

# Evaluation of Tensile Properties on Glass/Carbon Fiber Reinforced Hybrid Composite by Matrix Modification with Synergetic Impact of Hybrid Nanofillers

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## Abstract

Epoxy, known for its binding strength and stability, can become brittle due to low stiffness and toughness. To mitigate the inherent brittleness of epoxy, nanofillers are introduced to improve the properties of Fiber Reinforced Polymer (FRP) composite. In demand for tailored strength, hybrid fiber polymer composites unite robust fibers with a less resilient matrix, offering versatility in lightweight, cost-effective applications. This work aims to investigate the synergistic impact of Multi Walled Carbon Nanotubes (MWCNTs) and Halloysite Nanotubes (HNTs) hybrid nanofillers on epoxy-based glass/carbon composites. The epoxy is altered by introducing MWCNTs and HNTs nanofillers at concentrations of 1 wt.% (comprising 0.5 wt.% MWCNTs and 0.5 wt.% HNTs) and 2 wt.% (comprising 1 wt.% MWCNTs and 1 wt.% HNTs), respectively. Mechanical tests following ASTM standards reveal that the 1 wt.% MWCNT + 1 wt.% HNT hybrid composites exhibit a notable 23.7% increase in tensile strength, and 6.7% in hardness compared to unmodified counterparts. Fracture surface analysis indicates improved fiber-matrix interactions, toughening the matrix, and enhancing resin-fiber adhesion. MWCNTs contribute to pullout/bridging effects, while HNTs aid crack deflection, resulting in superior tensile properties for the hybrid nanocomposites.

**Keywords:** Hybrid glass/carbon fibers reinforced polymer; Multiwall carbon nanotubes (MWCNTs); Halloysite nanotubes (HNTs); Tensile properties, Hardness behaviour, Morphological Characteristics

## 1.Introduction

Hybridization in advanced fiber-reinforced composites combines diverse fibers within a matrix to tailor material properties, addressing challenges that single reinforcements can't. Combining glass and carbon fibers in polymers enhances strength, while retaining durability [1]. Hybrid composites offer benefits like weight reduction, fuel savings, increased cargo capacity, longer life cycles, and reduced maintenance costs, making them valuable in aerospace, marine, infrastructure, transportation, and energy sectors [2],[3],[4]. However, ensuring their durability in various environmental conditions, such as temperature fluctuations, moisture, oxidation, and mechanical loads, is a key challenge for applications like wind turbines, ship hulls, and airframes [5], [6]. Maintaining consistent material functionality in such conditions is crucial for their practical viability.

Throughout the year, there has been significant attention devoted to exploring the mechanical properties of hybrid composites comprising glass and carbon fibers within the realm of research [7], [8]. Zhang et al.[2] explored the epoxy composites with equal glass/carbon ratios, finding outer-layer carbon reinforcement improves flexural properties; alternating fibers maximizes compressive strength. Similarly, N V Pujar et al.[9] improved tensile strength and modulus by placing carbon fibers strategically in 20% carbon and 80% glass interlayer hybrid composites compared to glass fiber composite.. Jagannatha et al.[10] achieved a 65.24% increase in tensile strength with 60% carbon fiber compared to 60% glass. Dong et al.[11] explored the optimal design of hybrid

composite materials, leading to improved flexural behavior by strategically placing glass on top and carbon below. Additionally, Dong et al.[12] demonstrated higher flexural strength in composites with glass on the compressive side. The results showed a 40% enhancement in flexural properties compared to plain carbon composites and a 9% improvement over glass reinforcement. Similarly, Pandya et al.[13] observed a remarkable increase in tensile strength (35.40%) compared to glass fiber composites by strategically distributing glass fibers in surface layers and carbon fibers in the core. Jesthi and Nayak [14] investigated stacking sequences, including [GCGGC]<sub>s</sub>, [GGGCC]<sub>s</sub>, and [GGCCG]<sub>s</sub>, in hybrid composites tailored for marine applications, incorporating glass fiber in the top layer and carbon fiber. The results underscored that, when compared to other hybrid composites, the [GCGGC]<sub>s</sub> hybrid composite exhibited superior mechanical properties across the board. Recently, Weili Wu [15] found higher carbon content improved tensile strength in interlayer and intralayer hybrid composites. The effectiveness of hybrid composites relies on variables such as thickness, fabric structure, matrix toughness, stacking sequence, and fiber-matrix hybridization [8]. Despite glass and carbon fiber hybrid composites exhibiting significant strength, ongoing research aims to improve their susceptibility to out-of-plane loads and interlayer characteristics. Emphasis is placed on enhancing these composites through interlayer reinforcement, considering factors like filler type, arrangement, volume fraction, polymer type, and incorporating nanofillers via nanotechnology to optimize mechanical properties [16].

