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EXPERIMENTAL EVALUATION AND NUMERICAL VALIDATION OF BENDING BEHAVIOURS OF GLASS/CARBON REINFORCED EPOXY HYBRID COMPOSITES THROUGH HYBRID NANOFILLERS

Praful Choudhari[1], Vivek Kulkarni[2], Sanjeevakumar Khandal[3] [1,2,3]Department of Mechanical Engineering, Sanjay Ghodawat University, Atigre, Kolhapur-416118, Maharashtra, India

Abstract
In addressing the brittleness and limited stiffness/toughness of epoxy in long-term applications such as marine, aerospace, and energy sectors, a novel approach integrates two nanofillers into an epoxy matrix to explore synergistic effects. This study explores the impact of MWCNTs and HNTs hybrid nanofillers on the flexural characteristics of glass/carbon/epoxy-based composites. Three hybrid composites with equal-weight proportions of two nanofillers were formulated: no fillers, 0.5 wt. % MWCNT + 0.5 wt. % HNT, and 1 wt. % MWCNT + 1 wt. % HNT. Analysis of flexural strength reveals that the combination of 1 wt. % MWCNT and 1 wt. % HNT produces the most significant positive hybrid effect, exhibiting substantial enhancements in flexural strength (684 MPa) compared to other compositions. This research underscores the crucial role of synergistic nanofiller interactions in optimizing glass/carbon fiber hybrid composites for diverse industry applications. To confirm the flexural strength under defined experimental loads, 3D solid models of the composite specimens were introduced into Finite Element Analysis (FEA) software for validation. A comparison between experimental and predicted FEA results demonstrates a good agreement, reinforcing the validity of the findings.

Keywords: Hybrid glass/carbon fibers reinforced polymer; Multiwall carbon nanotubes (MWCNTs); Halloysite nanotubes (HNTs); Flexural properties; Finite element analysis

1.Introduction
Hybrid fiber-reinforced composites, uniting glass and carbon fibers in polymers, bolster strength and durability [1], yielding weight reduction and cost savings across aerospace, marine, and other sectors. This results in diminished fuel consumption, driving increased demand for this innovative technology [2], [3], [4]. Ensuring the robustness of applications such as wind turbines, ship hulls, and airframes in various environmental conditions remains a significant challenge. Additionally, addressing their vulnerability to impact and bending events represents an ongoing issue that has yet to be resolved. Research has extensively explored the impact and bending properties of glass/carbon (G/C) fiber hybrid composites [5], [6], [7]. Carbon fibers offer greater strength and stiffness with low density but lack damage tolerance due to brittleness. In contrast, glass fibers are cost-effective with lower mechanical properties but superior impact resistance. A studies by Zhang et al. [2] found that outer-layer carbon reinforcement in glass/carbon (G/C) fiber hybrid composites improves flexural properties, while Dong et al. [8] optimized flexural strength with glass on top surface and carbon at below in glass/carbon (G/C) fiber hybrid composites. Jesthi and Nayak [9] improved marine hybrid composites with [GCGGC]s stacking, enhancing flexural and impact properties. Another study of Jesthi and Nayak [10], [GGGCC]s showed better flexural and impact extension but [CCGGG]s had higher toughness and strength. Investigation into G/C hybrid composites for structural applications like automobiles and construction reveals enhanced toughness, impact resistance, and energy absorption. Hybrid composite performance hinges on various factors, including thickness, fabric structure, matrix toughness, stacking sequence, and fiber-matrix hybridization.

Despite glass and carbon fibers hybrid composites displaying impressive impact and flexural properties, their lateral and interlayer characteristics are deficient, making them prone to fracture under out-of-plane loads. Researchers seek improvement through interlayer reinforcement, considering factors like reinforcement type, arrangement, volume fraction, polymer type, and fillers. Nanofillers introduced through nanotechnology present an affordable and safe solution [11], optimizing mechanical properties, particularly bending and toughness. Ongoing research concentrates on developing polymer composites with diverse fillers for stronger, safer, and cost-effective structures.

