

# Influence of GNP and CB Toughening on the Physical and Mechanical Properties of Hybrid Fiber Epoxy Composites Fabricated with Vacuum Infusion Molding

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## ABSTRACT

The development of lightweight, high-performance composite materials is essential for advancing the structural applications across various engineering fields. This study explores the production of hybrid fiber materials incorporating Graphene Nanoplatelets (GNP) and Carbon Blacks (CB) as reinforcements to improve the mechanical performance. The hybrid fiber materials were manufactured using a Vacuum Infusion Molding (VIM) technique. The mechanical properties, including the tensile and flexural strength, were measured according to ASTM standards. The results show that the composites with 0.1% GNP and 0.1% CB exhibited an optimal mechanical performance, with a highest tensile strength of 354.741 MPa and a balanced failure mode. The higher filler content of 0.2% GNP and 0.2% CB, however, resulted in increased brittleness and poor interfacial bonding, indicated by the higher void content (3%) and reduced fiber-matrix adhesion. The SEM fracture analysis revealed that moderate nanoparticle levels improve the fiber-matrix adhesion, but excessive amounts cause agglomeration and early failure. These findings emphasize the importance of filler optimization to balance the strength, toughness, and durability, paving the way for flax/glass hybrid composites optimized for lightweight, high-performance structural applications.

*Keywords-glass fiber; flax fiber; graphene nanoplatelets; carbon black; flexural properties; vacuum infusing molding; tensile strength; flexural strength*

## I. INTRODUCTION

Fiber-Reinforced Polymer (FRP) composites have attracted significant interest due to their advantageous properties [1]. Natural fibers offer several benefits over synthetic fibers, such as carbon and glass, including low cost, biodegradability, non-toxicity, and a reduced carbon footprint, which also makes their manufacturing process safer [2, 3]. However, their tendency to absorb moisture reduces the mechanical performance and limits their use in outdoor structural applications [4, 5]. Despite this drawback, flax fibers are being actively studied for lightweight and corrosion-resistant applications, such as stairs, walkways, and bridges. Compared to the kenaf/glass composites, flax/glass composites exhibit higher strength and modulus [6], while flax plants themselves grow rapidly under diverse conditions with relatively low water requirements [7]. Additionally, flax yields about 40% usable fiber, like hemp or jute, and requires only around 15 MJ of energy to process 1 kg of flax yields [8]. These advantages have led to the widespread use of flax fibers in semi-structural and non-structural polymer composites; though their structural applications remain limited due to the moisture sensitivity and relatively lower strength [9]. One effective solution is the hybridization with synthetic fibers, such as glass, which can improve the cost-effectiveness, mechanical strength, and sustainability [9, 10]. For instance, authors in [11] reported that carbon/glass hybridization in flax/epoxy composites with stacking sequences (G/F/G and C/F/C) enhanced the impact and flexural strength by 214% and 335%, respectively, compared to the kenaf/epoxy composites (impact strength 9.1 J/m<sup>2</sup>, flexural strength 69.4 MPa). Similarly, authors in [12] reported a tensile strength of 127 GPa  $\pm$  9 MPa, modulus of 8.7 GPa  $\pm$  0.8 GPa, flexural strength of 201 MPa  $\pm$  5 MPa, and flexural modulus of 7.8 GPa  $\pm$  0.5 GPa for G/F/G flax/glass hybrids with 29.7% fiber content, including 12.3% glass fiber. The performance strongly depends on the stacking sequence, with kenaf and glass being proposed as outer layers and jute as the core to optimize the shear strength. Furthermore, authors in [13] found that adding Kevlar as outer layers improved the tensile and flexural properties, while the fracture toughness remains a critical factor for the structural durability, as cracks may occur under stress or due to design flaws [14]. Authors in [15] showed that flax/glass composites arranged in the sequence G/G/G/F/F/G/G/G exhibited the best mechanical properties. Although research on glass/flax epoxy hybrids has been conducted, the specific field remains underdeveloped. To address this gap, the present study investigates a new flax/glass hybrid composite reinforced with nanofillers and epoxy, evaluating its density, tensile, and flexural properties in accordance with ASTM standards.

