela, C., Pravin, J., Mauro, G., Stefano, B. and Alberto, T., (2011). Electrical Properties of CNT-Based Polymeric Matrix Nanocomposites, Carbon Nanotubes–Polymer Nanocomposites, Dr. Siva Yellampalli (Ed.), ISBN: 978-953-307-498-6, InTech.

Anika Zafiah, M. R., (2010). Polymer from renewable materials, *Science Progress*, 93(3): 16-1.

Anika Zafiah, M. R., (2009). Degradation studies of polyurethanes based on vegetables oils, Part I; Photodegradation, *Progress in Reaction Kinetic and Mechanism*, 33: 391-363.

ASTM International Standard Test Method for standard terminology relating to plastics, United states : ASTM D883-2012



Bachari, T. S., (2014). Electric properties of Polyvinyl Acetate (PVA)- Polyol and prepared Sulfonated Phenol formaldehyde Resin (SPF) bulk samples composite, *Asian Journal of Applied Science and Engineering*, Volume 3, No 2 ISSN 2305- 915X.

Cai, D., Yusoh, K. and Song, M., (2009). The mechanical properties and morphology of a graphite oxide nanoplatelet/polyurethane composite, *Nanotechnology* 20 085712 (5pp)

Li, J. H., Li, J. and Li, M., (2007). Preparation of expandable graphite with ultrasound irradiation, *Materials Letters* 61, pp.5070–5073.

Murariu, M., Dechief, A. L. Bonnaud, L., Paint, Y., Gallos, A., Fontaine G., Bourbigot, S., (2010). Polym. Degrad. Stab., pp.95.

Naebe, M., Wang, J., Amini, A., Khayyam, H., Hameed, N. Hua Li, L., Chen, Y. and B. Fox, (2014). Mechanical Property and Structure of Covalent Functionalised Graphene/Epoxy Nanocomposites. *Sci. Rep.* 4, pp.4375.

Narimissa, E., Gupta, R. K., Choi, H. J., Kao, N. and Jollands, M., (2012). Morphological, mechanical, and thermal characterization of biopolymer composites based on polylactide and nanographite platelets, *POLYMER COMPOSITES*.

Pillai, S. K. and Sinha Ray, S., (2011). Epoxy-based Carbon Nanotubes Reinforced Composites, *Advances in Nanocomposites - Synthesis, Characterization and Industrial Applications*, Dr. Boreddy Reddy (Ed.), ISBN: 978-953-307-165-7, InTech.

Senguptaa, R., Bhattacharyaa, M., Bandyopadhyayb, S., Bhowmicka, A. K., (2011). A review on the mechanical and electrical properties of graphite and modified graphite reinforced polymer composites, *Progress in Polymer Science* 36 pp.638–670.

Sridhar, V., Lee, I., Chun, H. H., and Park, H., (2013). Graphene reinforced biodegradable poly(3hydroxybutyrate-co-4 hydroxybutyrate) nano-composites, *eXPRESS Polymer Letters* Vol.7, No.4 pp.320–328.

Yasmin, A., Luo, J. J., and Daniel, I. M., (2006). Processing of expanded graphite reinforced polymer nanocomposites, *Composites Science and Technology* 66 pp.1179–1186.