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Effects of Multiwalled Carbon Nanotubes and Graphene Nanoplatelets Filled Hybrid Epoxy Nanocomposites on Electrical and Mechanical Properties

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Abstract

The material properties can be significantly changed, with the addition of low concentration of nanofillers. With the integration of two nanofillers into the epoxy matrix, new hybrid nanocomposites can be obtained to suit requirements of the high voltage applications by improving desired electrical, thermal and mechanical properties. Indeed, hybrid nanocomposites are the fastest growing areas of material research. In the present work, the feasibility of multi-scale hybrid nanocomposite for conductor core applications was discussed. Composite sheets were fabricated with a very low wt.% of Multi-Walled Carbon Nano Tube(MWCNT) with an average size of 6-13nm and Graphene Nanoplatelets (GNP) with a particle size of 15μm were mixed in to commercially available bisphenol-A epoxy resin reinforced with E-Glassfiber. A homogeneous epoxy-nanofiller mixture is obtained using mechanical stirring followed by slow pultrusion technique. The resulting MWCNT/GNP reinforced epoxy resin was used to fabricate hybrid nanocomposite sheets with 70 wt.% unidirectional Glass Fiber(GF) with density of 400 g/m2. The hybrid nanocomposite sections were processed using pultrusion technique. With this former process glass fiber-epoxy (70:30), GF-epoxy/MWCNT (70:28:2) and GF-Epoxy/ MWCNT/GNP (70:27:2:1, 70:26:2:2, 70:25:2:3) weight percent composite sections were obtained. Dielectric properties were studied using LCR meter in the frequency range from 10Hz to 8MHz and mechanical properties were investigated using Universal Testing Machine. The mechanical tests revealed that the addition of MWCNT improves the tensile strength, cross breaking strength and the GNP enhances the tensile elastic modulus of the hybrid composite sheets. Electrical properties such as dielectric constant and impedance were found to be improved with filler addition. Finally, this work seeks to address the need of hybrid nanocomposite core in high voltage applications and need to understand the changes in electrical and mechanical properties with the addition of hybrid nanofillers.

Keywords: Electrical and Mechanical Properties, Graphene Nano Platelets, GFRP Hybrid Nanocomposites, Multi Walled Carbon Nanotubes

1. Introduction

Hybrid nano composites are the new class of materials which are widely used in numerous advanced applications of electrical, mechanical, civil, aerospace engineering fields. Advantage of using this new class of materials is that tailored/customized functionalities could be achieved with the use of nano fillers and processing techniques. Emerging technologies in the areas of Electrical transmission and distribution and energy storage applications require robust, light weight material having high dielectric constant which could withstand high operating temperatures. Thermo setting epoxy polymer matrix along with the glass fiber offers exceptional mechanical, thermal and chemical resistant properties and also provides high strength to mass and high stiffness to mass ratios in addition to flexibility and low density compared with metals[1,2]. The Glass Fiber Reinforced Polymer composite (GFRP) epoxy composites with the reinforcement of nanofillers like MWCNT, GNP could

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provide the multifunctional Hybrid Nano Composites (HNCs) materials. HNCs can have a significant mass saving in production due to inherent high strength to mass and high stiffness to mass ratios compared with metals, best suited to attain functional properties for the electrical applications. Addition of very low volume fraction of MWCNT increases dielectric constant, reduces the impedance, and improves mechanical strength and tensile modulus[1–3]. GNP improves the current density, thermal conductivity and stiffness compared to that of the copper and aluminium being non-reactive to atmospheric gases, also reduces brittleness and ductility[4,5] also significantly increases fracture toughness and fatigue strength. Though reinforcement of MWCNT and GNP could results in high performance due strong van der Waals forces between the fillers tend to agglomerate and result in uneven dispersion, where as a well-dispersed network of GNP provides a conductive path and a steady improvement in thermal conductivity with increasing concentration of GNP[6–9]. The dispersion and processing techniques play a major role for good adhesion/ bonding between the fillers and matrix to infuse the attributes of carbon nanofillers for making advanced materials. Among the various fabrication techniques pultrusion process[10] has been widely used for fabricating high performance advanced composite products.