## II. METHODOLOGY

### A. Materials

The current study employed plain-woven E-glass and flax fibers as reinforcing materials, while GNP and CB were used as nanoparticle fillers. The primary reinforcements consisted of twill flax and twill-woven glass fabrics. Although natural fibers alone cannot match the mechanical performance required for advanced applications in aerospace, marine, and automotive industries [16], their properties can be significantly enhanced

through hybridization. In this work, hybrid composites were fabricated by combining four layers of glass fiber with three layers of flax fiber, reinforced with epoxy resin containing GNP and CB. The stacking sequence of the fibers was carefully designed to optimize the performance. The detailed specifications of the fibers, GNP, and CB are presented in Table I and Table II.

TABLE I. FIBER PROPERTIES

Properties	Flax	Glass
Type	2x2 Twill	Fabric
Density g/cm <sup>3</sup>	1.5	2.54
Areal density g/cm <sup>2</sup>	222.1	700
Fiber bundle Dia, (mm)	(0.8 – 1.2)	13
Ply thickness (mm)	(0.25 – 0.4)	0.5
TS (MPa)	(500 – 1500)	2482.11
Young's modulus (GPa)	50	78.6
Elongation of break (%)	2	3

TABLE II. NANOFILLER PROPERTIES

Product type	GNPs	CB
Appearance (Color)	Dark Grey	Black
Appearance (Form)	Powder	Powder
Particle size ( $\mu$ m)	2 – 8	4
Density (g/cm <sup>3</sup> )	(0.03 – 0.1)	1.79 g/cm <sup>3</sup>

### 1) Epoxy

The composites were fabricated using EPOLAM 2040 resin and EPOLAM 2042 hardener. As summarized in Table III, the epoxy system is composed of these two components, with their respective properties listed.

TABLE III. EPOXY PROPERTIES (RESIN AND HARDENER)

Properties	Resin	Hardener
Type	EPOLAM 2040	EPOLAM 2042
Density (g/cm <sup>3</sup> )	1.16	0.95
Viscosity (mPa.s)	1300	15
Gel time (min)	100	-
TS (MPa)	75	-
Elongation at break (%)	8.7	-

### 2) Epoxy and Nano Filler Mixtures

Exfoliation and functionalization are crucial for enhancing the interfacial bonding of nanomaterials with epoxy polymer, especially for less reactive materials, like GNP and CB. To achieve this, sonication was employed. An Ultrasonic cleaner (model: 040S) with an ultrasonic power of 240 W and a frequency of 40 Hz was utilized to disperse graphene and CB in ethanol to create a dispersion of GNP and CB in an epoxy matrix, as shown in Figure 1(a). This investigation identified ethanol as a low-boiling-point organic solvent. The mixture of GNP/Ethanol and CB/Ethanol underwent sonication in a bath ultrasonic sonicator for 60 min at a temperature range of 30 °C-45 °C. The ethanol was evaporated after sonication by heating in an open beaker at 80 °C to 120 °C. Sonicated GNP nanoparticles and CB in powder form were mixed with a certain quantity of epoxy using mechanical stirring for 30 min at 200 revolutions/min at 25 °C, as depicted in Figure 1(b). The specified amount of hardener was included in the mixture using

mechanical agitation for 7 min at 120 revolutions/min at ambient temperature. The mixture was allowed to degas for 10 min before being infused into the fibers during the VIM process [17].

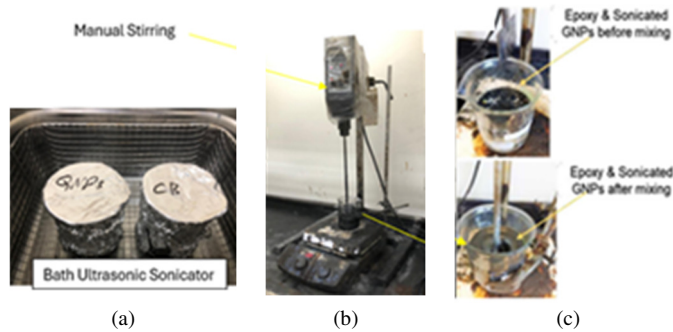


Fig. 1. Preparation of GNP and CB: (a) bath-type ultrasonic dispersion instrument and (b) mechanical stirring and heater.