In recent years, there has been a research emphasis on improving epoxy properties through the incorporation of nanofillers, with carbon nanotubes (CNTs) and halloysite nanotubes (HNTs) demonstrating significant potential. Carbon nanotubes (CNTs) are prominent nanofillers addressing epoxy resin brittleness in fiber polymer composites. Their dense crosslinked structure, strong adhesion, customizable properties, high modulus, temperature resistance, and low creep make them valuable. With elastic moduli (0.3-1.0 TPa) and tensile strength (10-500 GPa) [17], [18], [19]. CNTs enhance mechanical properties at low loadings, employing toughening mechanisms like bridging and crack pinning. A recent study by M.D. Kiran et al. [20], achieved a 77% tensile strength increase with 0.75% MWCNT in epoxy-based carbon fiber compared to untreated composites. However, Dehrooyeh et al. [21] determined that the optimal concentration for peak performance in CNT/epoxy nanocomposites is 0.5 wt. %. This optimal condition was subsequently applied to boost the mechanical characteristics of glass fiber in CNT/epoxy nanocomposites, leading to substantial enhancements. In a similar vein, Lee et al. [22] grafted CNTs onto carbon fibers, noting a 22% improvement in tensile strength compared to untreated composites.

Halloysite nanotubes (HNTs), analogous to MWCNTs, offer a natural, cost-effective filler for composites, explored for biocompatibility, dispersion ease, and enhancing thermal, fire, and mechanical properties [23]. Deng et al. [24] noted an 11.5% flexural strength increase and a 32.8% rise in modulus with 10 wt.% HNTs in epoxy. Y. Ye et al. [25] improved impact strength in epoxy-based carbon composites by 25% with 2 wt.% HNTs. Md. Shahneel Sahrudin et al. [26] found 0.2 wt.% HNTs yielded superior flexural modulus and strength compared to CNT-epoxy composites. Fang Liu et al. [27] enhanced carbon fiber/epoxy composites with HNTs, improving compressive and flexural characteristics. M. D. Kiran et al. [28] demonstrated varying concentrations (0.1-0.75 wt. %) of HNTs in epoxy-based carbon fiber composites, resulting in notable enhancements in tensile strength and modulus, flexural strength, and modulus. A review by T.S. Gaaz et al. [29], emphasized the significant flexural strength improvement due to HNTs' high aspect ratios, suggesting promising enhancements, with the highest tensile strength increase occurring at a 7% HNT addition.

To avoid challenges, optimal composite filler content is usually lower, benefiting from enhanced nanofiller-matrix interactions. Researchers explore blending multiple fillers to strengthen polymer matrices, with simultaneous use of two nanofillers showing promise for synergistic effects in hybrid materials. Mohd Shahneel Sahrudin et al. [30] demonstrated synergies with low-weight nanofillers (HNTs + CNTs) in epoxy, increasing flexural strength (by around 46%) and modulus (by around 17%). Ling Jiang et al.[31] exhibited improved strength and toughness in polyurethane composites with silane-treated-HNT/acid-treated-MWCNT hybrid nanofillers. Limited literature exists on the synergistic effects of introducing hybrid nanofillers, specifically MWCNT and HNT, into FRP hybrid composites. In a recent examination led by P. Choudhari et al.[32], it was emphasized that there is a demand for hybrid nanofillers to boost the mechanical properties of FRP composites, predominantly in the context of long-term marine applications.