In this work, HNCs with higher wt.% ratios of MWCNTs and GNPs were fabricated compared to the work carried out previously[11–14], a homogenous suspension was prepared by directly dispersing MWCNTs and GNPs into epoxy assisted with industrial mechanical stirrer with addition of fire retardant agent and transferred to Epoxy bath container. Glass fiber threads made to dip into epoxy bath and composite section is pulled thorough the mould using pultrusion technique. Composite Sections were cut according to test requirements for measurement of Mechanical and Electrical Properties.

2. Materials and Methods

2.1 Materials

In this study Graphene nanoplatelets 15μm particle size, surface area 120-150 m^2/g, bulk density 0.03-0.1 g/cm^3 of Sigma Aldrich Company was supplied by Ultra Lab products Bangalore. Material Multi Walled Carbon Nanotubes (MWNTs), O.D.xL 16nm x 10-20μm Specific Surface area 220m2/g, Bulk density 0.10-0.06 g/cm3 supplied by IENT Inc. Tamilnadu, India. Araldite MY 740 100pbw and Hardener HY 918 85pbw of HUNTSMAN co. and the plain glass fiber rovings with a density of 2.72 g/cm^3 procured by M/s. Ashok industries, Bangalore.

2.2 Preparation of the Epoxy/MWCNT/GNP Mixture and Fabrication

Epoxy, nano fillers, fire retardant agent (aluminium tri hydrate) ATH[15] along with the hardener were taken as per the supplier's recommended standards. Nanofillers, ATH were weighed according weight of epoxy. The different ratios are given in the Table 1. A homogeneous mixture is obtained after mechanical stirring; composite mixture is then transferred to temperature controlled Closed Container resin bath system in the pultrusion setup as shown in the Figure 1. The fiber glass is pulled with the speed of 5-6meters/Hour with the die temperature maintained at 145^{o}C-170^{o}C for curing. A rectangular section of 3.7mm thickness with 40mm width is pulled and sections of 1.5 meters were cut and transferred.

2.3 Experimental

Fully automated high-precession LCR meter (HIOKI-IM3536) and (HIOKIL2000) four-terminal probe in the frequency range from 10Hz to 8MHz is used at room temperature for electrical parameters measurements for all HNC samples. The capacitance (c), dissipation factor (tanδ) and resistance were recorded directly and from which dielectric constant (ε_r) and dielectric loss (ε") were calculated.

Tensile test was performed with tension fixture in Universal Testing Machine (UTM) in accordance to the ASTM D3039 standard samples to evaluate the tensile properties. Water absorption test conducted as per Indian Standard IS: 10192 standards.

2.4 Experimental

Fully automated high-precession LCR meter (HIOKI-IM3536) and (HIOKIL2000) four-terminal probe in the frequency range from 10Hz to 8MHz is used for electrical parameters measurements at room temperature of all HNC samples. The capacitance(c), dissipation factor (tanδ) and resistance were recorded directly and from which dielectric constant (ε_r) and dielectric loss (ε") were calculated.

Table 1. Sample identification and composition

Sample identification	Composition	MWCNT (wt.%)	GNP (wt.%)	ATH (wt.%)
A	Epoxy/GF	--	--	20%
B	Epoxy/GF/ MWCNT	2%	--	20%
C	Epoxy/GF/ GNP	--	1%	20%
D	Epoxy/GF/ GNP/MWCNT	2%	1%	20%
E	Epoxy/GF/ GNP/MWCNT	2%	2%	20%
F	Epoxy/GF/ GNP/MWCNT	2%	3%	20%

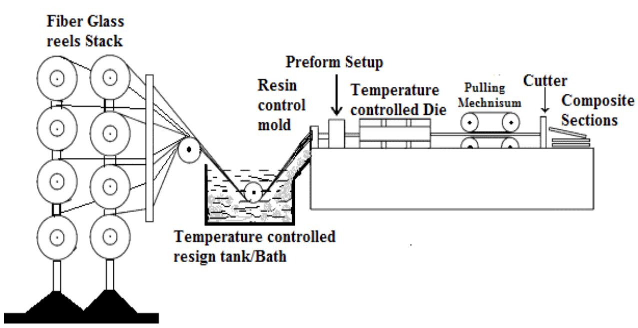


Figure 1. Pultrusion process.