**B. Fabrication Process**

The VIM technique is used to fabricate composites, allowing for parts with complex shapes. This study investigated hybrid fibers consisting of 7 layers with varying GNP and CB percentages. The fiber stack includes 3 layers of flax fiber and 4 layers of woven glass fiber, forming a 3.8 mm-thick composite laminate. Flax fibers were pre-dried at 60 °C for 3 h before resin infusion. The mold was treated with a wax-releasing agent and left for 15 min. Sealing tape was applied before laying the fiber layers. Peel-ply fabric and infusion mesh were added on top to avoid contact and ensure an even resin distribution. Spiral tubing was placed at the outlet, and inlet points connected to PVC hoses for resin flow. These were linked to a vacuum pump and catch-pot system. A baggy film was sealed using adhesive tape to prevent air leaks. Clamps controlled the resin flow at both ends. The vacuum pump ran until reaching 40 kPa–60 kPa. After verifying the airtightness for 15 min, a mixture of 0.1% GNP and 0.1% CB with epoxy and hardener (100:32 ratio) was prepared and rested for 10 min. Resin infusion began by unclamping the inlet. After infusion, the hoses were clamped, and the composite was cured for 24 h. Finally, a post-curing process was performed at 60 °C for 6 h, as portrayed in Figure 2, to relieve the internal stress.

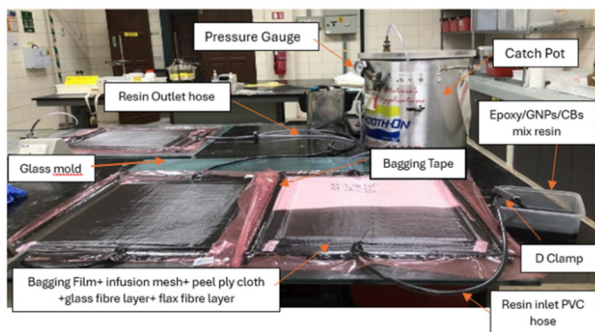


Fig. 2. VIM process.

**C. Physical Properties**

The density of the experimental composites was evaluated using a Mettler Toledo AX-205 device following the specifications of [24]. An abrasive diamond cutter was used to cut three 10 mm x 10 mm samples from each panel. To determine the test accuracy, the average reading of these samples was calculated. The fiber volume fraction for the fiber-reinforced hybrid composites is calculated by:

$$W_f = \frac{M_f}{M_{comp}}; M_f = M_{f1} + M_{f2} \tag{1}$$

$$M_{f1/f2} = W_f \times n \times A_{comp} \tag{2}$$

where a composite's fiber weight fraction ( $W_f$ ) is calculated using the ratio of the fibers' mass to the resulting mass. The designations  $M_f$ ,  $M_{comp}$ ,  $W_f$ ,  $n$ , and  $A$  represent the mass of fibers, the mass of the composite, the areal density of fibers, the number of layers, and the area of the composite, respectively, whereas  $f_1$  and  $f_2$  represent fiber one and fiber two in hybrid composites.

**D. The Measurement of Tensile Properties**

The tensile test was conducted using the Universal Testing Machine (UTM), as shown in Figure 3. Tensile tests were performed on the composites at 25 °C and 87% relative humidity [23]. The test scenarios required loading the specimens until they failed using a Zwick/Roell 50 kN computer-controlled UTM. The testing speed was constant at 3 mm/min for all specimens. The specimens measured 250 mm x 25 mm x 3.8 mm. The tensile properties were found using:

$$\sigma = \frac{P_{max}}{A}; \varepsilon = \frac{\delta}{L_0}; E_B = \frac{\sigma}{\varepsilon} \tag{3}$$

where  $\sigma$  is the stress,  $P$  is the load (N),  $\varepsilon$  is the strain,  $L$  is the gauge length (50 mm),  $A$  is the cross-section area (mm<sup>2</sup>), and  $E_B$  is the tensile modulus according to [23].

