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# Investigation of electrical-mechanical performance of epoxy-based nanocomposites filled with hybrid electrically conductive fillers

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Keywords: graphene nanoplatelets, carbon nanotubes, in-plane and through-plane electrical conducitivity, flexural strength and modulus, morphology

#### Abstract

This study aimed at investigating the influence of utilizing electrically conductive nanofillers such as graphene nanoplatelets (GNP) and carbon nanotubes (CNT) as secondary fillers on the electrical-mechanical behavior of epoxy/synthetic graphite (epoxy/SG) composites where SG was a microfiller. The composites were prepared by an internal mixer and compression molding and the electrical-mechanical behavior of the composites was characterized by the measurements of the in-plane and through-plane electrical conductivity as well as the flexural strength and modulus. The microstructure of the composites was examined by scanning electron microscopy (SEM). The results showed that the addition of GNP to epoxy/SG composites in the expense of SG had a negative impact on the electrical conductivity as the GNP content increased up to 10 wt%. Composites containing M-grade GNP with a particle size of 15  $\mu$ m showed higher in-plane and through-plane conductivity values compared to that having the C-grade GNP with particle size less than 2  $\mu$ m. The incorporation of CNT to the epoxy/SG/GNP composites had a pronounced improvement to the electrical and flexural properties. The maximum values of in-plane and through-plane conductivity reported were almost equal of about 41 S cm<sup>-1</sup> for composites containing a GNP:CNT weight ratio of 4:6. The effect of fillers morphologies and distribution on the behavior of the composites was also investigated.

## 1. Introduction

The conductive polymer composites (CPCs) which compose of a polymer matrix and conductive filler material (s) have gained a wide interest in many engineering applications such as bipolar plates for polymer electrolyte membrane fuel cells (PEMFC) [1-6]. The properties of CPCs depend mainly on the inherent properties of the individual constituents of the composites in addition to their processing techniques [7-10]. Graphene nanoplatelets (GNP) and carbon nanotubes (CNT) are widely used as conductive fillers besides the traditional filler which is graphite (G) because they have desirable properties such as: excellent thermal and electrical properties, and low gas permeability [11-14]. A combination of micro- and nano-sized conductive fillers in CPCs has been investigated by researchers who reported a positive impact on the properties of the resulting composites [15, 16]. However, a problem of the dispersion and distribution of fillers throughout the polymer matrix was usually encountered by the researchers which affecting the final properties of the obtained composites [17]. The conductive fillers had the tendency to agglomerate and they were difficult to distribute throughout the matrix, so that the desired properties were difficult to be achieved. Kakati et al [18] added graphene in small amounts of about 1 wt% to produce a composite material using phenol formaldehyde (resole) resin as a matrix and the results showed that a composite material with improved electrical conductivity and mechanical properties as well as the I-V performance was produced for the applications of bipolar plate in polymer electrolyte membrane (PEM) fuel cells. Liu et al [19] examined the mechanical properties of graphene nanoplatlets (GNP) reinforced alumina ceramic composites prepared by spark plasma sinetring and they found



Figure 1. SEM images of different types of electrically conductive fillers.

that GNP was successfully dispersed throughout the ceramic alumina matrix which contribute to the improvement in their flexural strength and fracture toughness. Dweiri *et al* [20, 21] also utilized GNP and CNT in corporation with synthetic graphite (SG) to produce polypropylene (PP) nanocomposites by compression molding technique and they noticed a minor improvement in electrical conductivity by using GNP as a secondary filler. Al Islam *et al* [22] investigated the morphology and mechanical properties of graphene/poly (vinyl alcohol) and found that the properties of CPCs could be enhanced by adding small amounts of GNP.