Despite existing research, there is limited exploration of the combined impact of structural rearrangement of hybrid fibers and hybrid nano-fillers on FRP composite mechanical performance. Most studies focus on these approaches in isolation. Therefore, exploring the combined effect of structural rearrangement of glass fiber/carbon fiber with the addition of (MWCNTs + HNTs) hybrid nano-fillers becomes a compelling area of interest. In the present work, symmetrical composites—plain glass fiber, carbon fiber, and hybrid composites with and without nanofillers—were fabricated using compression molding. The study aims to investigate the synergistic impacts of (MWCNTs + HNTs) hybrid nano-fillers on the tensile, hardness and morphological properties of glass/carbon-reinforced epoxy composites.

## 2. Materials and Methods

### 2.1. Materials used

In this study, primary reinforcements consisted of bi-directional woven fabric E-glass fibers (400 gsm) obtained from Valmiera Glass UK Ltd and bi-directional carbon fabric (3 k plain weave, 200 gsm) sourced from Marktech Composites Pvt. Ltd, Bangalore, India. The hybrid composite, composed of ten layers, preserved a weight distribution of 72% glass fibers to 28% carbon fibers. The overall weight ratio between the reinforcing fibers and epoxy polymer stood at 55:45. A mixture of LY-556 epoxy polymer and W152 LR hardener from CF Composites, Mumbai, India, was prepared in a proportion of 100:30. The detail material properties are given in the Table 1,

**Table 1: Physical and mechanical properties of carbon fiber, E-glass fiber, and Epoxy resin**

Properties	Carbon Fiber	E-glass Fiber	Epoxy
Density (g/cm <sup>3</sup> )	1.76	2.54	1.32
Tensile strength (MPa)	4900	3100-3800	83-93
Tensile modulus (GPa)	230	65.5-73.8	3.42
Elongation at break (%)	2.1	4.0	-

#### 2.1.1 Nanofillers

Nanofillers, namely MWCNTs and HNTs, served as secondary reinforcements. MWCNTs, obtained from AD-NANO Technologies Pvt. Ltd, Shivamogga, India feature dimensions according to the manufacturer's datasheet: outer diameter 10-30 nm, inner diameter 5-10 nm, density 2.1 g/cm<sup>3</sup>, surface area 110-350 m<sup>2</sup>/g, and length exceeding 10 μm, with a purity of 99%. Halloysite nanotubes (HNTs) from Sigma Aldrich Company, Bengaluru, India had diameters of 30-70 nm and lengths of 1–3 μm, exhibiting a tube-like structure with a density of 2.53 g/cm<sup>3</sup> and a surface area of 64 m<sup>2</sup>/g. Their high aspect ratio and low percolation characteristics make them suitable for reinforcing epoxy matrix composites.

## 2.2. Methods

### 2.2.1 Preparation of MWCNT/HNT modified epoxy

Overcoming challenges posed by increased viscosity in achieving uniform nanofiller dispersion in high-density epoxy, MWCNTs and HNTs powders were dried, combined in equal weights (1 wt.% with 0.5 wt.% MWCNT + 0.5 wt.% HNT, and 2 wt.% with 1 wt.% MWCNT + 1 wt.% HNT), and subjected to mechanical stirring, magnetic stirring, and sonication. The modified epoxy resin underwent preheating, additional sonication to break agglomerates, cooling, and gradual mixing with the hardener. Vacuum degassing was consistently applied to prevent bubble formation, ensuring a uniform distribution of nanofillers. The Figure 1 represent schematic diagram for preparation of the MWCNT/HNT blend in epoxy matrix by dispersion method.