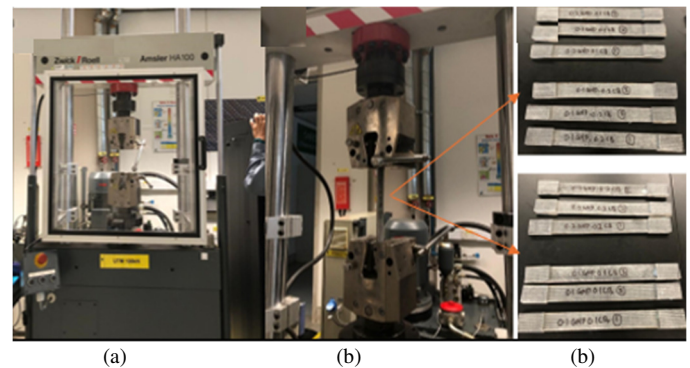


Fig. 3. Tensile testing: (a) UTM for tensile test, (b) machine setup, (c) samples of hybrid composite.

**E. Measurement of the Flexural Properties**

The flexural test was conducted with the 5 kN GOTECH UTM machine, controlled by a computer, following the specifications of [25]. Three specimens from each configuration were tested. The specimens were 3.9 mm in thickness and had a width of 15 mm. The specimen's length must be enough to have an overhang at each end, equal to at

least 10% of the support span, but never less than 6.4 mm on either end [18]. The test speed and flexural properties were calculated using:

$$R = \frac{ZL^2}{6d} ; \sigma_f = \frac{3PL}{2bd^2} ; \epsilon_f = \frac{6Dd}{L^2} ; E_B = \frac{L^3m}{4bd^3} \quad (4)$$

where  $R$  is the testing speed,  $Z$  is 0.1 [25],  $L$  is the supporting span ( $L = 16 \cdot d$ ),  $d$  is the composite thickness (mm),  $\sigma_f$  is the flexural strength (MPa),  $\epsilon_f$  is the flexural strain (%),  $E_B$  is the flexural modulus,  $P$  is the load (N),  $b$  is the width of the specimen (mm), and  $m$  is the slope (N/mm).

### III. RESULTS AND DISCUSSION

#### A. Physical Properties Results for Hybrid Composites

In this study, hybrid composite panels were fabricated using four layers of glass fiber and three layers of flax fiber, with varying percentages of GNP and CB fillers. Each panel measured 30 cm × 30 cm × 3.8 mm and a density of 1.4 g/cm<sup>3</sup>. To ensure proper impregnation and structural integrity, the fiber weight fraction was maintained at 41%, corresponding to a fiber volume fraction of 26.68%, and was consistently kept below 55% across all samples. The void content, a key factor influencing the composite quality, ranged from 1% to 3%. Typically, a low void content (<1%) indicates a strong fiber-matrix adhesion, while a high void content (>5%) suggests structural defects [19]. The lowest void content (1%) was observed in composites reinforced with 0.1% GNP + 0.1% CB, and 0.1% GNP + 0.2% CB, reflecting negligible defects and enhanced fiber-matrix interaction. In contrast, the highest void content (3%) occurred in composites with 0.2% GNP + 0.2% CB, likely due to the excessive filler loading, which promotes the agglomeration and air entrapment [20]. A moderate void content (2%) was recorded for composites containing 0.2% GNP + 0.1% CB, indicating an acceptable but slightly reduced quality. Overall, the results suggest that lower filler loadings (0.1%) produce superior composite quality by minimizing the void formation, whereas higher filler contents may enhance certain mechanical properties but increase the void formation, ultimately compromising the structural performance.

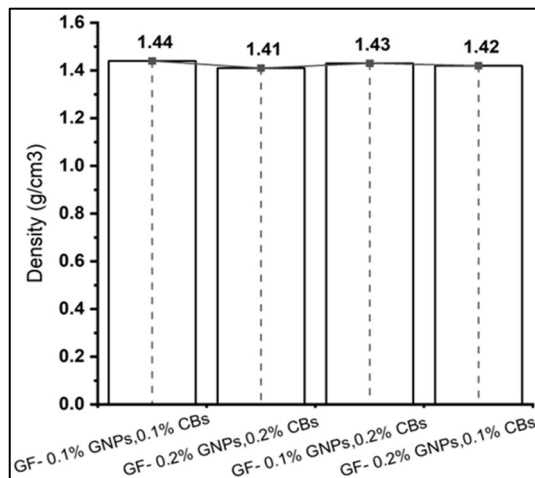


Fig. 4. Density results.

