EFFECT OF FIBER ORIENTATION ON THE MECHANICAL BEHAVIOR OF GLASS FIBER/EPOXY HYBRID NANOCOMPOSITES CONTAINING MULTI WALLED CARBON NANOTUBES AND GRAPHENE

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ABSTRACT

This paper deals with the influence of the lay-up configuration of glass fiber reinforced epoxy matrix hybrid nanocomposites which are developed containing multi-walled carbon nanotubes (MWCNTs) and graphene (GPN) to investigate merged effect of nanoreinforcements on the mechanical performance of nanocomposites. The accompanying laminates were produced by mechanical ultrasonicator with compression molding for this study: [0°]s, [90°]s, [0°/90°]s and [45°/45°]s oriented. Both the nanofillers were functionalized before incorporating into epoxy matrix to promote interfacial interactions. The concentrations of both MWCNTs and GPN filled in the reinforced glass epoxy matrix nanocomposites were increased systematically, i.e. 0.1wt.%, 0.2wt.% and 0.3wt.% while composites containing individual nano reinforcements were additionally manufactured for comparison. The developed nanocomposites were characterized microstructurally by scanning electron microscopy (SEM) and mechanically by tensile, flexural, and compressive tests. Homogeneous dispersion of MWCNTs and GPN was observed under SEM, which resulted in the revampment of mechanical properties of nanocomposites. The [0°]s glass/epoxy hybrid nanocomposites containing 0.2wt.% MWCNTs and 0.2wt.% GPN demonstrated 28.4% enhancement in tensile strength and modulus revamped to 38.4%, respectively. Flexural strength and modulus likewise demonstrated a rise of 84% and 64%, respectively. Compressive strength and modulus likewise demonstrated a rise of 480% and 74%, respectively. Strikingly, fracture strain likewise enhanced in both the tensile, flexural and Compressive testing. The impact resistance enhanced to 237% demonstrating a significant revampment in the toughness of hybrid nanocomposites.

Key words: E-glass fibre, MWCNTs, GPN, Mechanical properties, HR-SEM.
1. INTRODUCTION

The two allotropic forms of carbon, i.e. graphite and diamond, in nanometer size, i.e. Graphene (GPN) and Multi Walled carbon nanotubes (MWCNTs), respectively have recently attained much attention as reinforcing medium in polymer matrices[1-2].These tubular and spherical structural types of carbon have appealing characteristics for use as nanoreinforcements in a spread of polymeric substances to overcome their inherent limitations and to develop a peculiar composition of composites with improved mechanical and purposeful properties [3].

In contrast, GPN consists of with an average thickness of the 5-10 nm [4] are supplied in varying sizes as much as 50 microns that have attracted attention as a promising candidate to create new polymer-nano composites due to its excellent properties and ready availability of its precursor, graphite. However, the inclusion of graphene (GPN) in epoxy has been proven to enhance mechanical and electrical properties with respect to the un-reinforced epoxy [5-10], thus showing promise for use of GNP-reinforced epoxy because of the matrix phase in a fiber composite. The resulting GNP/carbon fiber/epoxy hybrid composite potentially show upgrades in mechanical properties with respect to conventional carbon fiber/epoxy composites. It's been demonstrated [6-10] that the effect of GPN on GNP/polymer composite mechanical properties is administrated by the amount of GPN added to the polymer and the dispersion of the GPN inside the polymer. Accordingly GPN have been successfully incorporated in thermosetting (epoxy, phenolic, polyurethane etc.) and thermo polymers (polyethylene, polypropylene, polystyrene, nylon etc.) to fabricate composites [7].

CNTs are seamless hollow tubes of rolled up graphene sheets. An individual graphene sheet forms a single walled carbon nanotube (SWCNT) while multiple graphene sheets constitute a concentric multiwalled carbon nanotube (MWCNT). CNTs are generally produced by chemical vapor deposition technique. This technique produces CNTs in preponderance and usually used for the reinforcement in composite materials[11]. In any case, with no driving force, as-grown CNTs do not disperse well in polymeric matrices and therefore different processes have been developed to disperse CNTs in polymers including ultrasonication, calendaring, ball milling, shear mixing and extrusion, together with the physical and chemical functionalization techniques to increase the interfacial interactions between CNTs and polymers. As an outcome, CNTs have been effectively incorporated in different types of thermo and thermosetting polymers to produce their respective composites[1,12-14].