With the different types of composites which can be produced by different techniques composing different types of electrically conductive fillers and polymer matrices, the challenge is still existing to further understanding the relationship between electrical-mechanical-morphological behavior of polymer-based composites filled specially with the combination of micro- and nano-sized fillers. The combination of 3-d filler (SG), 2-d filler (GNP) and 1-d filler CNT) was also a point of interest. In this study, different grades of GNP (M-grade and C-grade) and Multi-wall CNT particles were used as secondary nanofillers in contact with primary synthetic graphite (SG) microfiller filled epoxy resin. GNP was nanoparticles consisting of short stacks of graphene sheets having a platelet shape. The in-plane and through-plane electrical conductivity, flexural strength and modulus, morphology, distribution and dispersion of fillers in epoxy resin were investigated.

#### 2. Experimental

#### 2.1. Materials

Three types of electrically conductive materials with different particle sizes and shapes were used as fillers in this study. Synthetic graphite (SG) supplied by Asbury Carbons Inc., from USA with flake shapes was used as a primary filling material. Exfoliated graphene nanoplatelets (GNP) supplied by XG Science, Inc., from USA and multi-wall carbon nanotubes (CNT) supplied by Nanocyl SA, from Belgium was utilized as secondary nanofillers. The morphologies of the fillers were examined by scanning electron microscopy (SEM) and shown in figure 1. A 635 thin epoxy resin was used as a matrix with a viscosity of 6 poise and the hardener supplied from US Composites. More details about the properties of the conductive fillers are provided in table 1.

#### 2.2. Processing of composites

Different types of composites were prepared: epoxy/SG/GNP and epoxy/SG/GNP/CNT. The fabrication process starts by ball milling the conductive fillers at a rotating speed of 200 rpm for 1 h to homogensize the

Table 1. Characteristics of the conductive filler	s.
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Conductive filler	Particle Size	Surface area $(m^2/gr)$	Density (gr/m <sup>3</sup> )	Purity (%)	
GNP(M-grade)	$15 \ \mu m$	120-150	2.2	>99.5	
GNP(C-grade)	$<2 \ \mu m$	500	2.2	>99.5	
CNT	9.5 nm	250-300	2.1	>90	
SG	74 $\mu \mathrm{m}$	1.5	1.8	99.7	

#### Table 2. Formulations of the produced composites.

	Composition (wt%)			
Sample	Epoxy	CNT	GNP	SG
Epoxy/SG/GNP	30		10	60
	30	_	7	63
	30	_	4	66
	30	_	1	69
Epoxy/SG/GNP/CNT	30	3	7	60
	30	6	4	60
	30	9	1	60

constituents. On the other hand, the epoxy and the hardener were mixed at a rotational speed of 1200 rpm for 40 s using a mechanical mixer (RW20-KIKA-WERK). The matrix and conductive fillers were then mixed at different weight ratios for 10 min at 30 °C using an internal mixer (Haake Reomix) at a rotational speed of 25 rpm. The mixtures were thereafter molded in a square steel mold at a pressure of 30 MPa and a temperature of 150 °C for 90 min. The compositions of the different types of composites are shown in table 2.

#### 2.3. Characterization of composites

In-plane electrical conductivity of the composites was measured using a Jandel Multi Height Four-point probe combined with RM3 Test Unit which had a constant current source and a digital voltmeter designed especially for the four point probe measurement. The through-plane electrical conductivity was measured by a through-plane electrical conductivity tester manufactured at ZBT in Duisburg, Germany. Flexural properties of the composites were examined using universal testing machine Model Instron 5567 according to ASTM D790-03 standard. Scanning electron microscopy (FESEM, Model Supra 55/55VP) was used to observe the surface of the fractured samples and the distribution and dispersion of the conductive fillers throughout the epoxy resin.

