Effect of hybrid mwcnts/graphene on tensile properties of reinforced unidirectional e-glass/epoxy composite

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Abstract: This paper presents an experimental investigation on the effect of tensile properties of unidirectional E-glass reinforced epoxy composites. Multi-walled carbon nanotubes (MWCNTs) and Graphene (Gnps) were dispersed as 0.1, 0.2, and 0.3 weight percentages in unidirectional E-glass fibre reinforced/epoxy composites were investigated. The result shows that 0.2 wt% MWCNTs/E-glass/epoxy resin composite gave better tensile properties when compared with E-glass/graphene/epoxy composite. The scanned electronic microscopic study for the fractured samples reveals a significant increase in the fibre-matrix interface adhesion and also decreases in fibre breakage, fiber pullout and debonding.

1. Introduction

Unidirectional E-glass-reinforced epoxy resin composites are often used in aerospace/defense applications. Because of their high specific mechanical properties, such as low density, light in weight, and stiffness, they are mainly used as structural components. Epoxy resin (LY556) is used as a matrix material in these composites. Graphene powder (Gnps) and multiwalled carbon nanotubes (MWCNTs) are new carbon materials that have recently been developed. Graphene has generated much more interest due to its high specific area and novel mechanical, electrical and thermal properties. Ravindranadh Bobbili et.al [1] combined E-glass-epoxy/MWCNTs (Multiwalled carbon nanotubes) composite and Epoxy/MWCNTs composite with four different reinforcements (0, 0.5, 1 and 1.5 wt%) of MWCNTs are dispersed into an epoxy resin and they were investigated for specific wear rate and friction coefficient can be reduced by addition of MWCNTs and also microscopic investigation of worn out sample fracture surface has revealed that fiber debonding happens when the stresses at the fiber matrix interface exceeds the interfacial strength, causing the fiber to debond from the matrix. M.S. Senthil Kumar et.al[2] has been observed that the various combinations of
nanomaterials and matrix materials which were used to fabricate the nanocomposite laminates with and without fiber and the impact of nano fillers were also studied. Md Musthak et.al [3] have studied the fabrication and characterization of E-glass fiber composites enhanced by carbon nanotubes with full factorial design experiments under tensile loading conditions. P.S. Shivakumar Gouda et.al[4] have fabricated and mechanical characterization of Graphene and Multi walled carbon nanotubes (MWCNTs) by direct mixing were employed to disperse these nanoparticles into a mono-component epoxy system and used as matrix for advanced composites with Carbon fiber reinforcement. M.B.A. Salam et.al[5] have fabricated and experimental characterization of carbon fibre/epoxy nanocomposites, with varying 0.1-0.4 wt.% carboxyl-functionalized multi-walled carbon nanotubes (MWCNTs-COOH) were incorporated in an epoxy system. Dilini Galpaya[6] et.al have fabricated and studied the mechanical characterization of graphene-polymer nanocomposites with suitable applications and advantages. Minh-Tai Le et.al[7] have fabricated and effect of a hybrid polymer nanocomposite containing epoxy/polyester blend resin and graphene nanoplatelets were used as fillers to enhance the properties of the epoxy matrix like tensile tests, dynamic mechanical analysis and thermo gravimetric analysis are performed on neat, 0.2 wt %, 0.5 wt %, 1 wt %, 1.5wt % and 2 wt % GNP-reinforced epoxy/polyester blend resin. S.H.Park et.al[8] have fabricated and experimental investigation on single-walled and multi-walled carbon nanotubes (CNTs) were functionalized with carboxyl groups and dispersed in a polymer containing an epoxide group. Mrinal Bhattacharya[9] have studied the processing of carbon nanotube, graphene and clay montmorillonite platelet as potential nanofillers to form nanocomposites and these nanofillers dispersion in the polymeric matrix was highlighted. Iman Taraghi et.al[10] combined of adding small amounts (< 1%) of Multi-walled carbon nanotubes (MWCNTs) addition on the tensile, flexural and impact properties of woven Kevlar fabric reinforced epoxy composites.

Based on the exhaustive literature review it was found that there is no report on the hybrid effect of MWCNTs and Gnp on mechanical properties of unidirectional E-glass reinforced polymer matrix composites. In this paper, we suggest a possible method to enhance the mechanical properties of MWCNT/Gnp/Uni directional E-glass reinforced epoxy polymer based composites. Standard tensile tests were performed on the nanocomposite samples were carried out as per ASTM 3039 standards and characterized using SEM.