TABLE IV. RESULTS OF PHYSICAL PROPERTIES

Composite	Thickness (mm <sup>2</sup> )	Weight percentage of filler (wt%)	$\rho_{comp}$ (g/cm <sup>3</sup> )	$W_f$ (%)	$V_f$ (%)	$V_m$ (%)	$V_{void}$ (%)
Hybrid Flax/glass	3.8	0.1 GNP and 0.1 CB	1.44	41	26.68	73.32	1
Hybrid Flax/glass	3.8	0.2 GNP and 0.2 CB	1.41	41	26.68	73.32	3
Hybrid Flax/glass	3.8	0.1 GNP and 0.2 CB	1.43	41	26.68	73.32	1
Hybrid Flax/glass	3.8	0.2 GNP and 0.1 CB	1.42	41	26.68	73.32	2

#### B. Strength Results

##### 1) Tensile Strength of Flax/Glass Reinforced Hybrid Composites

The tensile properties of flax/glass hybrid composites with varying GNP and CB loadings were evaluated in terms of tensile strength, strain (%), and tensile modulus, as shown in Figure 5. The highest average tensile strength of 354.741 MPa, along with a tensile modulus of 22.08 GPa, were obtained for composites containing 0.1% GNP and 0.1% CB. This indicates that a low, balanced filler content promotes the uniform dispersion, minimizes the agglomeration, and enhances the stress transfer within the matrix, complying with the improvements reported in [15]. In contrast, the lowest tensile strength (336.099 MPa) was recorded in composites with 0.2% GNP and 0.1% CB, likely due to the GNP agglomeration, which disrupts the fiber-matrix bonding and reduces the load transfer efficiency. Similarly, the composites with 0.2% GNP and 0.2% CB achieved a tensile strength of 344.762 MPa but exhibited the lowest tensile modulus (19.22 GPa), highlighting that the excessive filler loading negatively affects stiffness [21]. Overall, the results confirm that a lower filler content (0.1%) yields optimal tensile properties, whereas higher concentrations (0.2%) promote the aggregation and reduce both the strength and stiffness. Thus, the precise control of filler dispersion is critical for maximizing the mechanical performance.

##### 2) Flexural Strength of Flax/Glass Reinforced Hybrid Composites

As shown in Figure 6, the highest flexural strength of 438.674 MPa, was obtained in the hybrid composite reinforced with 0.1% GNP and 0.2% CB. A similarly high value of 428.513 MPa was achieved with 0.1% GNP and 0.1% CB. Composites with 0.2% GNP and 0.1% CB and with 0.2% GNP and 0.2% CB showed slightly lower strengths of 410.68 MPa and 340.707 MPa, respectively. In contrast, the pure glass/flax composite exhibited the lowest strength of 294.45 MPa, confirming that the addition of GNP and CB significantly improves performance [22]. These results indicate that the combination of 0.1% GNP and 0.1% CB provide the most effective reinforcement, while higher GNP content may cause agglomeration and reduce strength. The flexural modulus results in Figure 6(b) support this conclusion, showing a substantial increase in stiffness with the incorporation of GNP and CB. The pure GF composite had the lowest modulus (10.8 GPa), whereas the 0.1% GNP and 0.1% CB sample reached the highest value (21.66 GPa), demonstrating excellent load transfer, adhesion, and dispersion. Increasing CB to 0.2% (with

0.2% GNP) reduced the modulus to 20.958 GPa, likely due to agglomeration. Nevertheless, all modified composites nearly

doubled the stiffness of pure GF, confirming the role of GNP and CB in enhancing mechanical integrity.