### 3. Results and discussion

#### 3.1. Electrical conductivity measurements

The influence of the addition of two different grades of GNP (M-grade and C-grade) in the expense of SG particles on the in-plane and through-plane electrical conductivity of epoxy/SG composites is shown in figures 2(a) and (b). In a previous work by Suherman et al [7], the values of in-plane and through-plane electrical conductivity of epoxy/70 wt%SG composites without the addition of GNP were investigated and reported as 28 S cm<sup>-1</sup> and 23 S cm<sup>-1</sup> respectively. In general, a decrease in both types of electrical conductivities was observed in figure 2 by incorporating GNP to epoxy/SG composites which further decreased as the content of GNP increased in the epoxy by 1, 4, 7, and 10 wt% except for some composites containing low content of M-grade GNP. Composites of epoxy/SG/GNP showed higher values of in-plane and through-plane electrical conductivity when M-grade of GNP was used compared to that of their counterparts which containing C-grade of GNP. The highest value of the in-plane conductivity was 31 S cm<sup>-1</sup> for composites containing 4 wt% GNP and the highest value of the through-plane conductivity was about 42 and 35 S cm<sup>-1</sup> for composites containing 1 and 4 wt% M-grade GNP respectively. A comparison between the electrical and mechanical properties of graphite-polymer composites and graphene-polymer composites has been conducted by many researchers in previous studies. Onyu et al [23] reported an enhancement in both electrical conductivity and mechanical performance of PP/graphene nanocomposites compared to that of PP/graphite composites. Bastiurea et al [24] evaluated the three-point bending properties of graphene-polyester and graphite-polyester composites and better results were observed in case of graphene-polyester composites. However, most of the previous studies investigated on single-filler composites (i.e. either graphite-polymer or graphene-polymer composites) and very few investigated on hybrid-fillers polymer composites combining specifically both graphene and graphite. Levy





*et al* [25] studied graphene-graphite hybrid epoxy composites to improve their rheological properties for thermal applications and they haven't investigated their electrical-mechanical behavior. In our experiments the existence of minor quantities of GNP up to 10 wt% and major quantities of SG up to 70 wt% in fillers-hybrid epoxy composites didn't reflect the expected improvement in the electrical and mechanical properties by the addition of GNP. No significant improvement in the electrical conductivities or flexural strength by the addition of GNP in the expense of SG was also reported by Dweiri *et al* [21] for PP/SG/GNP composites. Hence the behavior of hybrid composites by integration of SG and GNP has to be explained by microstructural observations. For example, Levy and his colleagues observed that GNP in the epoxy composite was mostly edge-on, where graphite was thicker and rather bulky [25].

A clear improvement in the electrical properties was observed by incorporating CNT to the epoxy/SG/GNP composites as shown in figures 3(a) and (b). In a previous work by Suherman *et al* [7], the values of in-plane and through-plane conductivity of epoxy/SG/CNT composites increased noticeably with the replacement of SG by CNT up to 5 wt%. Dweiri *et al* also found a significant improvement in the electrical conductivities by the addition of CNT to PP/SG/GNP composites up to 7.5 wt% [20]. In this study, the addition of CNT at 3, 6, and 9 wt% to composites containing 30 wt%epoxy/SG/GNP with an overall weight percentage of GNP and CNT equal to 10 wt% has a clear improvement in the electrical properties compared to that of 30 wt%epoxy/SG/ 10 wt%GNP which was considered as a control sample. The composites containing GNP:CNT weight ratio of 4:6 represented a good compromise between the in-plane and through-plane conductivity values which were almost maximum an equal of about 41 S cm<sup>-1</sup> for M-Grade composites. The effect of CNT was more pronounced in case of the composites containing C-grade GNP compared to that containing M-grade. For example, in composites containing GNP:CNT weight ratio of 4:6, an increment of more than 15% and 100% in the through-plane conductivity for M-grade and C-grade composites respectively compared to that of the control sample.





