Graphene Enhanced Polymer Resins for High-Performance Structural Composites

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ABSTRACT

In this work, a low-cost and scalable manufacturing technique was developed to incorporate highly dispersed graphene into epoxy and polyurethane (PU), which are among the most widely used polymeric materials. The study covered the wide spectrum of graphene materials of different structures, both raw products of synthesis as well as a functionalized one, to be used as reinforced fillers of polymer resins. Additionally, a single or few-layer graphene, as produced by a plasma enhanced chemical vapor deposition (PECVD), and graphene nanoplatelets (GNPs), which are composed of a dozen or several dozen graphene layers produced from exfoliation of graphite, are also investigated. Furthermore, the properties of epoxy composites reinforced with hybrid graphene fillers and combinations of graphene materials with other fillers has also been discussed in the paper.

The influences of concentration and composition of hybrid graphene mixtures on tensile and flexural behaviour of epoxy resin were studied. Tensile and flexural properties of epoxy resin were improved significantly and reached maximum at low addition of graphene (i.e., below 0.1 wt%).

In comparison with pure polyurethane, the graphene-based polyurethane composites exhibited higher tensile strength and modulus improvements, even with a very small amount graphene added. The impact of PECVD graphenes with different flake sizes on tensile properties of PU were investigated. Unlike the graphene-enhanced epoxies, both the tensile strength and Young`s modulus of polyurethane improved greater with the addition of large flake-size graphene than with the presence of small flake-size one. Consequently, these graphene-based polymer resins have great potential in the preparation of various high-performance polymer composites for defence and military applications such as marine, automotive, aerospace, protective equipment, and structures by improving strength and ballistic performances, and reducing weight.

Keywords: *graphene, pristine graphene, PECVD graphene, hybrid graphene, polyurethane, epoxy, graphene polymer nanocomposite*

1.0 INTRODUCTION

In recent years, the polymer composite material has been widely used in various applications including automotive, construction, infrastructure, wind turbine blades, sporting goods, and aerospace [1]. The increasing demand of stronger and lighter polymer composite for innovative composite materials has promoted researchers to continuously update existing materials and develop new materials with improved properties and multifunctionalities, so they can be exploited for improving civilian and military applications [2].

The distinctive one-atom-thick planar structure, along with excellent mechanical strength, electronic transport properties, favourable thermal conductivity, and exceptionally high aspect ratio [3], make graphene an excellent structural and multifunctional nano-reinforcement for developing high-performance and

multifunctional polymer composites. Several promising results regarding improvements in the mechanical performance of graphene-based polymer composites such as fracture toughness, fatigue, impact strength, tensile strength, stiffness, and surface hardness etc. have already been reported by various research groups [4]-[12].

The main goal of this work is to improve mechanical performance of common resins, such as epoxy and polyurethane. The chosen resins are widely used as adhesives, coatings, construction materials and automotive materials for defence applications, industrial tooling and other general consumer products [13]-[15]. This is explored by utilizing different types of graphene materials and by tuning the FORZA technology for graphene production [16]. The reactor system, FORZA, is based on a scalable microwave plasma technology platform. The system can transform carbon containing gases and hydrogen to graphene which is grown vertically on a variety of substrates, such as stainless steel, inconel, aluminum, quartz, and copper, as well as more complicated structures, including metal meshes, carbon fiber cloths, and particles etc. The morphology and flake size of the produced graphene can be controlled by changing the growth conditions by varying the growth time, gas compositions, input energy, gas pressures, and temperatures.

The whole family of graphene materials, as well as their graphite or carbon precursors, can be used directly as fillers of polymer resins to provide efficient reinforcement and functional properties of the obtained composites [17]-[21]. Due to the multifunctional behaviour of these fillers, they can improve the chosen mechanical properties of their polymer composites, e.g., tensile strength, flexural strength, elongation, modulus, and/or toughness. This was the reason we have been conducting research concerning the behaviour of various graphene types on mechanical performance of polymer resins. Many critical factors affect the mechanical characteristics of graphene polymer composites, such as the size and aspect ratio of graphene flakes, its concentration within the composites and the graphene manufacture approach (e.g., liquid-phase exfoliation of graphite, chemically oxidation and reduction, PECVD etc).

To understand the contribution of the manufacturing techniques on the reinforced behaviour of the graphene used, a PECVD production of graphene (bottom-up) and graphene nanoplatelets produced by exfoliating graphite (top-down approach) were integrated into epoxy resin. The two as-produced graphene materials often have different morphology and number of layers. The former is single- or few-layer graphene, while graphene nanoplatelets is a multi-layer graphene which is available at low cost but often increase the tensile modulus of composite material [22]. Their mechanical properties were measured and compared.

The weak interfacial adhesion between the graphene layers and polymer matrix, resulting mainly from the atomically smooth surface and chemical inertness of graphene, may make the stress transfer from the matrix to graphene inefficient. Functionalization of graphene surfaces, by introducing suitable functional groups compatible with the polymer not only significantly improves the filler-polymer interfacial interactions but also increases the dispersion of graphene into the polymer [23]-[25], and thus make graphene materials very effective fillers in order to obtain mechanically reinforced composites. Epoxy composites with graphene fillers functionalized with amines were therefore prepared and mechanically tested.