In addition to CNTs and GPN, other forms of carbon, i.e. SWCNTs, MWCNTs, nanoclays, nanorods, nanodiamonds, carbon nanofibers, graphene, graphene oxide and graphite platelets and many other nanoreinforcements are used in thermo and thermosetting polymers for a variety of mechanical, thermal, electrical and functional properties including increased strength, enhanced toughness, electrical conductivity, shape memory effect, flame resistivity, rheological behavior and electromagnetic field absorption[15-26].
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Exclusively, CNTs and GPN have been utilized as a part of polymeric matrices, especially, in epoxy for an assortment of purposes including the increased functional and mechanical properties. In any case, their comprehensive combined effect has not yet been explored up on the mechanical performance of the composites, next to the receptiveness of a single report on polyvinyl alcohol composites containing SWCNTs and GPN wherein only hardness and tensile modulus were reported [3,19].

The reports of polymeric composites containing individual CNTs and GPNs demonstrates enhancement in mechanical properties [27–31]. For instance, improvement in tensile properties has been appeared in epoxy matrix composites containing CNTs in restricted quantities, i.e. up to 2.0 wt.% [32–35] and additionally in large quantities, i.e. up to 8.0 wt.% [35]. The three sorts of CNTs, i.e. SWCNTs, DWCNTs and MWCNTs have all exhibited to increase the tensile and modulus of composites [36,37]. Similarly, flexural properties of the composites have additionally indicated rises with the addition of CNTs; for instance, 120% improvement in flexural strength was seen in an investigation by including just 1 wt.% MWCNTs in epoxy matrix [38]. Different investigations have additionally detailed the enhancements in flexural strength and modulus of CNTs reinforced polymeric matrix composites [38–40]

As of late, binary nanoreinforcements have been incorporated in hybrid fibre polymeric matrices to build up a unique class of hybrid fibre nanocomposites [41]; the addition of individual and two nanoreinforcements demonstrates distinctive impacts on the mechanical and functional properties of composites. As section of the analyzed combinations of binary nanoreinforcements in polymeric matrices are: CNTs-GNPs [41], CNTs- Carbon black [42], CNTs-SiC [43], CNTs-Silica [44], CNTs-Copper [45].Utilizing these merges, the explored functional and mechanical properties are electrical and thermal conductivities, dielectric and microwave properties, depositary and defecit modulus, impact and fracture toughness, and tensile, flexural and tribological properties.

In the present investigation, MWCNTs, GPN and MWCNTs/GPN have been incorporated in various stacking sequence of glass fibre [0°]s, [90°]s, [0°/90°]s and [45°/45°]s reinforced epoxy matrix in constrained amounts to guarantee their uniform dispersion in order to accomplish the advantages of these three nanoreinforcements upon the mechanical performance of glass fibre/epoxy matrix. Epoxy was particularly chosen because of its wide applications in industries. Besides, epoxy resin offer low volume shrinkage amid curing and can be utilized without solvent and discharges no by-products. In any case, in spite of these attributes, the relatively weak mechanical properties of epoxy resins have confined its utilization in numerous potential applications. MWCNTs and GPNs were particularly chosen in blend with regards to the best of authors knowledge, this combinations has not been utilized before in glass fibre/epoxy matrix in spite of the fact that reports on CNTs/epoxy and GPNs/epoxy are available, as mentioned above and discussed about further beneath. The composites were prepared by in situ polymerization subsequent to blending the nanoreinforcements in epoxy utilizing ultrasonication technique to ensure their uniform dispersion. So as to create solid collaborations of nanoreinforcements with the glass fibre/epoxy matrix, MWCNTs and GPNs were functionalized. The tensile, flexural, and compressive properties were measured in the present investigation and the morphology of the developed composites was researched by scanning electron microscopy(SEM). The obtained results were compared with the available data on MWCNTs/Glass fibre/epoxy and GPNs/glass fibre/epoxy composite systems regarding relative mechanical improvements in connection to the loadings of nanoreinforcements and the quality of their dispersion.
2. EXPERIMENTAL