Shtien *et al* [26] studied the particle size and synergy effect of GNP-filled epoxy matrix hybrid composites and found that the composites needed to create additional contact between graphene platelets to increase their packing factor. Herein, the additional contacts between SG bulky particles couldn't be enhanced by adding further GNP and the filler-filler or the filler-matrix contact resistances couldn't be minimized. They also mentioned another critical parameter that primarily affects the formation of conductive paths which was the primary-to-secondary filler ratio. The effect of this parameter might been clearly noticed in our work where, for example, the value of the in-plane conductivity shifted to its highest value in composites containing GNP:CNT weight ratio of 4:6 before it dropped again. Dweiri [27] in his study of the effect of graphite shape and size on the electrical conductivity found that the small graphite particles (sphere-like) introduced more point-to-point contacts which decreased the electrical conductivity, while the flake-like particles resulted in higher conductivity values due to the introduction of more surface-to-surface contacts. The larger size of M-grade GNP which displayed higher values of conductivity in this study might result in more surface-to-surface contacts.

#### 3.2. Flexural properties of the composites

The flexural properties of the composites were investigated and shown in figures 4 and 5. The values of flexural strength and flexural modulus of epoxy/70 wt%SG were investigated previously and found to be 33 MPa and 6.5 GPa respectively. In general, the replacement of SG by GNP decreased the flexural strength and the flexural modulus of epoxy/SG composites. An increase in flexural modulus was observed mainly at 4 wt% GNP. There are some factors which usually controlling the mechanical properties of filled-polymer composites: the interfacial bonding between the filler and the matrix which, when it is good, helps to transmit stress from the matrix to the filler and improves the flexural strength [28]. Moreover, the flexural properties in hybrid-filled polymers depend on the size, shape, and dispersion of the filler particles: e.g. larger filler particles increase more the stiffness of the material [29]. This was not the case which seen for epoxy/SG/GNP composites since the stiffness for composites with small size of C-grade GNP was higher than that having larger M-grade size. Heo



**Figure 6.** SEM images of fracture surfaces of epoxy/SG/GNP composites containing (a) 4 wt% GNP (M-grade), (b) 4 wt% GNP (C-grade), (c) 10 wt% GNP (M-grade), and (d) 10 wt% GNP (C-grade).

*et al* [30] investigated the effect of particle shape and size on the mechanical and electrical behavior of graphite filled phenol resin. On the one hand, they reported that the smaller the particle size for sphere-like particles, the larger the specific area, the better the interfacial coherence between particles and resin which enhanced flexural strength. On the other hand, they found that the flake-like particles have higher flexural strength due to their higher specific area and their ability to form a layered structure. Researchers in the literature [31] found that if the conductive filler of a flake-like shape were layered perpendicularly to the compression direction and formed a layered microstructure; its resistance to the flexural loading would be higher than that of sphere-like shape. But the agglomeration of fillers was clearly observed in our composites and it was negatively affecting the mechanical properties mainly for smaller size GNP of C-grade composites. However, further study of the effect of size and shape from one side and the distribution and dispersion of fillers from the other side could explain the unexpected and complex mechanical and electrical behavior of hybrid systems. The addition of CNT to the epoxy/SG/GNP composites had a pronounced improvement on their flexural strength mainly at a GNP:CNT weight ratio of 7:3 mainly for composites containing C-grade GNP compared to that of epoxy/SG/10 wt%GNP composites.

#### 3.3. Morphological observations

A flake-like morphology of nanoparticles was clearly observed for M-grade GNP and aggregates of sub-micron platelets for C-grade GNP are shown in figure 1. The SEM images of SG showed an acicular, macro-morphology and CNT appeared in bundles and entanglements. SEM fracture surface images of the epoxy/SG/GNP and epoxy/SG/GNP/CNT composites are shown in figures 6 and 7 respectively. Figures 6(a) and (b) showed the fracture surface images of epoxy/SG/GNP composites containing 4 wt% of M-grade and C-grade GNP respectively. The GNP was well embedded in the epoxy and agglomeration of GNP was noticed in the microstructure (red circles). The embedment of GNP in the matrix as well as the presence of small GNP agglomerates in composites was also observed by Wang *et al* [32]. The presence and distribution of GNP in the composites had three possibilities: lying between SG plates which could improve the through-plane