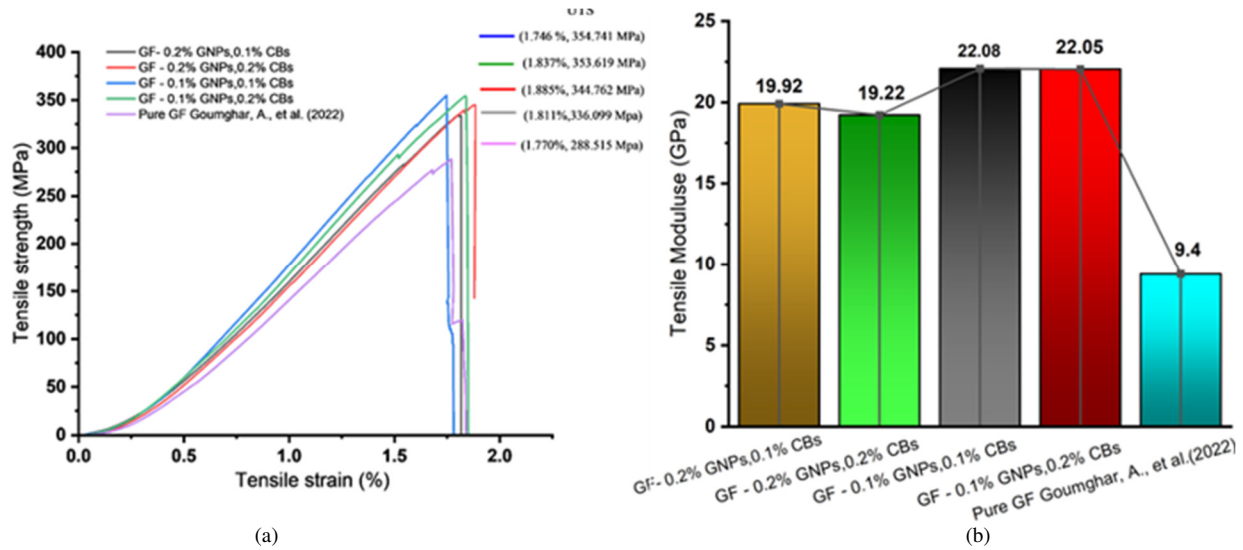


Fig. 5. (a) Tensile strength and strain, (b) tensile modulus.

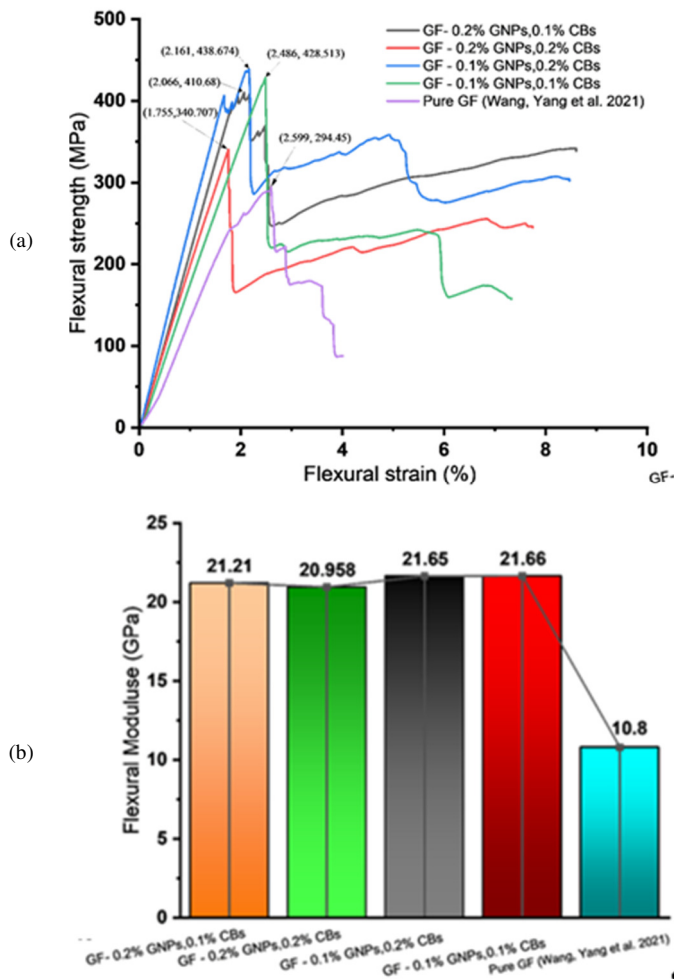


Fig. 6. Flexural results: (a) flexural stress and strain, (b) flexural modulus.