2.1. Materials

MWCNTs were procured from United Nano Tech innovations Pvt Ltd, Bangalore, India. The length of MWCNTs was 3–10 micron with the diameter of 12–15 nm; the purity level was >97%. MWCNTs were acid-refluxed in 40 ml of 3:1 volume ratio of concentrated sulphuric and nitric acids at 25 °C for 35 min (Fig.1a). The acid-treated MWCNTs were then washed with distilled water, sonicated and centrifuged at 2500 rpm for 20 min [44] GPN were provided by Bottom up Technologies Corporation, Jharkhand, India. The average GPN length size was 5-10 micron with particle size thickness of 5–10 nm; the purity level was >99%. GPN were air-oxidized at 440°C for 5 h to remove unwanted carbonaceous impurities and then treated with UV/O3 cleaner (Jelight 144AX-220) emitting radiation of 28 μW/cm2 by low pressure mercury vapor grid lamp[11] (Fig.1b). Plain weave 200 gsm E-glass fiber from Arun fabrics Pvt Ltd, Bangalore., The epoxy resin utilized in the present examination was Araldite®LY 556 with a curing agent Aradur® HY 951.

![Figure 1 Nanoreinforcements used in the glass reinforced epoxy matrix composites (a) MWCNTs and (b) GPN.](image)

2.2. Manufacturing

For the manufacturing impact of the lay-up designs of glass fibre reinforced epoxy hybrid nanocomposites, MWCNTs and GPN were dispersed in epoxy resin in ascertained amounts and ultrasonicated for 4h at room temperature. Six glass/epoxy composites containing individual 0.1-0.3wt.% MWCNTs and 0.1-0.3wt.% GPN, and three glass/epoxy composites containing 0.1wt.%, 0.2wt.% and 0.3wt% of each one of MWCNTs and GPN were manufactured. MWCNTs/GPN/epoxy(i.e. slurry of hybrid nanoreinforcement) blends were degassed for 10 min after sonication before reinforced glass fabric. Curing agent was added to the slurry of nano followed by mechanical blending. In parallel aluminum dies, which were coated with polyvinyl alcohol before pouring. The coating procedure was performed to keep away from the sticking of slurry of nanomixture to the die cavity and the simple expulsion of the cured composites. After that slurry of hybrid nanomixtures were poured in between the stacking sequence designs of (i) unidirectional at [0°]s, (ii) unidirectional at [90°]s (iii) balanced and symmetrical laminate with a combination of [0°/90°]s layers; and (iv) angle ply symmetric [45°/45°] glass fibre oriented then followed by compression molding to prepare hybrid nano glass/epoxy composites. The molded glass/epoxy composite specimens were dried for 24 h at room temperature and later expelled from the dies. Post-curing was performed at 100 °C for 4 h in an electric oven. The size of the prepared composites plates was 300 × 300 × 3 mm.
2.3. Characterization
The fabricated composites were cut into required dimensions for tensile, flexural, and compressive tests. ASTM standards of D3039, D790 and, D3410 were considered for tensile, flexural and compressive tests and the dimensions of the specimen were 250 × 25 × 3 mm, 125×12.7×3 mm, and 140×15×3 mm respectively. Tensile tests were performed on a tensile testing machine (Hydraulic Instron 3039) at a crosshead speed of 1.5 mm/min to acquire tensile strength, tensile modulus and fracture strain in the composites. Flexural tests were performed under three-point loading on a same testing machine at a crosshead speed of 1.5 mm/min to obtain flexural strength, flexural modulus and fracture strain and compressive tests were performed on a same testing machine (Hydraulic Instron 3039) at a cross head speed of 2mm/min to obtain compressive strength, compressive modulus and fracture strain with the maximum load capacity of 100KN At minimum of five tests were performed for each sort of composite system and for each sort of mechanical testing. High resolution Scanning electron microscopy (HR-SEM) was performed on glass fibre reinforced containing MWCNTs, GPNs and fractured surfaces of the composite specimens