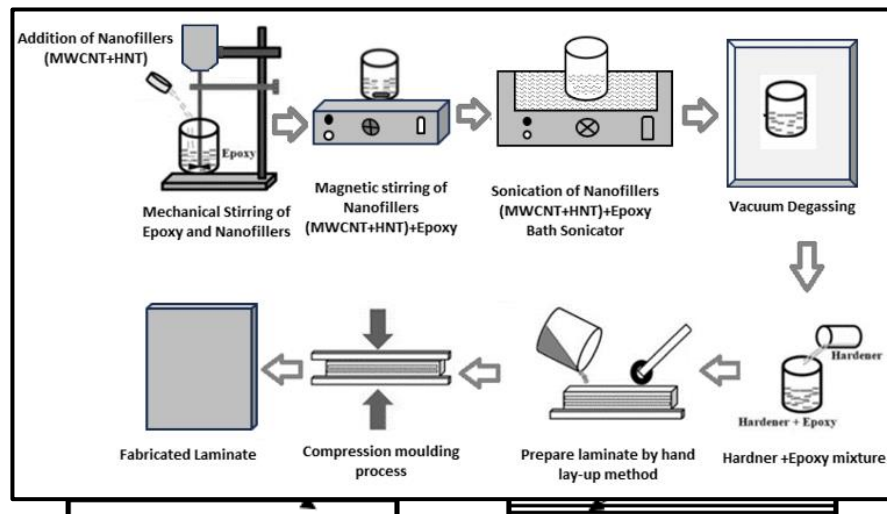


Figure: 1 Schematic diagram of the preparation of the MWCNT/HNT in glass fiber/carbon fiber epoxy composites by dispersion method

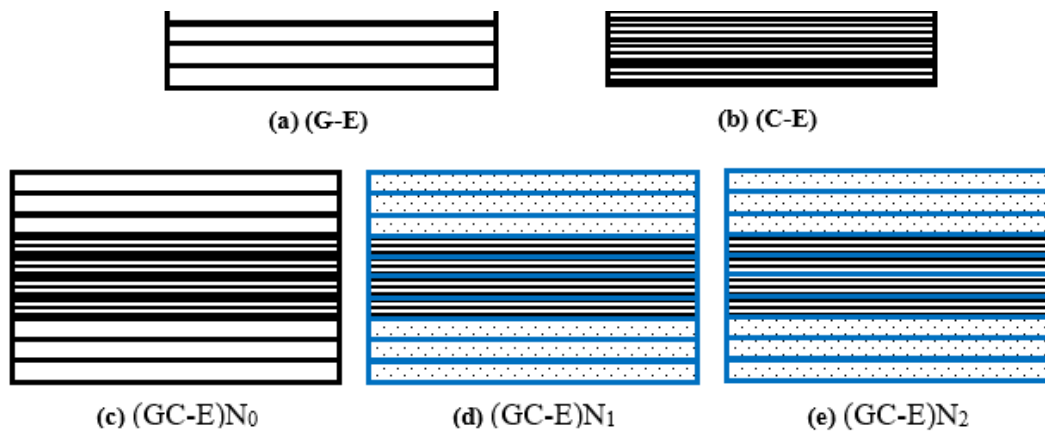


Figure: 2 Schematic diagram of composite laminates types with interply rearrangement

The glass/carbon fiber epoxy hybrid laminates were fabricated through compression molding. The mold, treated with a release agent, received epoxy applied to one side of the fiber mat, followed by layering another mat and compression. Hybrid composites with  $[G_3C_2]_s$  interply rearrangement comprised six glass layers and four carbon layers, while plain composites had ten layers of glass or carbon fiber. Five symmetrical composites were created as shown in Figure 2: plain glass fiber epoxy (G-E), carbon fiber epoxy (C-E), unmodified glass/carbon fiber epoxy (GC-E) $N_0$  with 0 wt.% (0 wt.% MWCNT + 0 wt.% HNT) nanofillers, modified (GC-E) $N_1$  with 1 wt.% (0.5 wt.% MWCNT + 0.5 wt.% HNT), and modified (GC-E) $N_2$  with 2 wt.% (1 wt.% MWCNT + 1 wt.% HNT). The resulting laminates were cut to ASTM standards for mechanical characterization using a water jet cutting machine.

### 2.3. Characterization of Composite

Mechanical properties play a pivotal role in product development as they determine the structural integrity and overall performance of the final product. To examine the combined influence of MWCNTs and HNTs hybrid nanofillers, the tensile, and hardness assessments were performed on an epoxy-based glass/carbon composite laminate. The tests were conducted according to established standards, and the ensuing results will be discussed in the following sections.