The mechanical performance of polymer composites can be further enhanced by combining graphene fillers which have structure difference. The combination need not necessarily be limited to only graphene fillers, but may be a combination of those from other carbon-based materials (e.g., CNT and C60 etc.), inorganic materials or metallic materials [26]. When the hybrid graphene filler is homogeneously dispersed in the polymer resins at an optimum concentration, their synergetic effect can bring multiple advantages that a single filler may not be capable of offering [27]. In addition to type of fillers added, the ratio of ingredients in the given hybrid set of fillers need to be optimized in order to achieve the desired mechanical properties [28]-[30]. In this paper, two sets of hybrid combinations, which are (G001 + CNT + G002) and (G001 + CNT + G003), at concentrations that varied from 0.01 wt% to 1 wt%, were investigated. The different proportion of each component in these hybrid fillers were also studied. Table 1 presents all the types of graphene and combinations used in this study.

Table 1: Graphene and hybrid graphene used in the study.

Graphene type	Description
G001	PECVD graphene, small flake size < 0.5μm
G002	PECVD graphene, large flake size < 7μm
G003	Amine-functionalized graphene oxide
GNPs	Graphene nanoplatelets, particle size from 2μm to 5μm
Gmix1	33.3% G001 + 33.3% CNT + 33.3%G002 (by weight)
Gmix2	85% G001 + 10% CNT + 5% G002 (by weight)
Gmix3	85% G001 + 10% CNT + 5% G003 (by weight)
Gmix4	75% G001 + 10% CNT + 15% G003 (by weight)

2.0 EXPERIMENTAL

2.1 Materials

G001 and G002 are single- or few-layer graphene produced by a PECVD method and was provided by CealTech AS, Norway. GNPs has particle sizes 2-5 μm and was purchased from Sigma-Aldrich. Graphene oxide (GO) with an oxygen atomic content of 37.6% was purchased from Abalonyx AS. Carbon nanotubes (CNTs) used in this study were (-OH) functionalized multiwalled carbon nanotubes purchased from Nanografi. The CNTs had a stated purity of > 96%, a 1 wt% content of (-OH), an inside diameter of 5-15 nm, an outside diameter of 28-48 nm, and a length of 10-25 μm. (3-aminopropyl) triethoxysilane (APTES) was kindly provided by Shin-Etsu Silicones.

The epoxy resin used for composite preparation was Epikote 828 and the curing agent used was Lindride 36K; both kindly provided by Comrod AS and used as received. Epikote 828 is a medium viscosity liquid epoxy resin produced from bisphenol A and epichlorohydrin, having a viscosity of 12-14 Pa·s at 25°C and epoxy group content of 5260-5420 mmol/kg. Lindride is a low viscosity and anhydride-based curing agent consisting primarily of methyltetrahydrophthalic anhydride and methylhexahydrophthalic anhydride.

The polyurethane (PU) used in this study was a HPE elastomer system, consisting of polyols (HPE 40A Polyol-SL240000 and HPE 85A Polyol-SL285000) and isocyanates (HPE 40-85A ISO-SL000105), and covers a large range of hardnesses, going from a 40 shore A elastomer to a 55 Shore D semi-rigid resin. All the polyol and isocyanate components were manufactured by Synthene and distributed by Lindberg & Lund AS. All chemicals were used without any further purification.

2.2 Functionalization of Graphene Oxide

0.1 g of graphene oxide powder was added into 100 ml of anhydrous ethanol. The solution was sonicated for 1 h before 2 g of APTES was added, and the mixture was stirred vigorously at a temperature of 78 °C for 18 hrs. After the reaction, the mixture was filtered and washed several times with deionized water and ethanol. Finally, the amine-functionalized GO was obtained after drying at 50 °C in vacuum for 12 hrs.

2.3 Preparation of Graphene-Based Resin Samples

2.3.1 Graphene-Based Epoxy Composites

First, graphene was dispersed homogenously into Epikote 828 resin by using ball-miller Retsch PM100 machine at the speed of 250 rpm. The mixture was then well mixed with hardener Lindride 36K at the weight ratio of 100:84 and then heated up to 40°C for 30 min under vacuum to get ideal viscosity and to remove bubbles. Finally, the resin was poured into a stainless-steel mould. Prior to pouring, the releasing agent was applied gently to the mould surface before the mould was preheated at 60°C for an hour to remove the moisture content. The samples were finally poured into the mould and cured at 80°C for 3hrs and then 130°C for another 3hrs.

2.3.2 Graphene-Based Polyurethane (G/PU) Composites

G/PU composites were prepared at 3 different graphene concentrations: 0.01 wt%, 0.05 wt%, and 0.1 wt%. To prepare the composites, a certain amount of graphene was added into 49g of polyols. The mixture was ball-milled at the speed of 250 rpm using a Retsch PM100 machine. Thereafter 100g of isocyanates HPE 40-85A ISO-SL000105 were added into the G/polyols. Prior to moulding, the resulting mixture was degassed in vacuum for 15 mins, and a thin layer of Extra M Release Agent from ChemTrend was applied to the mould, followed by drying at 70°C to remove moisture completely. Finally, the G/PU was casted in the mould. The composites were fully cured after 7 days at room temperature.