3. RESULTS AND DISCUSSION
3.1. Microstructures of Composites
The morphology of the fractured surfaces of neat glass/epoxy composite and [0°]s, [90°]s, [0°/90°]s, and [45°/45°]s glass fiber reinforced epoxy matrix along with MWCNTs and GPNs nanocomposites were investigated utilizing SEM (Fig.2). An increase in the contents of nanofillers can easily be observed in the images (Fig.2d, g and j) and a change in fracture surface of the matrix was flat, and some cracks were seen in matrix side near the fibre-matrix morphology of composites is visible in comparison to neat glass/epoxy composite because of the agglomeration of MWCNTs, the matrix is absent on the fracture surface of nanocomposites, and only bridging of some nanoparticles between the fibers is clear (Fig.2a). The distribution of nanoreinforcements can be seen in Fig. 2d and f; no agglomerates of MWCNTs or GPNs were found on the polished surfaces of the composites. The functionalization of nanoreinforcements increased the adhesion of MWCNTs and GPNs with the epoxy matrix; subsequently, it can be seen that nanoreinforcements were held firmly in the epoxy matrix. Pullout phenomenon of MWCNTs is especially observable, which is an indication of increased toughness. The firmly held nanoreinforcements along with their uniform distribution without the presence of agglomerates foresee a significant effect of MWCNTs and GPN on the mechanical performance of composites.
Fig. 2. SEM images showing fractured surfaces of (a) neat glass/epoxy, (b) to (d) glass/epoxy composites containing 0.1-0.3wt% MWCNTs, (e) to (g) glass/epoxy containing 0.1-0.3wt.% GPN and (h) to (j) glass/epoxy containing 0.1-0.3wt.% MWCNTs/GPN. High magnification SEM images of polished surfaces of glass/epoxy composites containing (c), (f) 0.2wt.% MWCNTs and 0.2wt.% GPN and (i) 0.2wt.% MWCNTs/GPN, demonstrates the uniform distribution of nanoreinforcements.
3.2. Mechanical properties of composites

3.2.1. Tensile Properties

Unidirectional neat glass/epoxy composite exhibits a tensile strength of 604.8 ± 5.5 MPa, which increased to 624.3 ± 3.0 MPa (3.3% rise) by including 0.2wt.% MWCNTs-glass/epoxy composite and 515.3 ± 3.7MPa (14% decrease) by incorporating 0.2wt.% GPN-glass/epoxy composite. The consolidated inclusion of MWCNTs and GPN-glass/epoxy composite at the fractions of 0.1wt.%, 0.2wt.% and 0.3wt.% each, additionally increasing the tensile strength of composites, i.e., 612.2 ± 3.7 MPa, 774.8 ± 4.5 MPa and 259.1 ± 3.9 MPa (2%, 28%, rise and 57% decrease), at [0°]s specimens respectively, when contrasted with other stacking sequence lay-up i.e. [90°]s, [0°/90°]s and [45°/45°]s oriented nanocomposite shown in Fig.3a. Literature generally demonstrates a rising trend in tensile strength after incorporating CNTs in different fibre/epoxy matrix composites however the extent of improvement varies with the loading of CNTs in different investigations. For Instance, an increase in tensile strength of carbon fibre/epoxy matrix was observed by adding CNTs in limited quantities, i.e. 1wt.% [41]; however, the extent of improvement (28%) was not considerable as compared to that observed in the present study. Similarly, tensile strength increased in another study by adding large quantities of MWCNTs, in neat epoxy composite i.e. up to 8.0wt.% but the extent of improvement, i.e. 65% [46] and 80% [40] was not comparable in relation to the quantity of MWCNTs used. In contrast, reports are also available which show no significant effect upon tensile strength even after adding individual 0.2wt.% MWCNTs, 0.2wt% GPN-glass/epoxy composite and binary nanoreinforcement of 0.2wt% MWCNT/GPN-glass/epoxy composite [50].GPN are accounted for increasing the tensile strength of neat epoxy matrix [8,9,10]; for instance, a 52.7% increase in tensile strength was seen in an investigation by adding just up to 0.3 wt.% GPN [6,7].