In recent years, research has concentrated on augmenting epoxy properties by integrating nanofillers, with carbon nanotubes and halloysite nanotubes showing promise. Carbon nanotubes, particularly prized among nanofillers, effectively address brittleness and cracks in epoxy resin, commonly used as a matrix in fiber polymer composites. Valued for strong adhesion, customizable properties, high modulus, elevated temperature resistance, and low creep, even low levels of CNT incorporation enhance mechanical properties [12], [13], [14], bolstering toughening mechanisms [15] in fiber-reinforced polymer composite interfaces. In an experimental investigation, Rathore et al. [16] improved glass fiber-reinforced epoxy composites with 0.1 % MWCNTs, achieving an 11.5 % boost in flexural strength and a 32.8 % increase in flexural modulus. Naturally occurring halloysite nanotubes (HNTs), resembling MWCNTs, prove a cost-effective and promising particle filler. A recent research of , M. D. Kiran et al.[17] showed enhanced flexural strength and modulus in carbon fabric-reinforced epoxy composites at low wt.% of HNTs. Deng et al.[18] observed significant mechanical improvements in epoxy with 10 wt.% HNTs, attaining an 11.5 % elevation in flexural strength and a 32.8 % rise in flexural modulus. Studies by Md. Shahneel Sahrudin et al.[19] introducing HNTs and MWCNTs to epoxy at various levels, 0.2 wt.% HNTs yielded superior flexural modulus and strength, surpassing lower-concentration CNT-epoxy composites. Similarly, Fang Liu et al.[20] improved carbon fiber/epoxy composites using halloysite nanotubes, enhancing compressive and flexural characteristics. In a review by T.S. Gaaz et al. [21], the notable enhancement in flexural strength attributed to the high aspect ratios of HNTs was underscored, indicating promising improvements.

The previous literature study reflects that in order to optimize composite strength, filler reinforcement is typically kept at 1 wt.% or lower to prevent dispersion challenges and stress concentrations. Enhanced properties result from improved nanofiller-matrix interactions, driven by their high surface-to-volume ratio. Researchers explore blending multiple fillers to strengthen polymer matrices and create superior composites, with simultaneous use of two nanofillers showing promise for hybrid materials and potential synergistic effects, expanding the range of novel structural composites. Mohd Shahneel Sahrudin et al.[22] demonstrated synergistic effects of low-weight nanofillers (HNTs + CNTs) in epoxy, increasing flexural strength (up to 46 %), and modulus (up to 17 %). Similarly, Ling Jiang et al.[23] showed improved strength and toughness in polyurethane composites with silane-treated-HNT/acid-treated-MWCNT hybrid nanofillers. There is a scarcity of research in the existing literature concerning the synergistic effects of introducing hybrid nanofillers, specifically MWCNT and HNT, into fiber-reinforced hybrid composites.

In spite of the available research, the interaction between structural rearrangement in hybrid fibers and the synergistic effects of (MWCNT+ HNT) hybrid nanofillers approaches on the bending strength performance of fiber-reinforced composites has been inadequately explored in the literature. Many researchers have separately applied these approaches, making it crucial to investigate their combined influence for a comprehensive understanding of their effect on flexural properties. In the current investigation, the fabrication of symmetrical composite with plain glass fiber $[G_5]_S$, carbon fiber $[C_5]_S$, and hybrid composites having interply rearrangement $[G_3C_2]_S$ with and without nanofillers were done by using compression moulding techniques. The symmetrical hybrid composites were configured with carbon fibers on the inner side and external glass fibers on the outer side. The primary focus was on developing a superior and reliable hybrid composite suitable for various long-term applications. Therefore, the synergistic influence of MWCNTs and HNTs hybrid nanofillers on the flexural characteristics of glass/carbon/epoxy-based composites have been evaluated experimentally and validated numerically.