2. Fabrication of Composite laminates

Unidirectional E-Glass fabric areal density 200 GSM were used as reinforcement. Epoxy resin LY5052 premixed and homogenized with hardener Aradur 5052 in the ratio of 10:1 by volume was used as matrix. All these polymer products were supplied by Arun fabrics Pvt Ltd (Bangalore, India). Fillers like multi walled carbon nanotubes (MWCNTs) with carbon purity more than 90%, carbon nanotubes and Graphene of diameter 5-20nm and surface area 330m²/gm supplied by United nanotech Innovations Private Limited (Bangalore, India). and preparation of test nano polymer specimen’s unidirectional E-glass (0°) reinforced epoxy was studied in this paper. For the present investigation dedicated 14 layered reinforced unidirectional E-glass fabric laminates (0°) with variation of weight percentage ratio of nano fillers ranging from 0.1, 0.2 and 0.3 of MWCNTs and Graphene powder were considered as shown in Figure.1.
0.1 Wt% of MWCNTs/Unidirectional E-glass/epoxy composite

0.2 Wt% of MWCNTs/Unidirectional E-glass/epoxy composite

0.3 wt% MWCNTs/Unidirectional E-glass/epoxy composite

0.1 wt% Graphene/Unidirectional E-glass/epoxy composite

0.2 wt% Graphene/Unidirectional E-glass/epoxy composite

0.3 wt% Graphene/Unidirectional E-glass/epoxy composite

0.1 Wt% of MWCNTs+Graphene / Unidirectional E-glass/epoxy composite

0.2 Wt% of MWCNT+Graphene/Unidirectional E-glass/epoxy composite

0.3 Wt% of MWCNTs+Graphene/Unidirectional E-glass/epoxy composite

**Figure 1.** Image showing the different variation of weight percentage ratio of nano fillers/unidirectional E-glass/epoxy specimens fabricated by Vacuum Layup technique

All the different variation of weight percentage ratio of nano fillers types of composite laminates considered for investigation were fabricated by vacuum bagging layup technique as shown in Figure.2. The layers of unidirectional E-glass each 0.25mm thickness alternatively placed and having 350 mm length and 350 mm width was put together to form a block with dimension of 350×350×3 mm for mechanical characterization. A uniform fibre weight fraction (Wf) of 50% was controlled for all types of laminates in fabrication. This approximates to a overall fibre volume fraction (Vf) of 50%, considering the fibre density as 2.55 g/cc and the matrix density as 1.17 g/cc. Then the vacuum bagging is applied to the mold with a vacuum pressure 2.525 MPa for uniform distribution of resin and also to remove the entrapped air. The composite is cured at room temperature and the post curing was done at 80 °C for 4hours. By using diamond cutter the specimens were cut from the plates and was polished with the help of polishing machine.
E-glass /0.1 wt% of MWCNTs
E-glass /0.2 wt% of MWCNTs
E-glass /0.3, 0.1, 0.2 and 0.3 wt% of Graphene
Vacuum pipe

Figure 2. Shows vacuum pump connections for the different laminates variation of weight percentage ratio of nano fillers/unidirectional glass/epoxy samples.

3. Experimental set up

Five specimens for each configuration were tested under different loads like tensile, compressive and flexural with a servo-hydraulic testing machine (INSTRON-8501) with a capacity of 100 KN. Each specimen was clamped by means of hydraulic wedge grips. The machine was equipped with a standard load cell and a crosshead displacement measuring device. During the mount phase of the specimen, the maximum applied load was controlled and set lower than 0.2 KN in order to avoid specimen damage. According to ASTM D3039 were subjected to loading with a stroke rate of 1.5 mm/min.

4. Experimental Results and Discussion

4.1 Static Mechanical Properties

Tensile test specimens were cut according to ASTM D3039 standard. The size of the specimen for unidirectional is 250×15×3 mm³, for transverse directional laminates is 175×25×3 mm³. Tensile tests were conducted on servo hydraulic universal testing machine with extensometer as shown in Figure 3.