2.4 Characterization and Mechanical Testing

The graphene types used in this study were characterized by means of Raman spectroscopy, scanning electron microscopy (SEM), and transmission electron microscopy (TEM). Raman spectra were collected on a Renishaw inVia micro-Raman spectrometer with a 532 nm Ar laser excitation; the graphene powders were placed on microscope slides and a small glass cover placed on top, which were secured by adhesive tapes. TEM was conducted on a JEOL JEM-2100 microscope with 200 kV accelerating voltage; graphene was dispersed in IPA and the dispersed flakes were deposited on a TEM support grid. SEM was performed on a JEOL JSM-7500F microscope with 5 kV accelerating voltage in which graphene powders were placed on carbon tapes.

Mechanical properties of nanocomposite materials were measured by an Instron Applications Laboratory Model 5985. Tensile strength tests were performed according to the standard DIN EN ISO 527-2. Dog-bone shape samples were used for the testing. Samples have a gauge length of 50 mm, 10 mm width, and thickness of 4 mm. Tensile tests were performed with a constant cross head speed of 1 mm/min for epoxy and 100 mm/min for PU material. Flexural strength (3-points flexural) test were performed according to standard ASTM D790-03; specimen sizes for this study were: 80 x 10 x 4 mm. A minimum of seven specimens were tested for each formulation as required to reduce the uncertainty of the measurement.

3.0 RESULTS AND DISCUSSION

3.1 Graphene Characteristics

The characteristic quality of graphene was investigated by using Raman spectroscopy. The typical Raman spectra of graphene has D, G and 2D peaks (Figure 1). The D peak appears at $1320 - 1350$ cm^{-1} and is the defect band that represent a ring breathing mode from the sp^2 hybridized carbon rings. The G band appears around 1570-1585 cm^{-1} and represent an in in-plane vibrational mode involving the sp^2 hybridized carbon atoms. The 2D band peak appears around 2650-2700 cm^{-1} and can be used to investigate whether the layers of graphene are single, double or multi-layers. In some cases, also D' band arises, from a defect-induced double- resonance process [31].
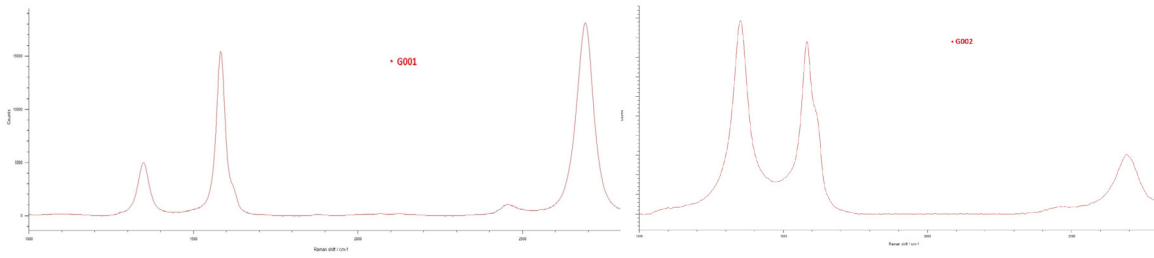
Figure 1: Raman spectra of G001 (left) and G002 (right).

Table 2: Raman characteristics of graphene G001 and G002.

Graphene type	D	G	2D	I_D/I_G	I_{2D}/I_G
G001	1345.0	1580.0	2690.3	0.38	1.16
G002	1348.6	1579.6	2691.2	1.71	0.59

The 2D band of G001 peak has a high intensity and a symmetric shape and a symmetric D peak which gives indication of a single layer graphene, which is further confirmed with the I_D/I_G ratio of 0.38 and the ratio I_{2D}/I_G of 1.16 as shown in Table 2. The higher ratio of I_D/I_G in G002 compared to that in G001 probably arises from the fact that G002 is more hydrogenated and has more interaction between the individual flakes, due to the production parameters in the PECVD process, possibly partly resulting in a change in hybridization, for example from sp^2 to sp^3, or a change in the sp^2 configuration. The strong D band peak and D' band peak also may suggest a more nanocrystalline structure and presence of graphene edges and defects such as distortion, vacancies, and straining to graphitic lattices, which are prevalent features of PECVD graphene; the high defect density in PECVD graphene is referred to energetic particles from the plasma interacting with the growing surface [32]-[35]. This is further seen by a lower I_{2D}/I_G in these samples. However, the electron diffraction pattern of G002, see Figure 5, shows characteristics of a highly crystalline material, which suggests it is indeed single layered graphene.

Figure 2 and Figure 3 represents a typical surface morphology of the large vertical graphene sheets as grown by PECVD [16] from a mixture of CH_4 and H_2 These SEM images show obvious differences in size and morphology between G001 and G002. These graphenes were produced using different process parameters (e.g., gas ratios, plasma densities, temperatures, pressures etc.). G001 exhibits a small flake size and densely packed structure. While G002 has a maze structure with a larger flake size and relatively wide spacings. The cross-section view in Figure 3 (right) represents the grown height of approx. 7 µm for G002.