C. Fracture Surface Analysis

1) Fracture Surface Analysis for Tensile Samples

The fracture behavior of the flax/glass hybrid reinforced GNP and CB composites after tensile testing was analyzed to assess how the nanofiller affects the failure mechanisms. The SEM images show the samples' response to the tensile stress, offering a detailed look at the fracture characteristics, according to Figure 7. Figure 8(a), containing 0.2% GNP and 0.1% CB, demonstrated a significant fiber pullout after testing, indicating weak fiber-matrix adhesion. This suggests that although the composite can withstand the tensile stress to some degree, failure mainly occurs through fiber detachment rather than breakage, which reduces the load-bearing efficiency.

In contrast, Figure 7(b), with 0.1% GNP and 0.2% CB, displayed a more uniform failure pattern with both the fiber pullout and breakage. The higher CB content seems to improve the fiber-matrix bonding, resulting in a more controlled failure mode and potentially better damage resistance. Figure 7(c), containing 0.2% GNP and 0.2% CB, showed a brittle failure mode involving fiber fracture and epoxy debonding. High concentrations of nanoparticles may cause agglomeration, which decreases the reinforcing efficiency. As a result, while the composite initially resists the external stress, failure occurs suddenly with rapid propagation, leading to abrupt breakage. Conversely, Figure 7(d) with 0.1% GNP and 0.1% CB exhibited a balanced failure mode with both fiber pullout and fracture, indicating an optimal strength and toughness balance.

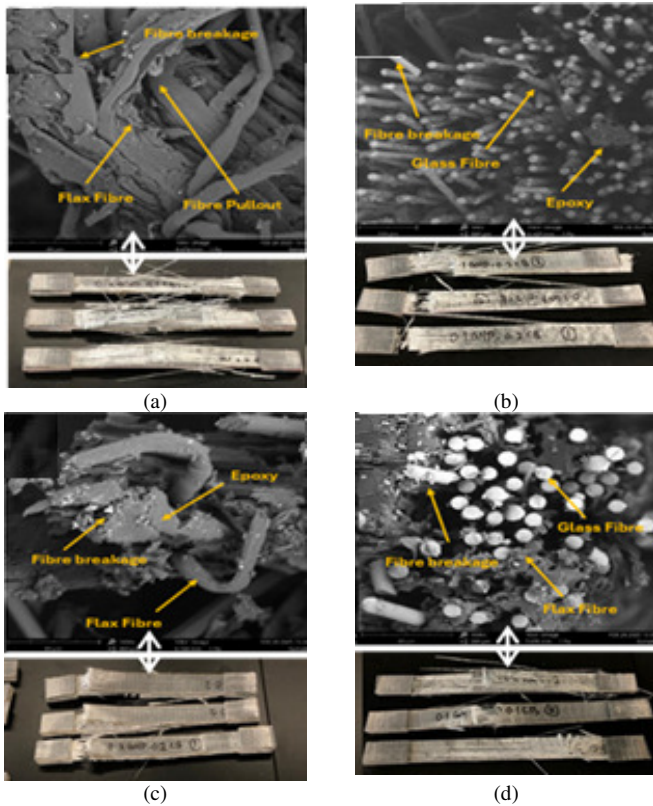


Fig. 7. Fracture surface of the flax/glass hybrid composite after flexural testing.

## 2) Fracture Surface Analysis for Flexural Samples

The fracture behavior of flax/glass hybrid composites after flexural testing, as illustrated in Figure 8, was examined to assess the influence of the GNP and CB fillers on the mechanical strength and failure mechanisms. In Figure 8(a), the composite containing 0.2% GNP and 0.1% CB exhibited extensive fiber breakage, particularly in the glass fibers. While this configuration demonstrated relatively high strength, it also showed increased brittleness, as the material was unable to absorb significant energy before sudden fracture. In contrast, Figure 8(b) shows that the 0.1% GNP and 0.2% CB composite failed with a bimodal mode, involving both fiber fracture and epoxy deformation. This mixed failure suggests improved toughness, as the material could withstand greater loads prior to failure. The composite with 0.2% GNP and 0.2% CB as Figure 8(c) shows, displayed fiber pullout and epoxy cracking, indicating reduced fiber–matrix bonding. This weak adhesion led to a stress redistribution within the laminate and made the composite more susceptible to interfacial failure. Meanwhile, the 0.1% GNP and 0.1% CB composite, as shown in Figure 8(d), exhibited a balanced failure mode involving both fiber fracture and epoxy deformation. This combination reflects an optimal balance of strength and toughness, enabling the material to resist the applied loads while maintaining structural integrity. Overall, this study demonstrates the potential of flax/glass hybrid composites reinforced with dual nanofillers (GNP and CB), fabricated via vacuum infusion. Unlike previous research that focused solely on the hybrid fibers or single fillers, the present work highlights the synergistic effect