2. Experimental Work
2.1. Materials
In the present investigation, the bi-directional woven fabric E-glass fiber of 400 gsm were supplied by Valmiera Glass UK Ltd, and the bi-directional carbon fabric of 200 gsm, 3 k plain waving were supplied by Marktech Composites Pvt. Ltd, Bangalore, India. The hybrid composite consists of a 72:28 weight ratio of glass to carbon fibers, with both the plain glass/carbon fiber composites and hybrid composites comprising ten fiber layers. The overall weight ratio between reinforcement fibers and epoxy polymer is 55:45. LY-556 epoxy polymer and W152 LR hardener, obtained from CF Composites, Mumbai, India, are mixed in a 100:30 ratio. The properties of the reinforcement and matrix materials are detailed in Table 1.

2.1.1 Nanofillers.
The use of MWCNTs and HNTs as secondary reinforcements are illustrated in Figure 1 (c), and (d) respectively. The MWCNTs, synthesized through chemical vapor deposition by AD-NANO Technologies Pvt. Ltd, Shivamogga, India, were employed. These MWCNTs have an outer diameter of 10 to 30 nm, inner diameter of 5 to 10 nm, density of 2.1 g/cm^3, surface area of 110 to 350 m^2/g, and a length exceeding 10 µm with 99 % purity. Halloysite nanotubes (HNT), obtained from Sigma Aldrich Company, Bengaluru, India, have diameters of 30 to 70 nm, lengths of 1–3 µm, a tube-like structure, density of 2.53 g/cm^3, and surface area of 64 m^2/g, making them suitable for reinforcing epoxy matrix composites due to their elevated aspect ratio and minimal percolation characteristics. The elastic modulus for HNTs was determined to be approximately130-140 GPa [24],[25].

Table: 1 Mechanical properties of fibers, matrix, and nanofillers

Parameters	Carbon Fiber	E-Glass Fiber	Epoxy	MWCNTs	HNTs
Density (g/cm^3)	1.76	2.54	1.32	2.1	2.53
Tensile strength (MPa)	4900	3100-3800	83-93	10–500	10.8
Tensile modulus (GPa)	230	65.5-73.8	3.42	300-1000	140
Elongation at break (%)	2.1	4.0	-		-
Poisson's Ratio	0.30	0.22	0.35	0.26	-

2.2. Methods
2.2.1 Preparation of MWCNT/HNT modified epoxy
Achieving uniform dispersion of nanofillers in high-density epoxy is challenging due to increased viscosity. To address this, nanofiller powders (MWCNTs and HNTs) were dried, combined in equal weights such as 1 wt.% (0.5 wt. % MWCNT + 0.5 wt. % HNT), and 2 wt.% (1 wt. % MWCNT + 1 wt. % HNT), mechanically stirred, magnetically stirred, and sonicated. The modified epoxy resin was preheated, further sonicated to break agglomerates, cooled, and gradually mixed with hardener. Vacuum degassing was employed to prevent bubble formation during the entire process, ensuring even nanofiller distribution.
2.2.2. Preparation of glass/carbon modified epoxy-based laminates
The glass/carbon fiber epoxy laminates were made via compression molding. A release agent was applied to the mould, and epoxy was spread on one side of the fiber mat, layered with another mat, and compressed. Six layers of glass and four layers of carbon were used in hybrid composites, while plain composites had ten layers of either glass or carbon. The stacking sequence of fabricated composite laminates. The Figure 1 illustrates five symmetrical composites: (G-E) for the plain glass fiber epoxy composite, (C-E) for carbon fiber epoxy composite (C-E), (GC-E)N_0 for unmodified glass

fiber/carbon fiber epoxy composite with 0 wt.% of (0 wt.% MWCNT + 0 wt.% HNT) nanofillers, (GC-E)N_1 for modified glass fiber/carbon fiber epoxy composite with 1 wt.% of (0.5 wt. % MWCNT + 0.5 wt. % HNT) nanofillers, and (GC-E)N_2 modified glass fiber/carbon fiber epoxy composite with 2 wt.% of (1 wt. % MWCNT + 1 wt. % HNT) nanofillers. The acquired composite laminates were trimmed to meet the required dimensions as specified by ASTM standards, utilizing a water jet cutting machine.