Comparable to tensile strength, the tensile modulus of the unidirectional glass hybrid nanocomposite[0°]s additionally increased by the inclusion of individual nanoreinforcements (MWCNTs,GPN) in glass/epoxy composite at a stacking of 0.2wt.% each, i.e. 14.5 ± 0.4 GPa and 18.3 ± 0.1 GPa, respectively, in comparison to neat glass/epoxy composite, i.e. 13.2 ± 0.2 GPa exhibiting a rise of 9.8% and 38%. The consolidated effect of MWCNTs and GPN-glass/epoxy composite likewise exhibits a rise in the tensile modulus values, i.e. 14.3 ± 0.4 GPa, 16.4 ± 0.3 GPa and 14.9 ± 0.3 GPa; at the loading points of 0.1wt.%, 0.2wt.% and 0.3 wt.% each indicating a rise of 8.3%, 24% and 12.8% individually, when compared with other orientations of glass/epoxy nanocomposite shown in Fig.3b. Literature generally for the most part a rising trend is observed in tensile modulus of neat epoxy matrix composites by including CNTs [39,40]; for instance, the elastic modulus increased continuously in composites containing coiled CNTs [47] and SWCNTs, DWCNTs and MWCNTs, both un-treated and NH2-treated, increased the tensile modulus of epoxy matrix composites [41,42]. In a riveting examinations, 0.5wt.% MWCNTs were incorporated in epoxy matrix with various matrix stiffness values by varying the quantity of curing agent; it was discovered that the effect of MWCNTs was noteworthy in ductile matrix which gradually diminished with an increase in the stiffness of the matrix [48]. The incorporation of 0.3wt.% GPN in epoxy matrix has likewise brought about enhancing the tensile modulus of epoxy matrix composites however further loadings of GPN switched the rising pattern [18]. In contrast, a reverse pattern is additionally seen in different investigations where elastic modulus diminished ceaselessly in epoxy matrix composites by including CNTs [49,50] and reports are additionally accessible, which demonstrate no significant improvement in tensile modulus even after the loading of SWCNTs at 0.1wt.% [51] and MWCNTs up to 1wt.% [38]. In an alternate study, the
expansion of MWCNTs increased the tensile modulus more than GPN when incorporated in the same loadings \[48\], as additionally observed in the present investigation.

A comparative pattern was observed elsewhere; however, maximum tensile modulus was observed at 0.2wt.% MWCNTs/GPN-glass/epoxy composite, which at that point diminished continuously up to 0.3 wt.% MWCNTs/GPN \[12,13\]

In addition to the increase of tensile strength and modulus of the composites the fracture strain is also increased by including 0.2wt.% MWCNTs-glass/epoxy composite, i.e. 8.2 ± 0.2 (6.36% rise) in comparison to neat glass/epoxy, i.e. 5.0 ± 0.1, which is an indication of enhanced ductility and toughness of the composites at \([0^\circ]s\). However, fracture strain diminished in composites containing 0.1wt.% GPN-glass/epoxy composite, i.e. 3.1 ± 0.1 demonstrating a fall of 3.8%. The incorporation of both nanoreinforcements, i.e. MWCNTs and GPN in glass/epoxy composite at the fractions of 0.1wt.% , 0.2wt% and 0.3wt.% each, however, it demonstrates a rising pattern, i.e. 5.34 ± 0.1, 9.3 ± 0.2 and 5.54 ± 0.1 however it is not exceptionally significant and demonstrating a rise of 1.64% and 8.56% and 1.05% but loading of 0.2wt.% to each filler demonstrated an value of 6.52 ± 0.25, which is a 30% rise in fracture strain (Fig. 3c).
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Both increasing [49] and diminishing [10] patterns have been accounted for the fracture strain in the wake of incorporating MWCNTs and GPNs in polymeric matrices. For instance, an increase in fracture strain was observed by adding 1wt.% MWCNTs [38] and in another investigation, surprisingly, the rise of fracture strain up to 20% was seen in epoxy matrix composites containing 8.0wt.% MWCNTs, which increased persistently from ~9% in neat epoxy specimens [50]. In the present investigation, the most astounding fracture strain of unidirectional glass/epoxy[0°]s composites containing MWCNTs increased while composites containing GPN demonstrated a decrease at [90°]s. The composites containing both nanofillers (MWCNTs/GPN) in [0°]s, glass/epoxy composite demonstrated rise in the fracture strain values despite the presence of GPN.