3. Results and Discussion

3.1 Water Absorption of GF-Epoxy/GNP-MWNT HNC Samples

HNCs specimens were cut into square piece of 40mm±0.5mm; the thickness of the specimen is less than 25mm as per Indian Standard IS: 10192 standards. First weight (W_1) of the specimen was first measured in air and then the specimen was immersed in distilled water for a period of 24hours at room temperature. Before immersion, care was taken to remove any air bubbles sticking to specimen before taking the weight in water. Specimens were removed from the water wiped properly using absorbing paper and weighed immediately and the weight (W_2) was recorded. The water absorption ratio was calculated by eq.1

$$\text{Water Absorption} = \frac{W_2 + W_1}{W_1} \times 100 \qquad (1)$$

Figure 2 shows the water absorption percentage of Neat and HNC samples with different nanofiller wt.% is as shown in Table.2. The water absorbency of HNC increased in the case of sample with single nanofiller, when both the fillers were added it reduces. Absorption in the case of Epoxy composite is smaller than that of Epoxy/GNP nanocomposite and higher than that of Epoxy/MWNTs, because GNPs have natural tendency to absorb water more than MWNTs and also due to higher exposed surface area of nanoplatelets. Epoxy/GNP-MWNT nanocomposite is more hydrophobic than Epoxy/MWNTs and Epoxy/GNPs nanocomposite. Water absorption on Epoxy/MWCNT-GNP nanocomposite surfaces decreases with increasing nanofillers[16].

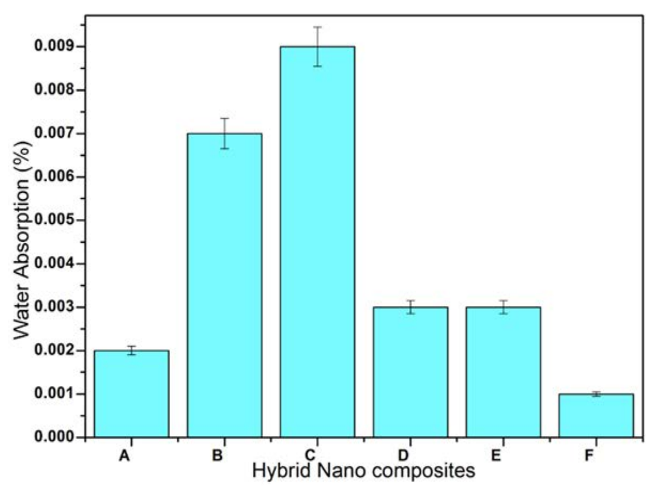


Figure 2. Water Absorption percentage in HNCs.

Table 2. Physical and mechanical properties test results

Sample	Composition	Physical properties		Mechanical properties	
		Specific gravity (g/cc)	Water absorption	Tensile strength (N/mm^2)	Cross breaking strength (N/mm^2)
A	Epoxy/GF	2.6	0.002	451.3	1468.5
B	Epoxy/GF/2wt.%MWCNT	2.66	0.007	450.62	1323.5
C	Epoxy/GF/1wt.%GNP	2.49	0.009	397.28	1298.8
D	Epoxy/GF/1wt.%GNP/2wt.%MWCNT	2.49	0.003	350.35	1228.89
E	Epoxy/GF/2wt.%GNP/2wt.%MWCNT	2.61	0.003	394.74	1484.34
F	Epoxy/GF/3wt.%GNP/2wt.%MWCNT	2.49	0.001	426.53	1515.11

3.2 Tensile Properties of Epoxy/GNPs-MWNTs HNC Samples

The flat specimens of required size were fixed between the grips of each head of the testing machine in a way that the direction of force applied to the specimen is coincident along with the axis of the specimen. A very low rate of tensile speed 0.5 mm/min is applied at room temperature of 28°C. For each type of HNC of varying filler content, the tensile test was carried out on at least five specimens; the result was the average value of the five measurements.

$$\textit{Tensile Strength} = \frac{W}{BD} \tag{2}$$

where, W = Load in Kgs, B= Breadth in m, and D = thickness in m.