### 2.3.1 Tensile strength test

The tensile strength and modulus were determined according to ASTM D3039 standards, using specimens with dimensions of 250 mm (length), 25 mm (width), and 3 mm (thickness). Testing was performed with a Universal Testing Machine (Star Testing Systems, India, STS 248) at a crosshead speed of 5 mm/min and a 150 mm gauge length at room temperature. Three specimens of each composite material laminate were tested, and reported values are the averages.

### 2.3.2 Hardness Testing

The hardness of five distinct composite types was evaluated with a portable Barcol hardness tester, following the ASTM D2583 standard. This approach is commonly employed to evaluate the hardness of both unreinforced and reinforced rigid plastics. It involves pressing an indenter tip against the material, with the depth of indentation determined by the material's hardness. The resulting Barcol Hardness Numbers (BHN) were calculated for each composite, and average values were recorded. Barcol hardness test is inexpensive, portable, and highly user-friendly

### 2.3.3 Morphological Studies

The structures of specimens that experienced tensile failure were examined using Field Emission Scanning Electron Microscopy (FESEM) with a Nova Nano SEM 450. Furthermore, it was utilized to assess the dispersion of MWCNT and HNT nanofillers in the matrix, aimed at comprehending the failure mechanisms.

## 3. Results and discussions

### 3.1 Tensile testing result

Tensile tests serve as pivotal assessments for anticipating the practical applications of materials. They primarily focus on assessing the strength characteristics of composite materials. Figure 3 presents a stress-strain curve obtained from tensile testing for all five distinct composites, demonstrating the relationship between the applied load and the deformation of these materials.

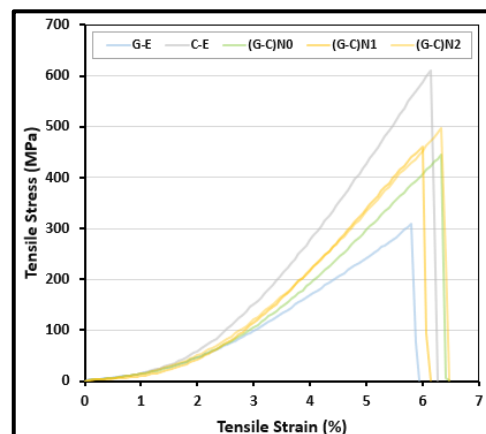


Figure: 3 Tensile stress-strain curves for different composites

Within the array of composites, the (C-E) composites, with 100% carbon fiber reinforcement, exhibited the most superior load-bearing characteristics. Figure 4 displays the tensile strength and tensile modulus of diverse composites. Carbon fiber epoxy composites (C-E) achieved peak tensile strength (526.4 MPa) and modulus (15.6 GPa), surpassing the glass fiber epoxy composites (G-E). The (G-C-E)<sub>N0</sub> hybrid composite, with [G<sub>3</sub>C<sub>2</sub>]<sub>s</sub> stacking, reached 399.9 MPa tensile strength and 11 GPa modulus. These values fall between the strengths of the (G-E) and (C-E) composites, which can be credited to the use of bidirectional fibers, ensuring consistent properties in both horizontal directions. Additionally, the high stiffness of the carbon fiber at the hybrid composite's core significantly contributes to this outcome. These findings align well with the enhancements in tensile strength and strain associated with the incorporation of internal carbon layers in hybrid composites.[14], [34], [35], [36] The introduction of a combination of MWCNTs and HNTs nanofillers, with varying weight percentages (0.5% and