TEM images of G001 Graphene at low magnification showed wrinkled graphene sheet structures with a flake sizes around 500nm (Figure 4). The electron diffraction (not shown) pattern indicates single or few-layer graphene. The electron diffraction pattern of G002 in Figure 4 confirms the hexagonal-packed lattice structure of single-layer graphene. The TEM images of GNPs on the contrary suggest a presence of multiple graphene layers with relatively smooth surfaces (Figure 5).
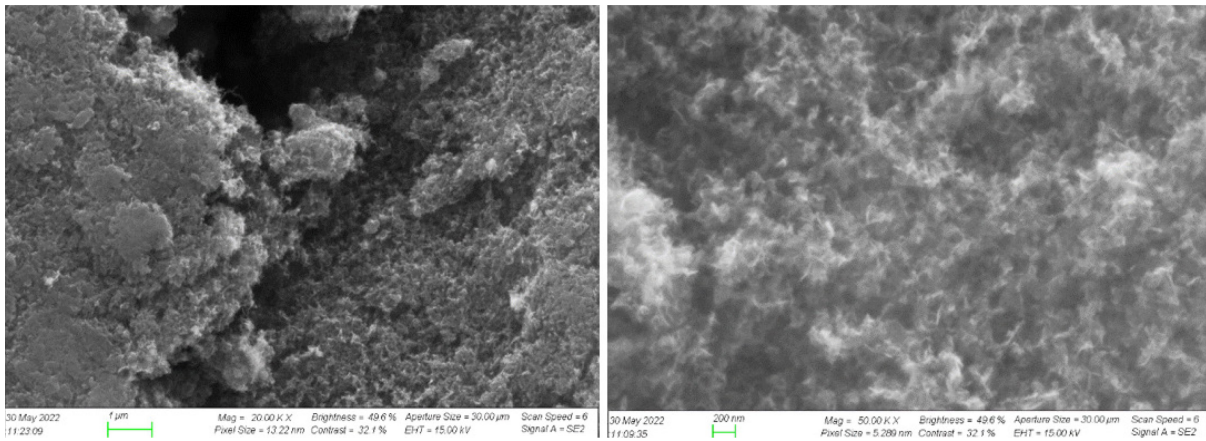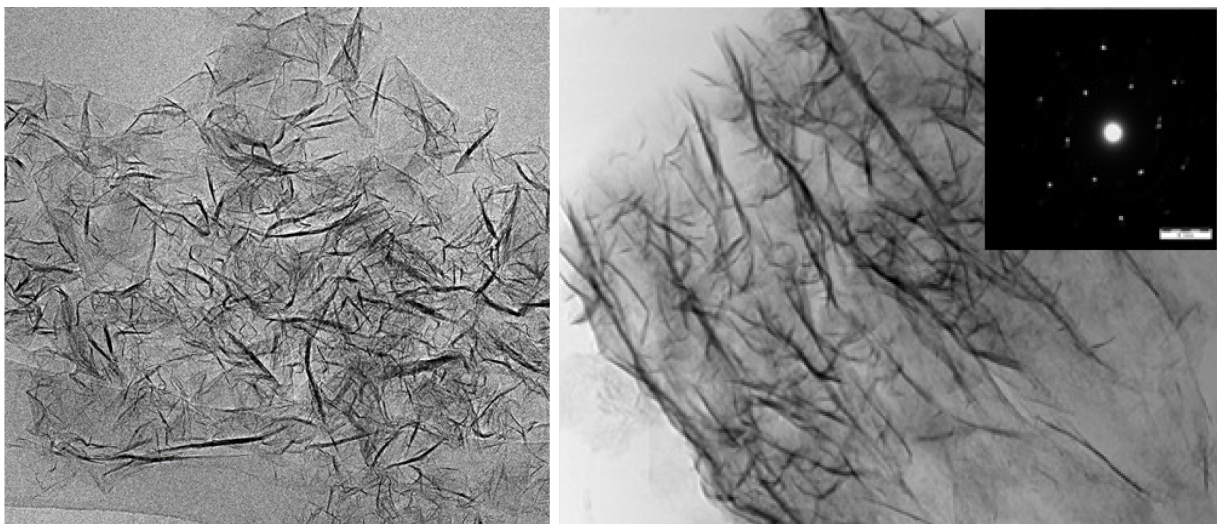
Figure 2: The SEM images of graphene G001.

Figure 3: The SEM images of graphene G002: top-view of the grown vertical graphene sheet (left) and cross section-view of graphene sheets (right).
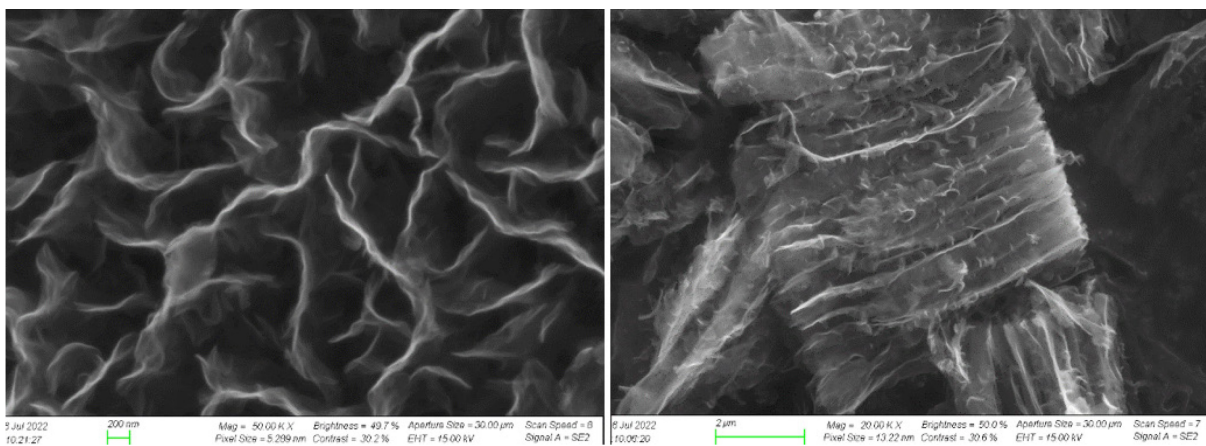