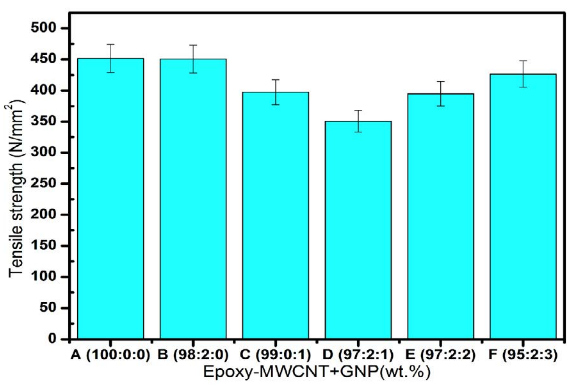


Figure 3. Tensile strength of epoxy-glass fiber composite and HNC

The tensile strength of the pure epoxy and their nanocomposites with different various filler loading of MWCNTs and GNPs is shown in Figure 3. With the addition of MWCNTs there is no significant change in the tensile strength, but as the content of GNPs increased to a range of 0 to 1%, the tensile strength decreased sharply. In a weight percentage range of 1% to 3%, the change in tensile strength was increased slightly but there is a considerable improvement in cross breaking strength as listed in Table 2.

3.3 Electrical Properties

The capacitance, loss tangent and impedance were measured at room temperature in the applied electric field with the frequency range from 10Hz to 8MHz. The complex impedance of the sample with the real (Z_r) and imaginary components (Z_i) can be calculated by using (3) and (4).

$$Z_r = |Z| \cos \varphi \tag{3}$$

$$Z_i = |Z| \sin \varphi \tag{4}$$

The variation of total impedance of the HNC with frequency is as shown in Figure 4. The addition of MWCNT and GNP decreases the impedance of the epoxy resin by a few orders of magnitude. This signifies that with the addition of nanofillers the conductivity of the composite increases.

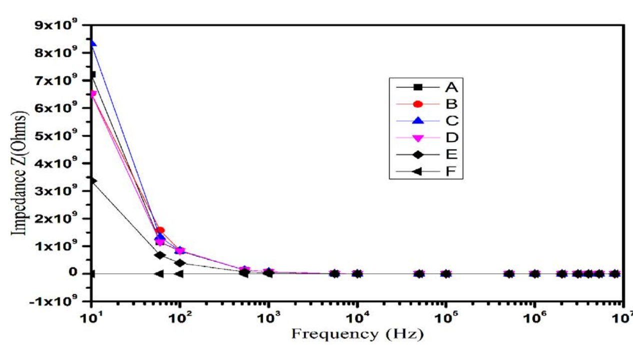


Figure 4. Impedance of HNCs versus the applied frequency.

Figure 5 shows Cole–Cole plot of the imaginary component Z_i versus the real component Z_r of complex impedance Z for all HNC samples. The constructed plots are distorted semicircles with maxima shifted toward the negative Z_r axis, indicating the decrease in bulk resistance (R_b) of the HNC, with increasing MWCNT and GNP. Therefore, the nanocomposites will be less resistive or more conductive in nature. The appearance of spikes in low frequency response region is attributed to additional capacitance and resistance contribution arising from dielectric relaxation in the HNCs structure[17,18].

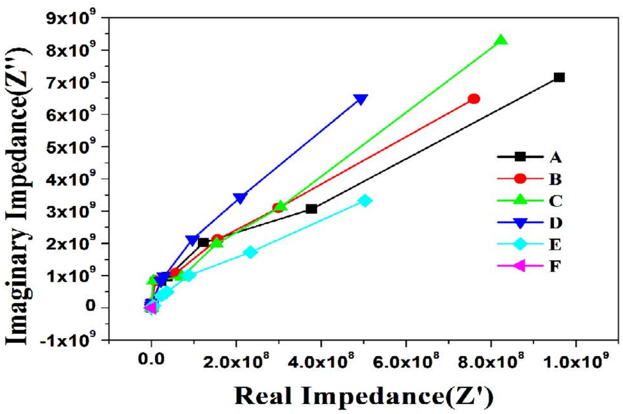
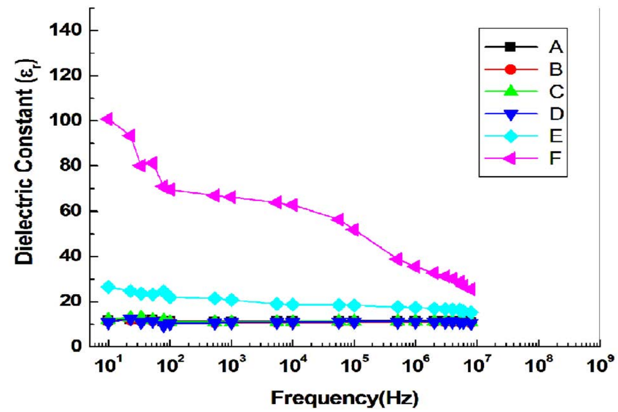