1%), resulted in an improved loading response in the  $[G_3C_2]_S$  hybrid composite. The addition of 0.5 wt.% MWCNTs and 0.5 wt.% HNTs resulted in a significant improvement in the (GC-E) $N_0$  hybrid composite. It showed a 15.2% increase in tensile strength and a 1.7% increase in tensile strain, as shown in Figure 3. Increasing the nanofiller content to 1 wt.% MWCNTs and 1 wt.% HNTs in the (GC-E) $N_0$  hybrid composite resulted in a notable improvement. Tensile strength and strain increased by 23.7% and 6.8%, respectively, compared to the neat (GC-E) $N_0$  hybrid composite. This enhancement is attributed to amended nanofiller dispersion in the epoxy matrix, enhancing interfacial bonding, consistent with previous research [30]. Tensile modulus values for (GC-E) $N_1$  and (GC-E) $N_2$ , with two nanofillers, measured at 12.7 GPa and 11.7 GPa, respectively, marking a 15.1% and 6.1% increase compared to the unmodified (GC-E) $N_0$  composite. Figure 4 presents a bar graph showing the tensile strength and modulus of different composites. Notably, (GC-E) $N_2$  outperforms other hybrid composites in terms of tensile properties. The stress-strain curves reveal a linear and brittle response in all composite specimens up to the point of failure.

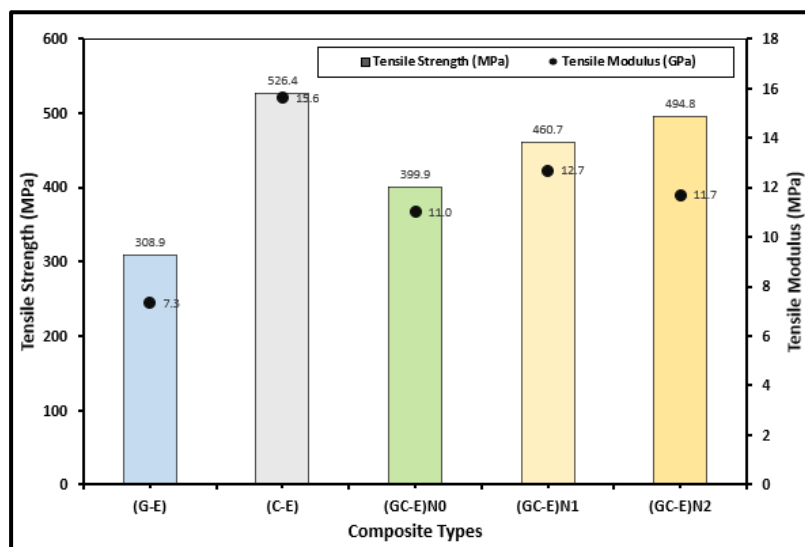
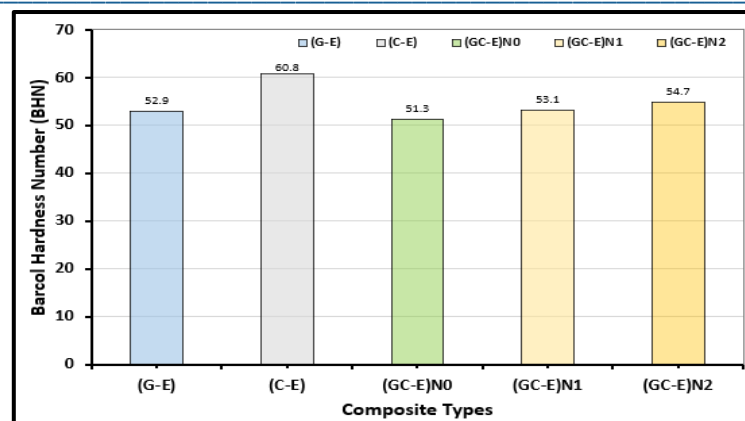


Figure: 4 Tensile strength and tensile modulus of different composites

In general, the Figure 3 demonstrates that all test specimens exhibited abrupt failure after reaching maximum tensile strength, characterized by a sudden drop. Glass fiber epoxy composites (G-E) exhibited greater tensile strain, while carbon fiber epoxy composites (C-E) displayed a reduced tensile strain in comparison to other composite materials. Notably, the addition of nanofillers improved the tensile strength and ultimate strain of (GC-E) $N_0$  hybrid composites, confirming their uniform dispersion without agglomeration during mixing. Uniform dispersion improved interlocking between (G/C) hybrid fibers and the matrix, effectively restricting crack development and enhancing load-carrying capacity during tensile loading.