3.2.2. Flexural Properties

Similar to tensile properties, the addition of nanoreinforcements increased the flexural properties of in [00]s glass/epoxy. Neat glass/epoxy demonstrated the flexural strength of 129.5 ± 6.2 MPa, which increased to 654.6 ± 5.0 MPa (80% rise) by including 0.2wt.% MWCNT- glass/epoxy and 598.2 ± 4.5 MPa by addition of 0.2wt.% GPN-glass/epoxy exhibits a rise in the flexural strength, i.e. (78.42%). (Fig. 4a). The addition of both nanofillers at 0.1wt.%, 0.2wt.% and 0.3wt.% each, in any case, it increases the values, i.e. 593.4 ± 7.2 MPa, 682 ± 8.5 ± 4.0 MPa, 350 ± 5.2 MPa indicating a rise of 79%, 81% and 64%, respectively. Insignificant increase in flexural strength was observed in an alternate study on epoxy matrix composites containing 2.3wt.% halloysite nanotubes [52]. However, up to 120% change in flexural strength was observed in another investigation by including 1wt.% MWCNTs in epoxy matrix which diminished to 100% by increasing the MWCNTs substance to 2wt.% [43]. Rather than utilizing high loadings of nanoreinforcements, the flexural strength also increased after including low contents of MWCNTs [44] and GPNs [17] individually in epoxy matrix, i.e. 0.4wt.% and 0.1wt.%, respectively.

The flexural modulus likewise enhanced with the addition of MWCNTs and GPN in [0°]s glass/epoxy individually at 0.2wt.%, i.e. 25 ± 0.8 GPa and 25 ± 0.8 GPa in contrast with neat glass/epoxy, i.e. 9 ± 0.1 GPa demonstrating a rise of 64% and 60% respectively (Fig. 4b).
The addition of both MWCNTs and GPN in the [0°]s glass/epoxy at 0.1wt.%, 0.2wt.% and 0.3 wt.% loadings resulted in the increased flexural modulus values, i.e. 22.1 ± 0.1 GPa, 30.7 ±0.4 GPa and 10.2 ± 0.2 GPa, which demonstrate a rise of 59%, 70% and 10%, respectively. The incorporation of MWCNTs and GPN in various stacking sequence lay-up of 90°, 0°/90° and 45°/45° glass/epoxy matrix has demonstrated for the most part an increase in flexural modulus in small fractions; however, the pattern reverses after increasing the content of nanoreinforcements. For instance, in an investigation flexural modulus increased with the addition of 0.1wt.% MWCNTs in epoxy that diminished significantly by increasing the MWCNTs content up to 0.35wt.% in the epoxy [45]. In another examination, flexural modulus is accounted for a similar pattern, i.e. sharp increment after including ~0.5wt.% MWCNTs followed by a decrease when their content was increased to 3.5wt.% in epoxy matrix [46].
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The maximum fracture strain in flexural testing increased when 0.2wt.% MWCNTs and 0.2 wt% GPN were included in unidirectional glass/epoxy composite [0°]s, i.e. 6.2 ± 0.15%, a (244.4 % rise), 6.5 ± 0.11% (261.1% rise) when compared with neat glass/epoxy 1.8 ± 0.1% yet GPNs affected adversely and reduced the flexural strain, i.e. 1.5 ± 0.15 (27.7% fall). The binary addition of MWCNTs and GPN in glass/epoxy composite at 0.2wt.% and 0.1wt.% each resulted in modest increase in flexural strain, 6.1 ± 0.18 (238.8% rise) and 7.2 ± 0.16 (300% rise). However, composites containing 0.2 wt.% MWCNTs and GPN each, demonstrated a significant rise, i.e. 5.6 ± 0.19% (211% rise), as shown in Fig. 4c. The addition of 0.4 wt.% MWCNTs had demonstrated the increase in flexural strain [44] at the effect of GPN on the flexural strain isn't accessible in literature.