Figure 5. Cole-Cole plot of complex impedance components for HNCs.

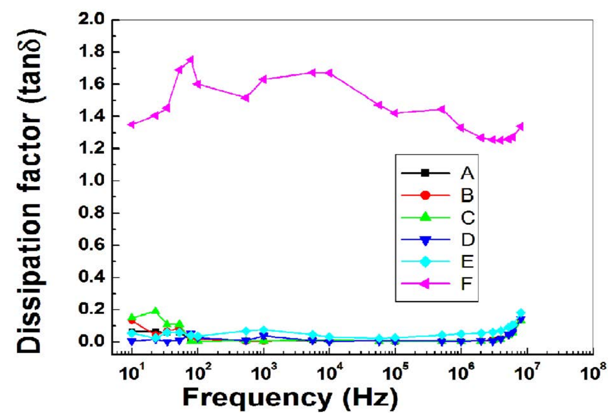
From the measured value of capacitance, the dielectric constant (ε_r) is calculated from (5)

$$\varepsilon_r = \frac{C * t}{\varepsilon_o * A} \tag{5}$$

where, C is the capacitance in farads, ε_o is the dielectric constant ($8.8549X10^{-12}$F/m) in free vacuum, t is the thickness of the specimen in m, A is the area of the electrode in m^2. The variations of the impedance and dielectric properties with increase in frequency ranging from 10Hz to 8MHz for Epoxy-GF and HNC are shown in Figure 6(a) and (b) respectively. At lower frequency range (10-10^4Hz), the dielectric constant of HNC decreased, and above which it decreases up to 10^8Hz. The dissipation factor shown in Figure 6(b) of the composite was lower than 0.2 for all HNC except for the HNC with the 2wt.%MWCNT+1wt.%GNP, this shows promising HNC with low dielectric loss (D). As mentioned in the percolation theory, both the dielectric constant and loss tangent exhibit transition behavior at vicinity of percolation. Thus, considerably high loss tangent of the composites with higher volume fraction of MWCNT and GNP could be observed in Figure 6(b).



(a)



(b)

Figure 6. **(a)** Frequency dependency of dielectric constant (ε_r). **(b)** Frequency Vs Dissipation factor of HNCs.

4. Conclusion

The individual and simultaneous effects of MWNT and GNP on the mechanical properties and electrical properties of an epoxy glass fiber HNCs were studied. It can be concluded from the results that the addition of both MWNT and GNP simultaneously improves the electrical properties. But introducing GNP into the MWCNT/epoxy nanocomposites reduces the mechanical properties. The dielectric properties of the composite are improved significantly with the combined effect of MWNT and GNP. It was also found that the simultaneous presence of MWNT and GNP can improve the cross breaking strength but it reduces the ultimate tensile strength. The presence of MWCNT and GNP is found to decrease the water uptake of nanocomposites compared to neat epoxy due to the excellent barrier properties of these nano-fillers.