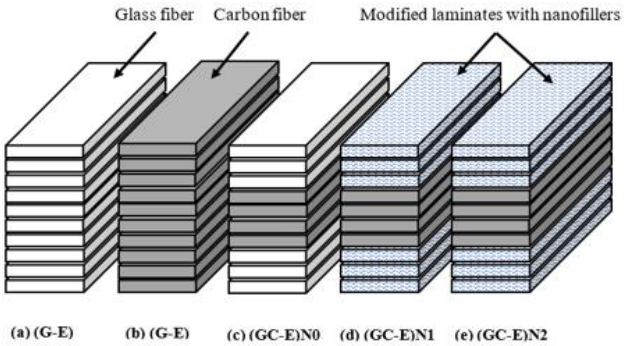

Figure: 1 Schematic diagram of composite laminates types with interply rearrangement

2.3. Characterization of Composite

To explore the synergistic effect of hybrid nanofillers, MWCNTs, and HNTs on the flexural properties of glass/carbon/epoxy-based composite laminates, the study adhered to ASTM standards, and the findings are elaborated in the subsequent sections.

2.3.1 Flexural Testing

All five composite types underwent ASTM D7264 flexural testing with 70 mm x 13 mm x 3 mm specimens and 60 mm support spans, as shown in Figure 2 (a), and (b) respectively. The Universal Testing Machine (TINIUS OLSEN H25KT) conducted flexural tests with a 60 mm span, 2 mm/min crosshead speed. Three specimens per composite were tested, and the average result was recorded.

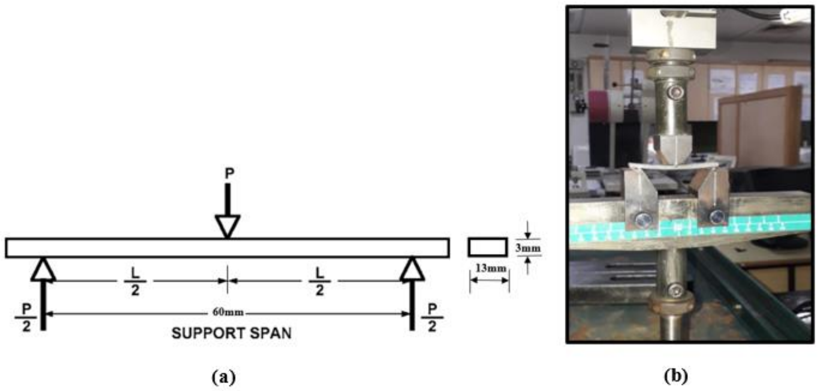

Figure: 2 (a) Loading diagram of three-point bending test arrangement, and (b) Flexural test specimens during the test

2.3.2 Numerical simulation

An elastic behavior and flexural property prediction study of hybrid composites were carried out using ANSYS software through finite element analysis. Experimental methods are effective for obtaining most properties, but studying the elastic-plastic transition in composites is challenging, necessitating the use of finite element modelling. Theoretical predictions serve to validate experimental results and accelerate the process of determining optimal filler quantities to maximize composite properties. Finite

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element models were created based on ASTM D7264 standards for the test coupons and setup dimensions, as illustrated in Figure 3.