| Composite specimen
| Strain gauge Extensometer

Figure 3. Servo Hydraulic Universal Testing Machine

Table 1 shows there is considerable ultimate tensile strength variation between the reinforced longitudinal(0°) glass/epoxy laminate and Transverse directional(90°) laminate with addition of variation of weight percentage ratio of 0.1, 0.2 and 0.3 of MWCNTs nano fillers. From these results it is concluded that the variation of average modulus elasticity was found to be 19.61 Gpa for 0.3 wt% of MWCNTs when compared with in 0.1 and 0.2 weight percentage of MWCNTs.
Table 1. Average tensile test results of Uni directional E-glass/epoxy reinforced with 0.1wt%, 0.2wt% and 0.3wt% of MWCNTs laminates

| Fibre orientation | Addition of nanofiller (%wt) | Load at Break (KN) | UTS (MPa) | Tensile stress at Yield (Offset 0.2%) (MPa) | Modulus (E-modulus) (GPa) |
|-------------------|-------------------------------|--------------------|----------|------------------------------------------|---------------------------|
| 0°                | 0.1                           | 43.48              | 611.12   | 242.14                                   | 19.27                     |
| Std. Dev          | 23.37                         | 323.98             | 132.28   | 10.32                                    | 6.91                      |
| 90°               | 9.42                          | 128.40             | 125.78   | 6.68                                     | 3.606                     |
| Std. Dev          | 4.96                          | 67.88              | 66.48    | 3.07                                     | 2.88                      |
| 0°                | 0.2                           | 46.01              | 628.40   | 243.39                                   | 19.47                     |
| Std. Dev          | 21.78                         | 333.29             | 132.26   | 10.82                                    | 7.26                      |
| 90°               | 10.29                         | 140.22             | 106.49   | 7.62                                     | 3.58                      |
| Std. Dev          | 5.21                          | 71.007             | 57.37    | 3.58                                     | 2.88                      |
| 0°                | 0.3                           | 43.00              | 586.88   | 241.05                                   | 19.61                     |
| Std. Dev          | 23.34                         | 318.25             | 132.61   | 10.23                                    | 7.77                      |
| 90°               | 10.19                         | 139.28             | 119.32   | 7.77                                     | 3.97                      |
| Std. Dev          | 5.47                          | 74.72              | 65.34    | 3.97                                     | 2.88                      |

The stress-strain curves of the unidirectional E-glass fabric/epoxy and transverse E-glass fabric/epoxy composites reinforced with three different percentage weight ratio of nano fillers sequences. The maximum stress is found for dedicated Longitudinal(0°) directional fabric laminate reinforced with addition of 0.2wt% MWCNTs arrangement as shown Figures from 4-6.
From Table 2 these results it is concluded that the variation of average modulus elasticity was found to be 18.88 Gpa for 0.1 wt% of graphene when compared with in 0.2 and 0.3 weight percentage of graphene.

Table 2. Average tensile test results of Unidirectional E-glass/epoxy reinforced with 0.1wt%, 0.2wt% and 0.3wt% of Graphene laminates

| Fibre orientation | Addition of nanofiller (%wt) | Load at Break (KN) | UTS (MPa) | Tensile stress at Yield (Offset 0.2 %) (MPa) | Young's modulus(E) (GPa) |
|-------------------|-------------------------------|--------------------|-----------|------------------------------------------|--------------------------|
| 0°                | 0.1                           | 38.45              | 512.67    | 245.89                                   | 18.86                    |
| Std. Dev          |                               | 20.41              | 272.86    | 134.15                                   | 10.086                   |
| 90°               |                               | 8.42               | 113.00    | 116.89                                   | 7.76                     |
| Std. Dev          |                               | 4.52               | 60.46     | 62.72                                    | 4.201                    |
| 0°                | 0.2                           | 39.19              | 526.40    | 220.24                                   | 22.24                    |
| Std. Dev          |                               | 20.07              | 268.89    | 120.27                                   | 11.66                    |
| 90°               |                               | 7.40               | 98.79     | 114.68                                   | 6.76                     |
| Std. Dev          |                               | 4.009              | 53.49     | 62.23                                    | 4.22                     |
| 0°                | 0.3                           | 27.31              | 419.538   | 334.98                                   | 11.55                    |
| Std. Dev          |                               | 13.95              | 216.55    | 172.87                                   | 5.98                     |
| 90°               |                               | 6.36               | 82.133    | 107.35                                   | 5.935                    |
| Std. Dev          |                               | 3.47               | 46.79     | 58.66                                    | 3.88                     |

The stress-strain curves of the unidirectional E-glass/epoxy and transverse fabric/epoxy composites reinforced with three different percentage weight ratio of nano fillers sequences. The maximum stress is found for dedicated longitudinal(0°) directional fabric laminate reinforced with addition of 0.2wt% graphene arrangement as shown in Figures 7-9.
From Table 3 these results it is concluded that the variation of average modulus elasticity was found to be 21.22 Gpa for 0.1wt% mixture of MWCNTs and graphene when compared with other mixture 0.2 and 0.3 weight percentage of MWCNTs and graphene.