Figure 4: TEM images of G001 graphene (left) and G002 graphene (right). Inserted SAED pattern showing high crystallinity from single layer graphene.
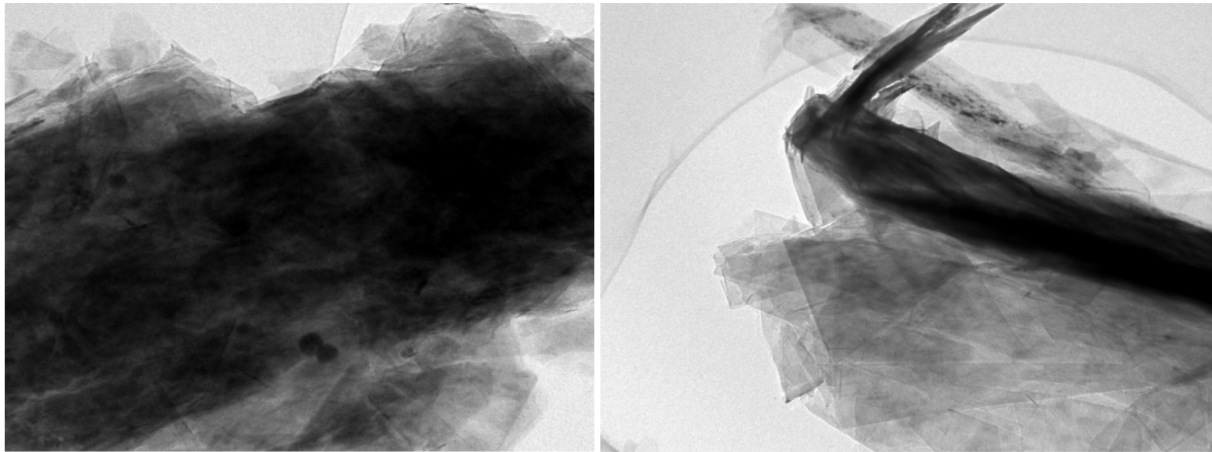
Figure 5: TEM images of GNPs.

3.2   Mechanical Properties of Graphene Epoxy Nanocomposites

3.2.1   Impact of Graphene Type on Mechanical Properties of Epoxy

3.2.1.1   Tensile Properties

Five (5) graphene and hybrid graphene types were selected and incorporated into epoxy Epikote 828 at concentration of 0.3 wt%. For each sample, 7 specimens were tested in tensile strain until rupture of the specimen. Average standard tensile properties at break for all samples is illustrated in Figure 7.

All the graphene-containing epoxy resin samples developed in the project showed significantly increased both tensile strength and elongation at graphene concentrations of 0.3 wt%. The small-size PECVD graphene G001 showed the highest improvement with an increase of up to 88% in tensile strength and 182% in elongation compared to the pure epoxy resin sample, followed in performance by the larger-sized PECVD graphene G002. The enhancement is explainable with the large aspect ratio and high interfacial contact area of the graphene. Moreover, an increase in tensile strength and elongation of epoxy resins containing graphene resulted due to the surface roughness of graphene (wrinkled surface), which caused an enhanced mechanical interlocking with the epoxy chains and, consequently, better adhesion [36].
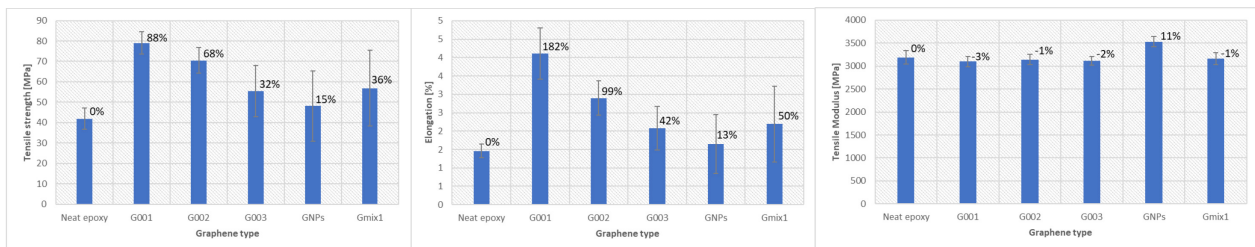

Figure 6: Tensile properties of neat epoxy and graphene-based epoxy resin at graphene concentration of 0.3 wt%.

Among the graphene samples, multi-layer GNPs showed the lowest improvement in tensile strength (15%) and elongation (13%). However, as expected, GNPs has the highest tensile modulus with an increasing of 11% compared to neat epoxy (not shown). Other graphene composites showed insignificant or negative improvement in modulus. It is interesting that when adding a nanofiller it results in improving strength and elongation, but give minor impact to the modulus. This probably refers to the improved toughness of resins

by these graphene fillers. The epoxy resin material fails due to the creation of cracks in the matrix, and further crack length increases followed by failure of samples. It is well known that the tougher materials have an efficient mechanism of internal energy absorption, which restricts the crack propagation, thus allows it to maintain the desired level of durability in the real field of application under massive variations in operating conditions.