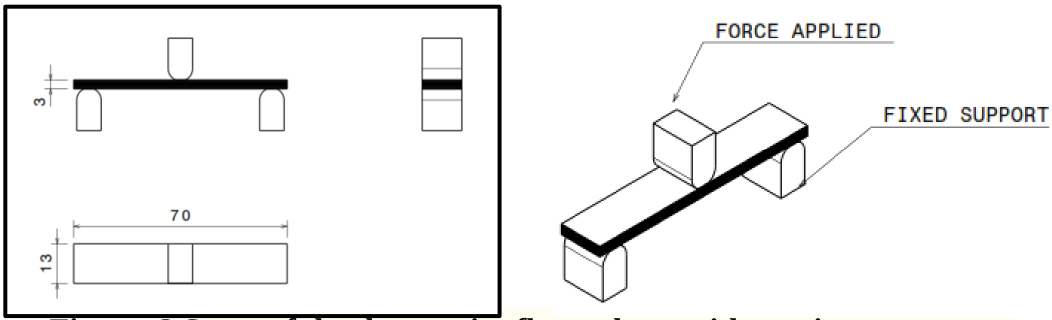

Figure: 3 Setup of the three-point flexural test with specimen geometry

The study involved creating a finite element model using the tetrahedron meshing method to simulate a composite plate with applied loads and constraints. A mesh size of 1 mm was chosen based on a convergence study, and the thickness of the finite element was set equal to the ply thickness. Each node had translations in x, y, and z directions. The model comprised 23,887 elements and 51,850 nodes. Failure modes, including elastic behavior, were analyzed. Meshed models in Figure 4 underwent static analysis to determine flexural responses, utilizing composite mechanical properties from Table 1.

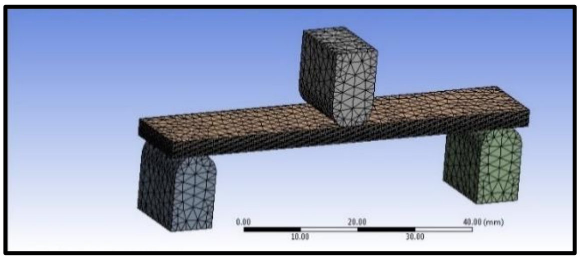

Figure: 4 Meshed model of the flexural test specimen

3. Results and discussion

3.1 Flexural Behaviour

The condition of the specimens before and after the flexural test is depicted in Figure 6. The provided data in Figure 5 (a) and (b) presents the results of 3-point bending flexural tests conducted on a range of composite specimens. These specimens include pure glass fiber epoxy (G-E), pure carbon fiber epoxy (C-E), and hybrid glass fiber/carbon fiber epoxy (GC-E) composites, both with and without the inclusion of nanofillers. The Figure 5 also include the average values for flexural strength, flexural modulus, and flexural extension for each of these composite types. The results of the flexural tests revealed notable variations among the composite specimens. The (G-E) composite specimens displayed the lowest flexural strength at 405 MPa and the lowest flexural modulus at 17.78 GPa. In contrast, the (C-E) composite specimens exhibited the highest flexural strength at 887.70 MPa and the highest flexural modulus at 74.13 GPa. Interestingly, the (G-E) composites demonstrated the greatest displacements before reaching failure compared to all other specimens. It was observed that a significant enhancement in flexural strength occurred when transitioning from (G-E) specimens, with a strength of 405 MPa, to (GC-E)N_0 hybrid specimens, with a strength of 656.30 MPa. This marked an improvement of nearly 62 %, as depicted in Figure 5 (a). Similarly, the flexural modulus of (GC-E)N_0 hybrid specimens increased to 36.30 GPa from 17.78 GPa, a remarkable boost of 103.93 % when compared to the plain (G-E) composites. The improved flexural characteristics of the (GC-E)N_0 hybrid composites were attained by a deliberate arrangement that positioned the glass fiber layers in the outer section, where greater ductility is present. This strategy resulted in a similar observation as reported for the interply rearrangement of glass fiber and carbon fiber layers in hybrid composites [9], [26].