3.2.3. Compressive Properties

Similar to tensile and flexural properties, neat glass/epoxy composite demonstrated a compressive strength of 14.6 ± 4.12MPa (Fig.5 a), which increased to 80.1 ± 3 (448% rise) by including 0.2 wt.% MWCNTs in glass/epoxy composite and 85.8 ± 0.6 MPa (487% rise) by the expansion of 0.2wt.% GPN in glass/epoxy composite. The effect of the incorporation of low content of MWCNTs in glass/epoxy matrix is more articulated in comparison with GPN/glass/epoxy matrix. Literature demonstrates the comparative outcomes when an comparison of increase in compressive strength is drawn between polymeric matrix composites containing GPN [7,8,9] and MWCNTs [50,51]; the influence of SWCNTs [3,50] is much more significant than MWCNTs. The combined effect of MWCNTs and GPN at a loading of 0.2wt.% each likewise demonstrated a rise in the compressive strength value, i.e. 84.6 ± 2.6 MPa (479% rise), which is higher than the solitary effect of MWCNTs and not as much as the individual influence of GPN on compressive strength. Additional rise in the contents of MWCNTs and GPN in glass/epoxy matrix, i.e. 0.1wt.% to 0.3wt.% each, increased the hardness of composites, i.e. 45.4 ± 1.7 MPa (210% rise), 80.3 ± 0.2MPa (450% rise) and 14.9 ± 2.7 MPa (20% rise). The extent of improvement in the compressive strength of epoxy matrix composites is tantamount with literature in similar concentrations of nanoreinforcements [49]. Both MWCNTs and GPN are accounted for increase the
compressive strength of epoxy matrix composites when incorporated independently in constrained amounts, i.e. 0.1 wt.% [52].

The Compressive modulus likewise enhanced with the addition of MWCNTs and GPN in glass/epoxy individually at 0.2 wt.%, i.e. 18.4 ± 0.5 GPa and 14.5 ± 0.3 GPa in comparison to neat glass/epoxy, i.e. 10.2 ± 0.4 GPa demonstrating the rises of 74% and 39% respectively (Fig. 5b). The addition of both MWCNTs and GPN in the glass/epoxy at 0.1 wt.%, 0.2 wt.% and 0.3 wt.% loadings also resulted in increased compressive modulus values, i.e. 11.3 ± 0.3 GPa, 12.5 ± 0.2 GPa and 7.5 ± 0.6 GPa, which exhibits a rise of 10%, 22% rise and 26% fall, respectively. The incorporation of MWCNTs and GPN in glass/epoxy matrix has for the most part demonstrated an increase in the flexural modulus in small fractions; however, the pattern reverses after increasing the content of nanoreinforcements. For instance, in an examination compressive modulus increased with the addition of 0.1 wt.% MWCNTs in epoxy that diminished significantly by increasing the MWCNTs and GPN content up to 0.35 wt.% in the epoxy [7,34]. In another examination, compressive modulus is accounted for a similar pattern, i.e. sharp increment subsequent to adding ~0.3 wt.% MWCNTs and GPN followed by a decrease when their content was increased to 3.5 wt.% in epoxy matrix [8,42].

The maximum fracture strain in compressive testing increased when 0.2 wt.% MWCNTs and 0.2 wt% GPN were included in unidirectional glass/epoxy composite [0°]s, i.e. 3.8 ± 0.14%, (125.4% rise), 1.5 ± 0.11 (261.1% rise) when compared with neat glass/epoxy 0.28 ± 0.1% however GPN were affected adversely and diminished the flexural strain, i.e. 1.5 ± 0.15% (27.7% fall). The binary addition of MWCNTs and GPN in glass/epoxy composite at 0.2 wt.% and 0.1 wt.% each resulted in modest increase in flexural strain, 6.1 ± 0.18 (238.8% rise) and 7.2 ± 0.16 (300% rise). However, composites containing 0.2 wt.% MWCNTs and GPN each, demonstrated a significant rise, i.e. 5.6 ± 0.19 (211% rise), as shown in Fig. 5c. The addition of 0.4 wt.% MWCNTs has just demonstrated the increase in flexural strain [44] however the effect of GPN on the flexural strain isn't accessible in literature.
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The change in the fracture morphology of composites due to the incorporation of nanoreinforcements seems to be conceivable reason of increased toughening effect. The roughness of the fractured nanocomposites increased significantly (Fig. 6) in comparison to the fractured surface of neat glass/epoxy composite (Fig. 2a). Neat glass/epoxy composite showed a typical brittle fracture, i.e. smooth surface, while the rough surface was observed in composites due to the phenomena of shear yielding and deformation of epoxy between the reinforcements, as also observed elsewhere [13]. The fracture morphology also shows the appearance of crazing due to nanoreinforcements, which limits the propagation of cracks and increases the strength of the composites [10].
Figure 6 SEM images of fracture morphology in hybrid nanocomposites containing e-glass fibre/epoxy composite after the incorporation of 0.2wt.%MWCNTs and 0.2wt.%GPN. The images show rougher surfaces compared to the fractured surface of neat glass/epoxy composite in Fig. 2a.