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6. References

1. Coleman JN, Khan U, Blau WJ, Gun'ko YK. Small but strong: A review of the mechanical properties of carbon nanotube-polymer composites. Carbon. 2006; 44(9):1624–52. https://doi.org/10.1016/j.carbon.2006.02.038
2. Simcha S, Dotan A, Kenig S, Dodiuk H. Characterization of hybrid epoxy nanocomposites. Nanomaterials. 2012; 2(4):348–65. https://doi.org/10.3390/nano2040348. PMid:28348313. PMCid:PMC5304600
3. Chatterjee S, Nafezarefi F, Tai NH, Schlagenhauf L, Nüesch FA, Chu BTT. Size and synergy effects of nanofiller hybrids including graphene nanoplatelets and carbon nanotubes in mechanical properties of epoxy composites. Carbon. 2012; 50(15):5380–6. https://doi.org/10.1016/j.carbon.2012.07.021
4. Li W, Dichiara A, Bai J. Carbon nanotube-graphene nanoplatelet hybrids as high-performance multifunctional reinforcements in epoxy composites. Composites Science and Technology. 2013; 74:221–7. https://doi.org/10.1016/j.compscitech.2012.11.015
5. Im H, Kim J. Thermal conductivity of a graphene oxide-carbon nanotube hybrid/epoxy composite. Carbon. 2012; 50(15):5429–40. https://doi.org/10.1016/j.carbon.2012.07.029
6. Geim AK. Graphene: Status and prospects. Science. 2014; 1530(2009):1530–4. https://doi.org/10.1126/science. 1158877. PMid:19541989
7. Shahil KMF, Balandin AA. Thermal properties of graphene and multilayer graphene: Applications in thermal interface materials. Solid State Communications. 2012; 152(15):1331–40. https://doi.org/10.1016/j.ssc.2012.04.034
8. Seshaiah T, Vijayakumarreddy K. Effect of hybrid mwcnts/graphene on tensile properties of reinforced unidirectional e-glass/epoxy composite. IOP Conference Series: Materials Science and Engineering. 2018; 377:012050. https://doi.org/10.1088/1757-899X/377/1/012050
9. Yue L, Pircheraghi G, Monemian SA, Manas-Zloczower I. Epoxy composites with carbon nanotubes and graphene nanoplatelets - Dispersion and synergy effects. Carbon. 2014; 78:268–78. https://doi.org/10.1016/j.carbon.2014.07.003
10. Ramesh B, Elayaperumal A, Satishkumar S. Drillability study of pultruded and sheet moulding compound thick polymeric composites. Proceedings of the Institution of Mechanical Engineers, Part B. 2015; 231(2):268–85. https://doi.org/10.1177/0954405415573678
11. Anwar Z, Kausar A, Rafique I, Muhammad B. Advances in epoxy/graphene nanoplatelet composite with enhanced physical properties: A review. Polymer-Plastics Technology and Engineering. 2016; 55(6):643–62. https://doi.org/10.1080/03602559.2015.1098695
12. Montazeri A, Montazeri N, Farzaneh S. Thermo-mechanical properties of multi-walled carbon nanotube (mwcnt)/epoxy composites. International Journal of Polymer Analysis and Characterization. 2011; 16(3):199–210. https://doi.org/10.1080/1023666X.2011.563595
13. Pandey G, Thostenson ET. Carbon nanotube-based multifunctional polymer nanocomposites. Polymer Reviews. 2012; 52(3–4):355–416. https://doi.org/10.1080/15583724.2012.703747
14. Marouf BT, Mai YW, Bagheri R, Pearson RA. Toughening of epoxy nanocomposites: Nano and hybrid effects. Polymer Reviews. 2016; 56(1):70–112. https://doi.org/10.1080/15583724.2015.1086368
15. Kausar A, Rafique I, Anwar Z, Muhammad B. Recent developments in different types of flame retardants and effect on fire retardancy of epoxy composite. Polymer-Plastics Technology and Engineering. 2016; 55(14):1512–35. https://doi.org/10.1080/03602559.2016.1163607
16. Zhao H, Li RKY. Effect of water absorption on the mechanical and dielectric properties of nano-alumina filled epoxy nanocomposites. Composites Part A: Applied Science and Manufacturing. 2008; 39(4):602–11. https://doi.org/10.1016/j.compositesa.2007.07.006
17. Elimat ZM, Zihlif AM, Ragosta G. Study of ac electrical properties of aluminium-epoxy composites. Journal of Physics D: Applied Physics. 2008; 41(16). https://doi.org/10.1088/0022-3727/41/16/165408
18. Abu-Jamous A, Zihlif AM. Study of the electrical conduction in poly(ethylene oxide) doped with iodine. Physica B: Condensed Matter. 2010; 405(13):2762–7. https://doi.org/10.1016/j.physb.2010.02.050