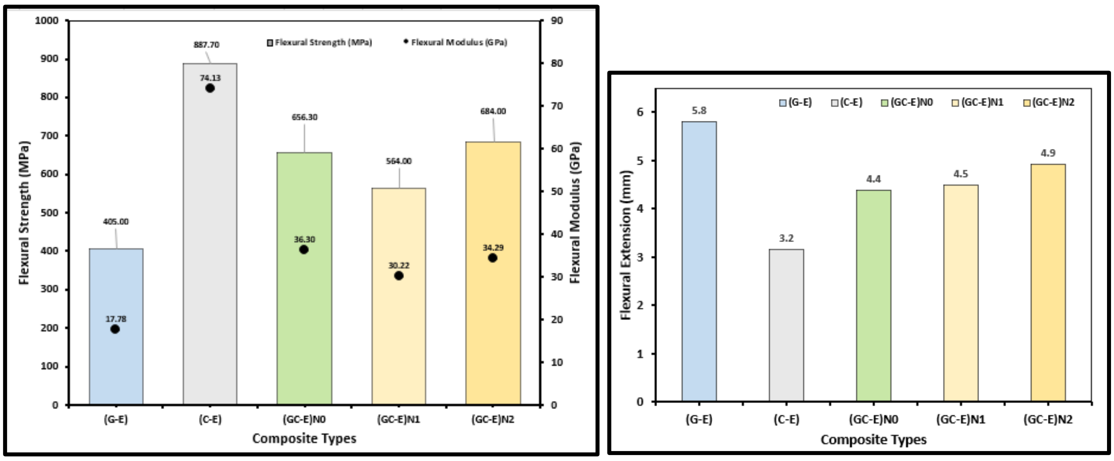

Figure: 5 (a) Flexural strength and flexural modulus, and (b) flexural extension of different composites

As depicted in Figure 5 (a), the incorporation of nanofillers in composite materials plays a vital role. The incorporation of MWCNTs and HNTs nano-fillers, particularly at 1 wt.% each, significantly outperforms the (GC-E)N0 hybrid compo-site. It results in a 4.22 % enhancement in flexural strength, and 11.37% higher flexural strain but decrease in modulus by 5.5%. Additionally, the flexural modulus of (GC-E)N_2 improved by 13.58 % compared to (GC-E)N1 hybrids. Conversely, Incorporation of 0.5 wt. % each for MWCNTs and HNTs nanofillers in (GC-E)N0, experiences a flexural strength and modulus decreased by 14 % and 16.7% but increase in flexural strain by 2.27 %. However, a positive trend emerged with a 2.27 % increase in flexural extension compared to unmodified (GC-E)N_0 hybrids, emphasizing nanofillers influence on strength and ductility in epoxy composites. The Figure 5 (b) illustrates the flexural extension verses the different composites types. The plain (G-E) composite had the highest flexural strain and the plain (C-E) composite recorded low strain value due to its brittleness nature. This improvement in flexural extension was consistent across all the modified and unmodified hybrid composites, as illustrated in Figure 5 (b). These findings underscore the significance of nanofillers in epoxy composites, demonstrating their capacity to influence both strength and ductility characteristics.

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3.2 FEA simulation results of flexural strength

The validation of the FEA model involved a comparison of the flexural stress-extension characteristics between the experimental and simulated flexural tests conducted on the composite laminates. The stacking sequence $[G_3C_2]_S$ composite type, namely (G-E)N_0, were considered for the further analysis under the synergistic effect of hybrid nanofillers. Similarly, the experimental efforts taken by Jesthi et al.[27],[9],[28], through interply sequence of fibers in the hybrid composite on the different combination of six glass fibers and four carbon fiber. The study resulted about the minimum flexural strength value for $[G_3C_2]_S$ as compared to other combinations.

Figure 6 illustrates the flexural stress versus extension curves for experimental and FEA simulated results in diagrams (a) (GC-E)N_0, (b) (GC-E)N_1, and (GC-E)N_2 test specimens. The numerical results depicting stress-deformation behavior closely align with the trends observed in the experimental data. Furthermore, the flexural stress obtained through simulation slightly exceeded that of the experimental method. This variance may be attributed to the simulation's assumption of homogeneous laminates without voids or crack formation, whereas actual laminates exhibit non-homogeneity, featuring initial voids and the development of fiber-matrix cracks during experimental flexural tests. The FEA simulation plots for flexural strength. and total deformation of the (a) (GC-E)N_0, (b) (GC-E)N_1, and (GC-E)N_2 test specimen of the generated numerical models are exemplified in Figure 7.