The tensile test results showed that the efficiency of filler induced increase in epoxy strength and elongation follows the trend: G001 > G002 > Gmix1 > G003 > GNPs.

3.2.1.2   *Flexural Properties*

Flexural properties of the 6 samples, including a pure epoxy resin and 5 graphene-containing epoxy resin samples at concentration of 0.3 wt%, were measured by using 3 points-flexural test. For each sample, 7 specimens were tested until rupture.

All 5 graphene-enhanced epoxy samples showed an increase in the average flexural strength and extension, as compared to the pure reference sample. A maximum increase of 74% in strength and 125% in extension was obtained by the addition of hybrid graphene Gmix1 whilst multi-layer GNPs showed the smallest increase in flexural strength (9%) and extension (11%). However, GNPs exhibited the highest improvement in flexural modulus with an increase of 13% (not shown).
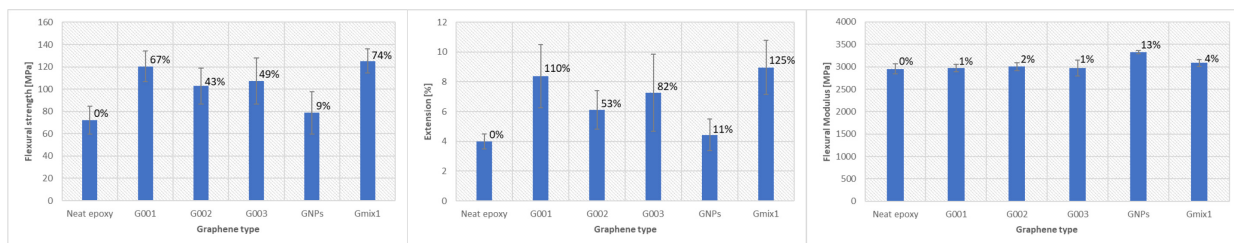

Figure 7: Flexural properties of neat epoxy and graphene-based epoxy resin at graphene concentration of 0.3 wt%.

In regard to the flexural strength and the flexural extension, the improving order was as follows: Gmix1 > G001 > G003 > G002 > GNPs.

3.2.2   Impact of Graphene Concentration on Mechanical Properties of Epoxy

Gmix2/epoxy and Gmix3/epoxy samples at various concentrations of 0.01 wt%; 0.1 wt%; 0.3 wt%; and 1 wt% were made. To understand the impact of ingredient proportion Gmix1/epoxy and Gmix4/epoxy were prepared at a graphene concentration of 0.3 wt%. The mechanical properties of these samples were then measured and compared.

3.2.2.1   *Tensile Properties*

The measurements for tensile properties of Gmix2- and Gmix3- based epoxy resins versus graphene concentration are shown in Figure 8. In general, elongation properties exhibited a similar trend as shown by tensile strength and both improved significantly with addition of graphene even with a very small amount (0.01 wt%). The optimal concentration of graphene falls into a low value range (0.01 – 0.1 wt%).

Neat epoxy resin demonstrated the tensile strength value of 42±5 MPa and an elongation value of 1.46±0.17%, which increased to 66±5 MPa and 2.57±0.32% respectively with the addition of 0.01 wt% Gmix3. It further reached a maximum at respectively 75±5 MPa and 3.14±0.57% at Gmix3 concentration of

0.1 wt%. A further increase in the Gmix3 concentration to 0.3 wt% and 1 wt% decreased the tensile strength to 70±4 MPa and 62±2 MPa, and the value of elongation to 2.74±0.25% and 2.31±0.08%, respectively. Apparently at above 0.1% weight fraction, the effectiveness of graphene diminishes, possibly due to the degradation in dispersion quality of graphene in the epoxy matrix. This highlights one of the disadvantages of graphene in that it is challenging to disperse graphene properly at the higher weight fractions.

Both the tensile strength and the elongation of resin containing 0.01 wt% Gmix2 exhibited the highest improvement with values of 77±2 MPa and 3.62±0.47%; respectively. Increasing the concentration of Gmix2 from 0.01 to 0.3% was accompanied by significantly decrease of both the tensile strength and elongation.