Figure 7. SEM images of fracture surface of epoxy/SG/GNP/CNT composites containing 4 wt% of CNT and (a,b) 6 wt% M-grade GNP, (c,d) 6 wt% C-grade GNP.

conductivity; at the edges of SG plates which could improve the in-plane conductivity; and the presence of isolated GNPs in separate entities which had minor contribution in improving either in-plane or through-plane conductivity. On the other hand, there were two factors affecting negatively the values of the electrical conductivity: the agglomeration of the GNP and the isolation of conductive particles with epoxy which can be exacerbated by using fillers with high surface area such as C-grade GNP. Both of these factors were observed in the composites and the agglomeration of GNP was clearly noticed by increasing the GNP content as shown in figures 6(c) and (d) at 10 wt%. Scattered sub-microns pores were found on the epoxy phase and its presence was clearly verified at higher magnification (figures 7(b) and (d)). The existence of porosity decreases the electrical conductivity. Other researchers [3, 4, 7] confirmed that the dispersion of conductive filler and the existence of voids controlled the values of the electrical conductivity and the flexural strength. On the other hand, the existence of pores in CPCs resulted in the stress concentration regions which deteriorated its mechanical properties [31].

The smooth and clean fracture surface of composites and the existence of clear gaps or cavities (white arrows in figures 6(a), (b) between fillers and matrix indicating poor interfacial adhesion between fillers and matrix which leaded to a catastrophic failure and hence degradation of the mechanical properties. This was also observed by Wang *et al* [32] for Polyethersulfone/Epoxy/GNP composites which also affected negatively the electrical properties of the composites. Moreover, the random located particles resulted in the formation of less stacking structures and the existence of more intra-particle pores which means less level of densification leaded to no further enhancement in the mechanical and electrical properties. The direct inter-particle contacts between GNP might be interrupted in the hybrid system, therefore, the in-plane conductivity more decreased. Oh *et al* [31] have noticed similar trend in their study of expanded graphite/flake-type graphite Filled phenol composites.

The SEM images of the fracture surface of epoxy/SG/GNP/CNT composites are shown in figure 7. CNT at a content of 6 wt% in figure 7(a) were well distributed throughout the matrix, nevertheless there were still several agglomerates. The presence and dispersion of CNT in the composites could be similar to that represented by the

three dimensional finite element (FE) model which was suggested by Sarasvand *et al* [33] in their investigation on the effects of GNP and CNT on the mechanical properties of epoxy-based nanocomposites.

# 4. Conclusion

The potential of using a hybrid system of nanometer- and micrometer-sized electrically conductive fillers in the preparation of epoxy-based nanocomposites had been investigated and some conclusions were drawn. The addition of GNP as a secondary filler to epoxy/SG composites was not effective, mainly at high level of loading, in enhancing either their electrical or mechanical properties. In general, the decrease in the in-plane and through-plane electrical conductivity by the replacement of SG with GNP could be explained by the agglomeration of GNP and the embedment, to some extent, of GNP agglomerates in the epoxy. There are other manufacturing constraints such as the presence of voids in the structure and the difficulty to obtain a layered structure. On the other hand, the increase in the in-plane and through-plane conductivity, if existed, could be explained by the improvement in the particle-to-particle contacts in the in-plane and through-plane directions respectively which might be attributed to the presence of nanofillers i.e. GNP and at the edges and between SG flaky plates. CNT had a significant enhancement in the properties which was common in the literature as a bridging filler between GNP. The values of electrical conductivity and flexural strength were still below the target of US Department of Energy (DOE) for bipolar plate of PEMFC of 100 S cm $^{-1}$  and 59 MPa respectively. The fracture surfaces of composite samples showed a brittle-like behavior. Finally, a study of three-dimensional model of the hybrid system is recommended in the future to further understand their electrical-mechanical behavior.

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