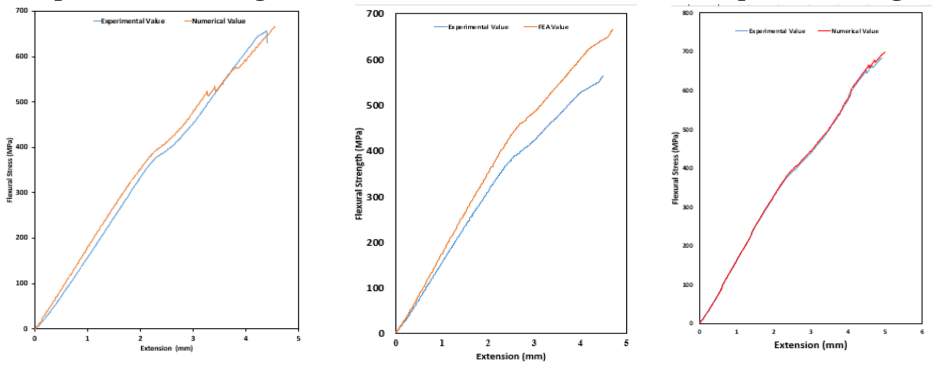

Figure: 6 Flexural stress verses extension curves for the experimental and numerical value of (a) (GC-E)N_0, (b) (GC-E)N_1, and (GC-E)N_2 test specimen

Figure 7 displays the experimental and FEA simulated flexural stress verses extension curve diagrams (a) (GC-E)N_0, (b) (GC-E)N_1, and (GC-E)N_2 test specimen. The numerical stress-deformation behavior closely mirrored the experimental results, with the simulated flexural stress slightly exceeding that observed in experiments. This discrepancy may be attributed to the simulation treating laminates as homogeneous, without voids or crack formation, whereas real-time conditions involve non-homogeneous laminates with initial voids and fiber-matrix cracks during experimental flexural tests. The FEA simulation plots for flexural strength. and total deformation of the (a) (GC-E)N_0, (b) (GC-E)N_1, and (GC-E)N_2 test specimen of the developed numerical models are illustrated in Figure 8.

Table: 2 Experimental and numerical results of flexural test for hybrid composite specimen

Specimen Types	Flexural Strength (MPa)			Extension (mm)		
	Expermental Value	Numerical Value	% Error	Expermental Value	Numerical Value	% Error
$[G_3C_2]S$ -((GC-E)N0)	656.3	664.76	**1.27**	4.39	4.55	**3.52**
$[G_3C_2]S$- ((GC-E)N1)	564	665.67	**15.27**	4.5	4.71	**4.46**
$[G_3C_2]S$ -((GC-E)N2)	684	720.77	**5.10**	4.92	4.99	**1.40**

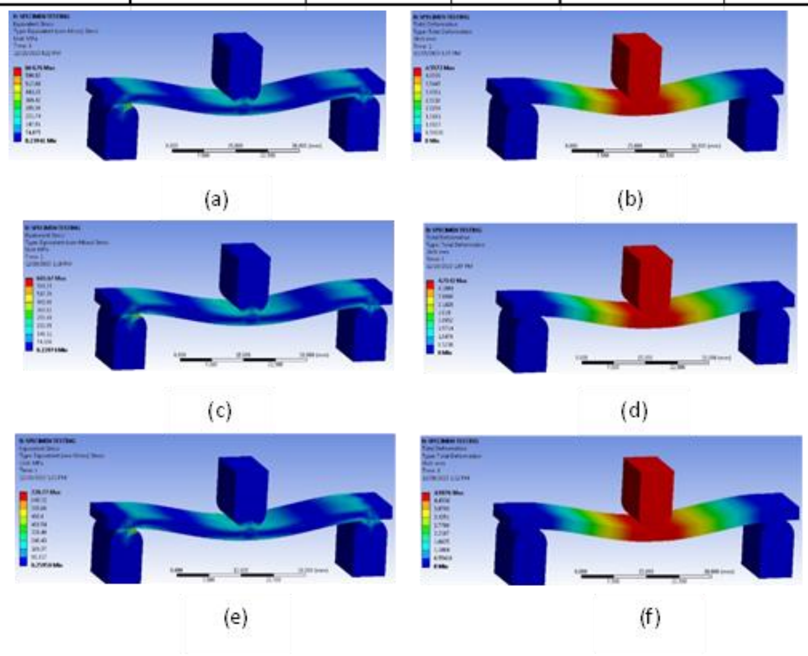

Figure: 7 FEA simulation plots for flexural strength. and total deformation of the (a)-(b) (GC-E)N_0, (c)-(d) (GC-E)N_1, and (e)-(f) (GC-E)N_2 test specimen