The toughening mechanisms introduced by incorporating MWCNTs and GPN may be the pullout of nanoreinforcements, glass fibre reinforcement/ epoxy matrix debonding, crack deflection, and MWCNTs bridging. A model has been shown in Fig. 7 demonstrating the presence of MWCNTs and GPN in the glass/epoxy matrix, both individually and combined. The presence of MWCNTs and GPN is expected to initiate toughening mechanisms in glass/epoxy matrix, as shown in Fig. 8, where an advancing crack is deflected numerous times because of MWCNTs and GPN. In the wake of the crack, the crack bridging effect because of MWCNTs is also shown. Pulled out MWCNTs are also visible in the crack; this phenomenon happens takes place after the debonding of MWCNTs from the matrix material. All these toughening mechanisms are probably to be the possible conceivable reasons for an increase in the tensile and flexural strength of the composites especially the impact resistance. Fig. 9 a and b demonstrate the presence of pull out and bridging phenomena (arrowed) in nanocomposites, respectively, because of MWCNTs; the hauled out phenomena strengthens in composites containing increased amounts of MWCNTs, as can be witnessed and compared in composites containing 0.1wt.% and 0.3wt.% MWCNTs in Fig. 9c and d. The crack development is all by account limited because of the bridging of MWCNTs (Fig. 9b)
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Figure 7 Models of glass/epoxy matrix hybrid nanocomposites (a) without reinforcements and containing (b) MWCNTs (c) GPN and (d) MWCNTs and GPN.

Figure 8 Model of glass/epoxy matrix hybrid nanocomposite showing the propagation of crack through epoxy matrix and the interaction of crack with MWCNTs and GPN producing toughening mechanisms including crack deflection, MWCNTs bridging and MWCNTs pullout.
Figure 9. Toughening mechanisms introduced in hybrid nanocomposites (a) MWCNTs pullout (b) MWCNTs bridging; pullout phenomenon in composites containing (c) 0.1wt.% MWCNTs and 0.1wt.% GPN, and (d) 0.2wt.% MWCNTs and 0.2wt.% GPN.

4. CONCLUSIONS
The individual and combined effect of MWCNTs and GPN on the mechanical properties of various stacking sequences lay-up glass/epoxy [0°]s,[90°]s, 0°/90°]s and [45°/45°]s matrix hybrid nanocomposites has been investigated. To ensure uniform dispersion of nanoreinforcements and optimum bonding with the [0°]s glass/epoxy hybrid nanocomposite, MWCNTs and GPN were functionalized before their incorporation into epoxy resin. The contents of each of MWCNTs and GPN were increased from 0.1wt.% to 0.3wt.% while the glass/epoxy composites containing individual reinforcement were also fabricated for comparison. Mechanical testing of the composites containing 0.2wt.% MWCNTs/[0°]s glass/epoxy composite and 0.2wt.% GPN/[0°]s glass demonstrated an increase in tensile (28.4%) flexural (84%) and compressive (480%) strengths along with tensile (38%) flexural (64%) and Compressive(74%) moduli in comparison to neat glass/epoxy composite; fracture strain increased in both the tensile, flexural and compressive testing. Uniform dispersion of MWCNTs and GPN were observed in SEM images without the presence of their agglomerates. The increase in mechanical properties of the composites can be related with the uniform dispersion of nanofillers and their strong interfacial adhesion with the epoxy matrix.

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