In comparison, at low concentration (i.e., 0.01 wt%), the Gmix2-based resin showed higher tensile strength than Gmix3-based resin; when increasing concentration to 0.1 wt% and higher, the Gmix3 is superior to Gmix2. The difference between the two hybrid graphenes is that the pristine graphene G002 was used in Gmix2 and the functionalized graphene oxide G003 was included in Gmix3. This indicates the functionalized graphene oxide G003 plays an important role in dispersing and stabilizing graphene in epoxy, especially at high graphene concentrations. In conclusion, it is recommended to use pristine graphene G002 in hybrid graphene set if a small quantity of graphene is used; However, when graphene concentration increases to a critical point where highly dispersed graphene becomes challenging, the presence of functionalized graphene oxide G003 will be favourable.
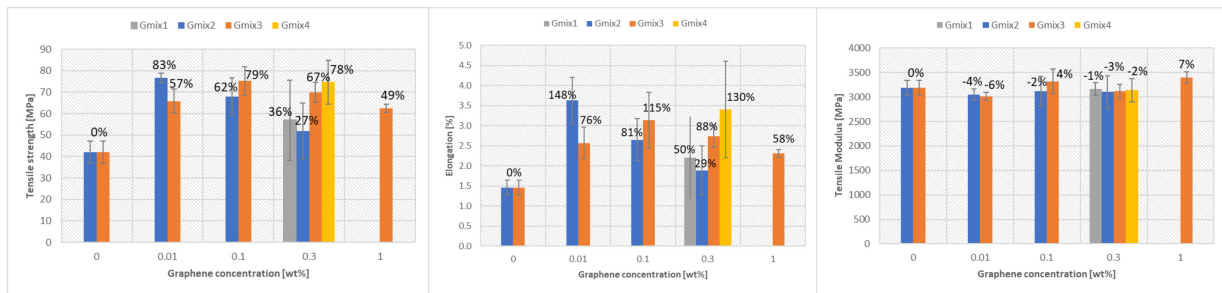

Figure 8: Tensile properties of hybrid graphene-based epoxy resin as function of graphene concentration.

At a concentration of 0.3 wt%, the improvement of tensile strength and elongation follows the order: Gmix4 > Gmix3 > Gmix1 > Gmix2, or in other word: (75%G001 + 10%CNT + 15% G003) > (85%G001 + 10%CNT + 5% G003) > (33.33%G001 + 33.33%CNT + 33.33% G003) > (85%G001 + 10%CNT + 5% G002). As mentioned above, it is explainable that the hybrid Gmix3/epoxy exhibits greater tensile properties than the hybrid Gmix2/epoxy at relatively high concentration of 0.3 wt%, Moreover, when the content of the functionalized graphene oxide G003 in the hybrid set increases from 5% (i.e., in Gmix3) to 15% (i.e., in Gmix4), the tensile properties of graphene-based epoxy increases. This is once again confirming the role of G003 for dispersing and stabilizing graphene in epoxy.

3.2.2.2   *Flexural Properties*

Typical average flexural properties of Gmix2- and Gmix3-based epoxy resins are shown in Figure 9. The flexural properties of resin increased by the presence of both graphene Gmix2 and Gmix3 and exhibited a similar trend as shown for tensile properties. A significant improvement in flexural strength was found for a very small loading of 0.01% by weight of Gmix2 and a decrease was observed with further loading. The flexural strength and extension were a maximum at loadings of 0.1% by weight of Gmix3.
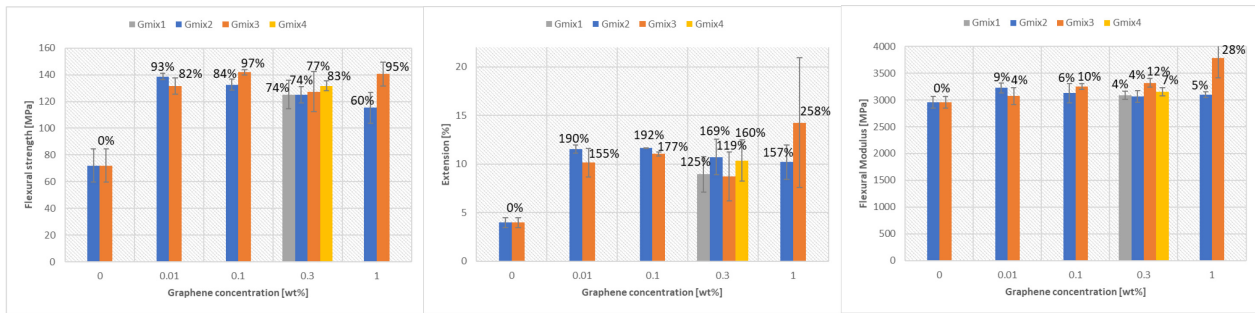
Figure 9: Flexural properties of Gmix2- and Gmix3- based epoxy resin.

The flexural strength and the flexural extension of the neat epoxy were found to be 72 MPa and 4 mm; respectively. With the addition of 0.01, 0.1, 0.3, and 1 wt% Gmix3, the flexural strength of resin increased by 82%, 97%, 77%, and 95% and the extension increased by 155%, 177%, 119%, and 258%; respectively.

Unlike tensile modulus, the flexural modulus of the resin improved with addition of graphene at any concentration. In compared with the neat epoxy, the flexural modulus of Gmix3/epoxy resin improved by 4%, 10%, 12% and 28% and that of Gmix2 epoxy resin increased by 9%, 6%; 4%, and 5% with the presence of 0.01%, 0.1%, 0.3%, and 1 wt%; respectively.

The behaviours of Gmix2 and Gmix3 on flexural strength at a given loading were similar to those on tensile strength. The Gmix2 is favourable at low loading while Gmix3 performs better at higher loading. At concentration of 0.3 wt%, the improvement of tensile strength follows the order: Gmix4 > Gmix3 > Gmix1 = Gmix2. Thus, this suggests that at high graphene concentration, the functionalized graphene oxide G003 should be added more in the hybrid graphene set.