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The overall improvement in the flexural characteristics of hybrid laminated composites is mentioned in Table 2. It is observed that the simulated flexural outcomes closely align with the results obtained through experimentation. Finite element models were successfully developed, showing good agreement with experimental data (maximum error of 15.27% in flexural strength, and 4.46% in elongation at break).

3.3 Failure mechanisms of flexural specimens

Figure 8 illustrates the condition of the specimens both before and after the flexural test. The failure of the composites is evident, resulting from the concurrent application of compression, shear, and tensile stresses during flexural loading conditions on the specimens. Moreover, it is evident that delamination took place in the pure (C-E) laminates, while surface cracking occurred on the tensile side of the hybrid (GC-E)N_0 laminates. Among the hybrid composite, (GC-E)N_1 shows the more deformation which results in reduction in flexural strength as shown in Figure 8 (b).

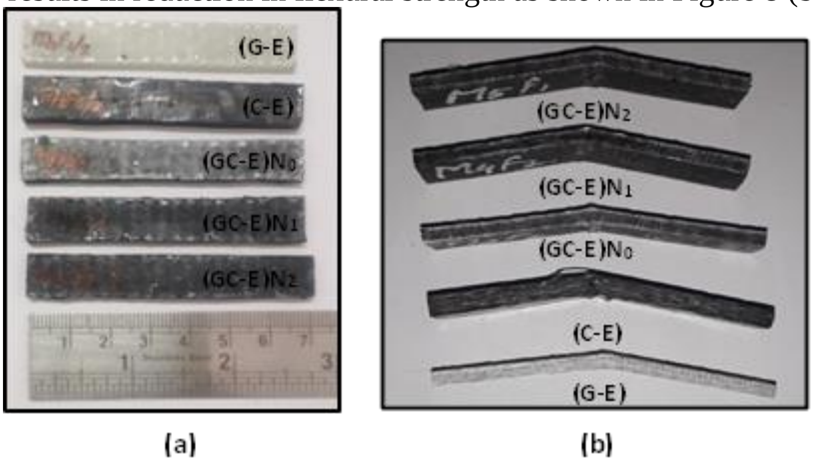

Figure: 8 Flexural test specimens (a) before and (b) after the test

4. Conclusions

Experimental investigations were conducted to examine the combined influence of multi-walled carbon nanotube and halloysite nanotube (MWCNT + HNT) hybrid nanofillers on glass/carbon epoxy composites. The results were validated through numerical means. The incorporation of 1 wt.% MWCNTs + 1 wt.% HNTs in the (GC-E)N_2 composite enhances flexural strength by 4.22 % compared to the unmodified (GC-E)N_0. Additionally, (GC-E)N_2 exhibits a 13.58 % improvement in flexural modulus over (GC-E)N_1, which, with 0.5 wt.% MWCNTs and 0.5 wt.% HNTs, undergoes a 14 % decrease in flexural strength but a 2.27 % increase in flexural strain compared to unmodified (GC-E)N_0.The enhancement in the flexural extension was observed in hybrid composites (GC-E)N_1 and (GC-E)N_2 as compared to (GC-E)N_0. The developed composite model underwent numerical validations through finite element analysis, revealing close coherence with the results. Minor variations in properties were noted, stemming from the assumption made in the FEA simulation that there is no void formation and perfect bonding between fiber and matrix in the composites. In future applications in marine, automotive, and aerospace sectors, hybridization can yield lightweight, cost-effective composites, minimizing environmental impact and lowering carbon footprints.

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