Table 3. Average tensile test results of Uni directional E-glass/epoxy reinforced with mixture 0.1wt%,0.2wt% and 0.3wt% of MWCNTs and Graphene laminates

| Fibre orientation | Addition of nanofiller (%wt) | Load at Break (KN) | UTS (MPa) | Tensile stress at Yield (Offset 0.2 %) (MPa) | Young's modulus(E) (GPa) |
|-------------------|-----------------------------|-------------------|----------|--------------------------------------------|-------------------------|
| 0                 | 0.1                         | 36.81             | 561.92   | 198.21                                     | 11.44                   |
| Std. Dev          | 19.07                       | 301.18            | 107.06   | 6.11                                       |
| 90                | 6.81                        | 151.92            | 119.73   | 6.56                                       |
| Std. Dev          | 3.17                        | 66.39             | 63.21    | 3.39                                       |
| 0                 | 0.2                         | 44.83             | 610.49   | 241.60                                     | 13.94                   |
| Std. Dev          | 24.16                       | 329.16            | 133.24   | 7.85                                       |
| 90                | 11.26                       | 153.40            | 120.98   | 4.73                                       |
| Std. Dev          | 5.91                        | 80.5              | 70.32    | 2.44                                       |
| 0                 | 0.3                         | 29.93             | 456.89   | 178.47                                     | 9.30                    |
| Std. Dev          | 15.75                       | 248.63            | 96.85    | 4.98                                       |
| 90                | 9.93                        | 166.73            | 130.89   | 5.89                                       |
| Std. Dev          | 5.17                        | 89.75             | 71.10    | 3.21                                       |
The stress-strain curves of the unidirectional E-glass/epoxy and transverse E-glass/epoxy composites reinforced with three different mixture percentage weight ratio of nano fillers sequences. The maximum stress is found for dedicated longitudinal(0°) directional fabric laminate reinforced with addition of 0.1wt% mixture of MWCNTs and graphene arrangement with other mixture weight percentages as shown in Figures 10-12.

Morphology of fractured specimens in tensile test

The investigations carried on the variations show that there is fiber pullout of longitudinal glass fibers observed for all variations of composites rather than transverse. The delamination is also seen in all variations except 0.2wt% of MWCNTs, 0.2wt % of graphene and 0.1 wt% mixture of graphene and MWCNTs series of specimens. The resulting microscopic images are shown in Figure 13 and Figure 14. Thereafter there was catastrophic failure along the width of the specimen as the load bearing area decreased. The test coupons exhibit a fracture path that is angled across the thickness of the sample. This angle was determined to be approximately 15° to the vertical in Figure 13( a), (b).
and 13(c), (d). The delamination and fiber pullout are significantly lower in case of composite samples processed using without graphene filler and with MWCNTs as witnessed by the microscope images shown in the Figure 13(a)-(b)

![SEM images](image1.png)

(a)                                                                                        (b)

![SEM images](image2.png)

(c)                                                                                        (d)

Figure. 13 SEM images of the fracture surface in MWCNT/woven E-glass fabric/Epoxy nanocomposites containing: (a) 0.1 wt.%; (b), (c) 0.2 wt.%; (d) 0.3 wt% MWCNTs

5.Conclusions

The aim of this study was to evaluate the effect of MWCNTs and graphene on the tensile properties of unidirectional E-glass fabric-epoxy laminated composites. The laminated nano polymer composites with different longitudinal and transverse fibre orientation of E-glass epoxy resin reinforcement along with fillers of 0.2 wt% of Graphene and MWCNTs was successfully processed and developed with minimum percentage of voids using vacuum bagging lay-up method of fabrication at room temperature. The experiments on tensile tests conducted with the chosen variations of glass, with 0.2 wt% Graphene and MWCNTs, show that there will be an increase in modulus of elasticity by 20 to 25% and also sustain greater loads. The effect of Graphene and MWCNTs on delamination, fiber pullout, fiber breakage and voids are studied by using optical microscope, in which it is observed that the delamination and fiber breakage are minimal by adding Graphene and MWCNTs fillers. The fiber pullout is seen in tensile, is less fiber pullout.

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