### 3.2 Hardness testing result

The indentation hardness of the composite material was assessed using a Barcol hardness tester in accordance with the ASTM D2583 standard. In Figure 5, the hardness levels of various composites are depicted, including glass fiber epoxy (G-E), carbon fiber epoxy (C-E), and glass fiber/carbon hybrid composites with and without the incorporation of nanofillers. It's worth noting that the highest hardness value was observed in the carbon fiber epoxy composite (C-E), while the lowest value was found in the glass fiber epoxy composites (G-E).



**Figure: 5 Comparison of Hardness of the different composites**

Nevertheless, there is an interesting observation when considering the synergistic effect of two nanofillers in (G-E) $N_1$  and (G-E) $N_2$  composites. These combinations led to an improvement in hardness values by 3.4% and 6.7%, respectively, when compared to the (G-E) $N_0$  composite. This indicates that both the modified hybrid composites exhibit a higher level of hardness in comparison to the glass fiber epoxy (G-E). The enhanced hardness value can be attributed to the well-established interfacial interactions of both the nanofillers HNTs and MWCNTs.

### 3.3 Morphological result

The FESEM images of fabricated composite laminates under a tensile behaviour gives the general information about the fracture surfaces of hybrid composites. From FESEM analyses from research, excellent dispersion of nanofillers (MWCNTs and HNTs) with no sign of agglomeration was observed in the both the modified hybrid composites specimens that is (GC-E) $N_1$  and (GC-E) $N_2$ .



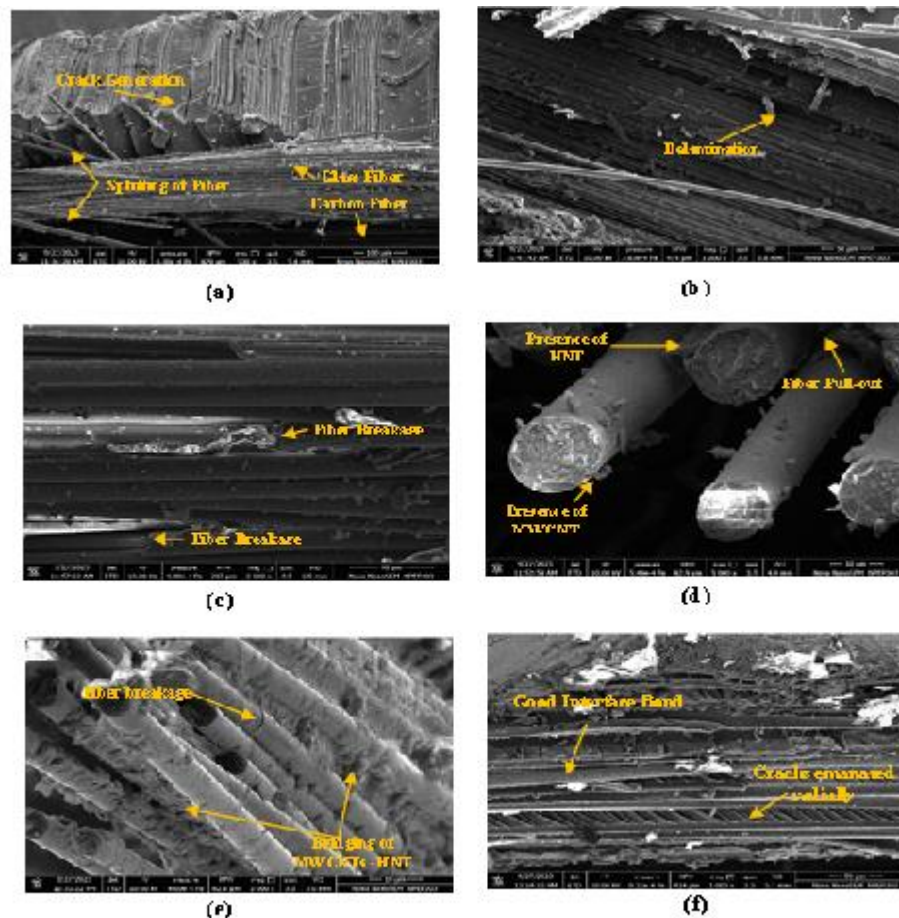


Figure 6 FESEM images (a)-(b) for (GC-E) $N_0$ , (c)-(d) for (GC-E) $N_1$  and, (e)-(f) for (GC-E) $N_2$  at fracture surface of tensile test specimen