of 0.1 wt% GNP and CB in enhancing the tensile and flexural properties while keeping the void content low. Specifically, the tensile and flexural performance improved by 18.67% and 33.65%, respectively, compared to the hybrid composites without nanofillers [15, 22].

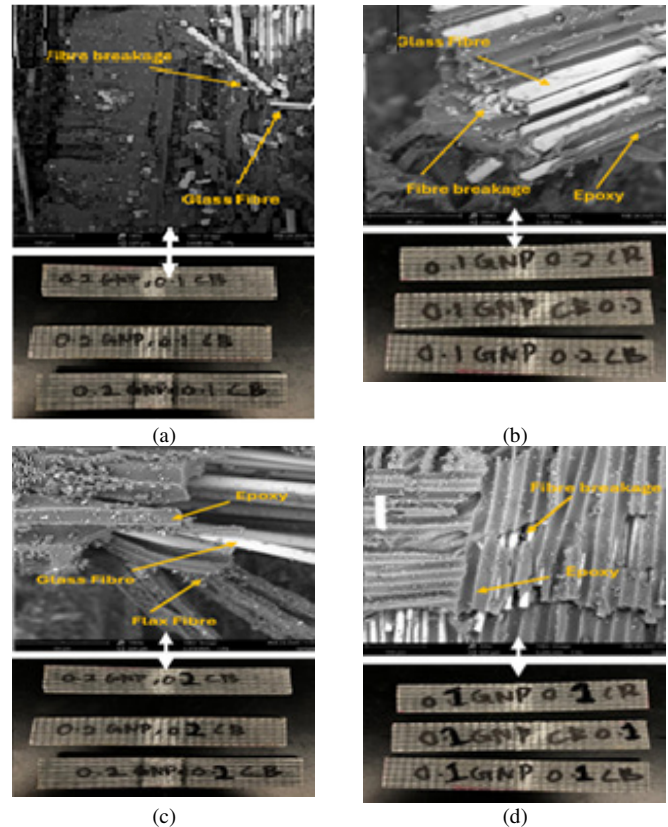


Fig. 8. Fracture surface of the flax/glass hybrid composite after flexural testing.

## IV. CONCLUSIONS

In this study, the mechanical properties of flax/glass hybrid composites reinforced with GNP and CB were investigated using VIM. The results showed that the optimum composition of 0.1% GNP and 0.1% CB achieved the highest tensile strength of 354.741 MPa and demonstrated a balanced failure mode, reflecting improved fiber–matrix adhesion and superior mechanical performance. In contrast, higher filler loading (0.2% GNP and 0.2% CB) led to increased void content (3%), reduced interfacial bonding, and greater brittleness due to the nanoparticle agglomeration. The SEM fracture analysis further confirmed that an optimal filler content enhances the fiber–matrix interaction, whereas excessive filler levels hinder the efficient stress transfer. The flexural testing revealed that the composites with 0.1% GNP and 0.2% CB exhibited the highest flexural strength (438.674 MPa), emphasizing the importance of optimizing the nanofiller ratios. Overall, the findings highlight the strong potential of hybrid natural fiber composites for structural applications where high strength, toughness, and lightweight properties are critical. However, precise control over nanofiller dispersion is essential to preserve the composite integrity and prevent aggregation. Future work should

investigate the long-term durability of these composites under varying environmental conditions to evaluate their suitability for real-world applications.

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#### DATA AVAILABILITY STATEMENT

The datasets generated and/or analyzed during the current study are available from the corresponding author upon reasonable request.

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