3.3   Mechanical Properties of Graphene/PU Nanocomposite

Figure 10 presents the tensile properties of the G001/PU and the G002/PU nanocomposites as a function of graphene concentration. The figure indicates that the incorporation of both G001 and G002 led to an increase in tensile strength and Young`s modulus of the composites as compared to the neat PU.
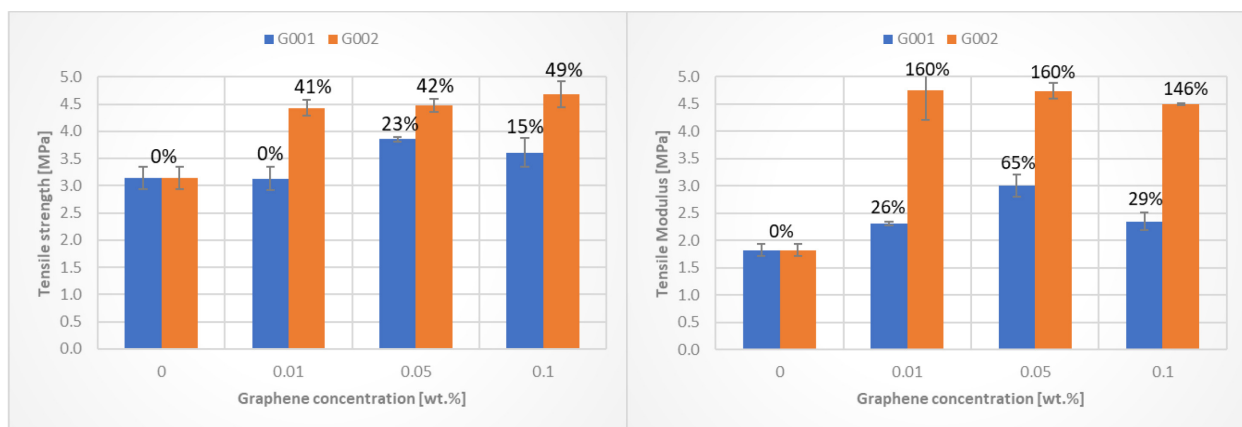

Figure 10: Tensile strengths and Young`s modulus of graphene-based PU nanocomposites.

The G002-based PU nanocomposites showed significant enhanced tensile strength at a very low graphene loading level of 0.01 wt%, and further increase in loading resulted in negligible further increase in the tensile strength. It was found that the elongation at break decreases with the addition of G001 and G002, while the tensile strength and Young's modulus were significantly improved and achieved a maximum at loadings of

around 0.01–0.1% by weight of G001. The data indicates that for the G002/PU composite, the maximal of Young's modulus could possibly be even below 0.01 wt%. The behaviour is typical of many graphene/polymer systems, of which there are a number of reports in the literature [36]. Both the Young's modulus and the tensile strength of the polymer are typically found to increase with the loading of graphene while the elongation at break is decreased.

It is clearly seen that PU nanocomposites prepared with the large flake size graphene G002 present higher tensile strengths and Young`s modulus (Figure 10) for all the nanofiller contents than the nanocomposites prepared with the small flake size graphene G001. These findings are consistent with those of Coleman et al. [37] and Gong et al. [38]. They also found that for a given loading of graphene, the level of reinforcement effect decreased as the graphene flake size was reduced.

4.0   CONCLUSION

The study shows that the magnitude of improvement of properties of composites with graphene depend on the structure of the graphene fillers, i.e., dimensions and number of graphene layers. The impacts of graphene flake size on mechanical properties of epoxy resin and PU resin were different. In contrast with graphene-base epoxy nanocomposites which showed the smaller flake size graphene having greater improvements for mechanical properties, the large flake size graphene-based PU nanocomposite exhibited much higher tensile properties. Introduction of hybrids and various combinations of graphene materials with other fillers of different structures and properties into the epoxy matrix can provide the composites with untypical properties and may also result in new functions based on synergistic effects. The properties of the hybrid graphene/epoxy composites were strongly associated with the functionalized surface of graphene toward improved dispersibility, compatibility and adhesion between epoxy phase and nanofillers, especially at high graphene loading.

The graphene content required to significantly improve mechanical properties of polymer resins has been as small as <0.01 wt%, which is orders of magnitude lower than other nanofillers such as nano-clays, CNT, nano-TiO2, metal/metal oxide nanoparticles, and silica nanoparticles. This such low weight fraction requirement of graphene suggests that graphene shows great promise as nanofiller for the next generations of advanced composite materials for both civilian and military applications. Because of these excellent enhancements in properties, graphene reinforced polymer nanocomposites possess great potential to be used in construction, automotive, electronics, aerospace and for other sectors. However, there are a number of fundamental scientific issues that need to be addressed in order to realize the full impact of this enabling enhanced materials technology. For example, more characterization efforts on graphene/polymer nanocomposites are still necessary to understand the issues related the correlation between the structure of polymer and graphene fillers, their dispersion and interaction. Other relevant mechanical properties such as fracture and fatigue properties, impact strength, damage tolerance, damping response, high strain rate response, torsional/flexural response, and so on are needed to complete the picture. It is also important to determine the impact of graphene on thermal and electrical properties of polymer composites. Finally, it could be at most valuable to study the apparent enhanced fire resistance property of graphene/polymer composites, since this property is one of the main obstacles for polymeric materials to replace metal or inorganic components.

5.0   ACKNOWLEDGEMENTS

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