Figures 6 (a) and (b) confirm the presence of both glass fiber and carbon fiber in the hybrid composites (GC-E) $N_0$ , showcasing strong matrix adhesion. The failure mechanism is readily noticeable through the development of cracks in the epoxy matrix, splitting and delamination of Fibers, which reduces the interfacial bond with the polymer matrix. Conversely, introducing the synergistic effect at loading content of 0.5 wt.% and 1 wt.% each reinforces the matrix and establishes a robust bond between the matrix and the (GC) hybrid fibers. The FESEM images of (GC-E) $N_2$  composite in the Figure 6 (e), (f) also showcases the crack-bridging and crack-deflection effects of MWCNT and HNT nanofillers, offering insights into the toughening mechanism. MWCNT fillers exhibit strong adhesion to the fibers, enhancing fiber-resin bonding. Furthermore, HNT fillers in the epoxy resin play a role in managing micro-crack development within the matrix by forming bridges with nanotubes. The enhancement in strength and modulus of the composites is attributed to the strong interface bond and matrix reinforcement. However, Figure 6 (c) and (d) show FESEM images of the (GC-E) $N_1$  composite, indicating the occurrence of deficiencies such as fiber breakage and fiber pull-out. Moreover, also provide the confirmation of the presence of MWCNT and HNT nanofillers within the polymer matrix

#### 4. Conclusions

In summary, the current study was focused on exploring the synergistic effects of (MWCNTs and HNTs) hybrid nanofillers in epoxy-based glass/carbon composites. The incorporation of nanofillers at low loading content in epoxy demonstrated enhanced mechanical properties, including improved tensile strength, and laminate hardness. The findings highlight the synergistic effects of fiber variations, matrix-nanofiller bonding, and hybridization, emphasizing the potential for advanced composite materials with superior performance.



- The hybrid composite (GC-E) $N_0$ , which incorporates a stacking sequence of  $[G_3C_2]_s$ , exhibited excellent tensile characteristics, boasting a tensile strength of 399.9 MPa and a tensile modulus of 11 GPa. These values fall within the range of tensile strengths observed in the (G-E) and (C-E) composites.
- Both MWCNTs and HNTs had a positive impact on the tensile strength and tensile modulus of the (GC-E) $N_0$  hybrid composite. Notably, when 1 wt.% of MWCNTs and 1 wt.% of HNTs were introduced, the (GC-E) $N_0$  hybrid composite, referred to as (GC-E) $N_2$ , exhibited remarkable improvements. These enhancements included a 23.7% increase in tensile strength, a 6.8% rise in tensile strain, and a 15.6% increase in tensile modulus in comparison to the pristine (GC-E) $N_0$  hybrid composite specimen.
- The Barcol hardness test proves valuable in evaluating material indentation hardness, indicating an increase in hardness values with higher dispersions of MWCNTs and HNTs. Specifically, in the nanofillers-modified hybrid composites (GC-E) $N_1$  and (GC-E) $N_2$ , Barcol Hardness Numbers demonstrated improvements of 3.6% and 6.7%, respectively, compared to (GC-E) $N_0$ .
- FESEM analysis reveals fiber breakage, effective interface bonding, and epoxy matrix toughening as key factors in composite reinforcement, leading to enhanced properties and novel high-performance materials with MWCNTs and HNTs. Common failure modes include fiber pull-out, breakage, and matrix damage in tensile behavior.
- Future research should delve into the performance of the hybrid nanocomposite under diverse and challenging conditions, examining its response to varying load levels. This innovative approach with advanced hybrid composites holds significant potential for exploration and application in diverse industries such as aerospace, automobile, and marine applications.

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#### Conflicts of Interest:

The authors declare that there is no conflict of interest.

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