Research Article

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Hybridization of biocomposites with oil palm cellulose nanofibrils/graphene nanoplatelets reinforcement in green epoxy: A study of physical, thermal, mechanical, and morphological properties

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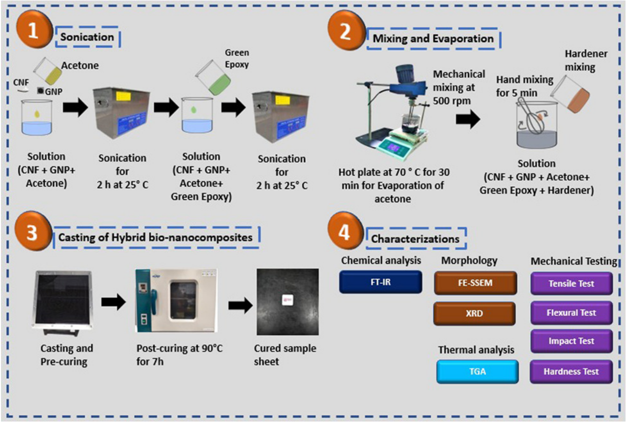
Abstract: With the rising demand for sustainable and high-performance materials, biobased nanocomposites have gained considerable attention as eco-friendly alternatives to conventional polymers. In this research, the effective incorporation of the oil palm cellulose nanofibrils (CNFs) and graphene nanoplatelets (GNPs) as nanofillers into biopolymer matrices made by biomass with 28% carbon content resulted in notable improvements in mechanical properties. The study aimed to fabricate green/bioepoxy hybrid nanocomposites by using acetone as the solvent in a solution blending procedure with varying CNF loadings (0.1, 0.25, and 0.5 wt%). The resulting nanocomposites were thoroughly analyzed and compared to unfilled green epoxy. The mechanical test results were consistent with previously published studies, demonstrating a 77% increase in tensile strength and 23.46% improvement in hardness when evaluated against unfilled green epoxy. However, the green epoxy hybrid nanocomposites' thermal degradation temperature (T_d) revealed a drop in temperature between 0.1 wt% of CNF/GNP loading and unfilled green epoxy (neat). Overall, the results demonstrated a significant improvement in the mechanical performance of green epoxy hybrid nanocomposites and showed the potential of using low-loading nanofillers in green epoxy. The hybrid composites can be used in electronics, marine, and biomedical applications.



Graphical abstract

Keywords: nanocellulose, graphene nanoplatelets, hybrid green nanocomposites, green epoxy, thermal properties, mechanical properties

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1 Introduction

Identifying effective, renewable, sustainable, and ecologically sound resources and materials exhibiting favorable properties represents a formidable challenge. It constitutes an essential pathway toward attaining sustainable advancement, particularly considering the precipitous depletion of non-renewable resources and the exacerbation of environmental degradation. These changes are driven by the rapidly developing social economy and accelerating industrialization [1,2]. Cellulose nanofibrils (CNFs) represent naturally occurring polymeric nanomaterials characterized by their superior biocompatibility and biodegradability, as well as possessing commendable mechanical properties, substantial strength, reduced density, extensive specific surface area, and adjustable chemical characteristics. Due to its biodegradable nature, sustainability, and biosafety features, cellulose is becoming popular. Cellulose, a biodegradable polymer derived from renewable sources like plants and algae, has applications in biomedical fields, paper production, textiles, and packaging [3,4]. However, issues like moisture absorption and poor mechanical properties for natural fiber-reinforced composites (NFRCs) pose a challenge to utilize on a large scale [5]. The problems result in fungal growth, poor bonding, and swelling. They are vulnerable to biological threats, ultraviolet (UV) deterioration [6], and limited thermal stability [7]. Low fatigue resistance and creep deformation also restrict long-term performance. Fiber hybridization, matrix modification, UV/fire retardants, hydrophobic coatings, and chemical treatments can all increase the durability of NFRCs [8].

It has been shown that CNF combined with other nanomaterials, such as graphene derivatives and graphene nanoplatelets (GNPs), is useful for developing hybrids with exceptional physicochemical features, high performance, and configurable functionality [9].

Recently, there has emerged significant scholarly interest in hybrid organic–inorganic nanocomposites synthesized *via* environmentally sustainable methods that utilize renewable intelligent materials. This increasing attention is attributable to their numerous advantages and unique properties. Given the dependence on limited and exhaustible non-renewable resources, the emphasis has transitioned toward the fabrication and optimization of hybrid composite materials [10]. Numerous scholars have utilized a diverse array of methodologies to synthesize graphene nanocellulose-reinforced hybrids, encompassing techniques such as layer-by-layer assembly, ultrasonication, ambient pressure drying, ball milling, and liquid-phase exfoliation [11]. The most common method is solution blending along with ultrasonication. Mohammadsalih *et al.* [12] used solution blending along with sonication for dispersing graphene nanosheets in a thermoplastic matrix. The resultant polystyrene/graphene oxide nanocomposites exhibited better dispersion and enhanced thermal properties.

Liu *et al.* [13] produced cellulose nanowhisker/GNP composite films using sonication mixing and solution casting. The composite films showed enhanced melting and decomposition temperatures, electrical conductivity, and tensile strength, making them suitable for bio-anti-static and electronic packaging applications. Many other researchers have also worked on the CNF/GNP, as shown in Table 1.

Epoxy is widely used for its mechanical properties and chemical resistance. However, it faces an environmental concern due to the toxic behavior of bisphenol A (BPA), resulting in endangering human and animals [19]. This is vital in the context of increasing concerns regarding environmental contamination and the discharge of hazardous substances into aquatic ecosystems attributable to synthetic polymers [20]. Thus, efforts are made to develop biopolymers from renewable resources like vegetable oils and lignin, which are aimed to replace BPA along with an added value [21] (Figure 1). Green epoxy resin is another alternative in a green matrix for producing NFRC. Green epoxy is produced from many renewable resources such as *Nahar* seed plant oil and bio tea leaf phenolic extract [21]. To be labeled as green epoxy, the resin needs at least 25% biocontent as per United States Department of Agriculture (USDA) standards. Researchers claim that green epoxy significantly reduces biohouse gas emissions at 30% biocontent.

Borah and Karak [23] developed bio-derived green epoxy composites using functionalized waste tea fibers. The results showed enhanced mechanical and thermal properties. The composites demonstrated high durability under harsh conditions, including UV exposure and extreme temperatures. Kumar *et al.* [24] used novel lignin-derived vanillyl alcohol epoxy (m&dVAE) thermoset to develop a unique mono- and diepoxy cross-linking structure, offering a sustainable alternative to petroleum-based epoxies. When cured with 4,4′-diaminodiphenyl methane (DDM), it outperformed commercial diglycidyl ether bisphenol A (DGEBA) in tensile strength, modulus, and adhesion with cellulose nanofibers. Zindani *et al.* [25] developed green biocomposites using short *Punica granatum* fibers (SPGF) reinforced in bioepoxy derived from cashew-nut shell oil. SPGFs were treated with sodium bicarbonate for varying durations to enhance the composite performance. While chemical treatment did not alter the cellulose structure, it improved physical and mechanical properties, especially after 5 days of treatment. The eco-friendly process supports sustainable composite fabrication with reduced environmental impact.

Table 1: Various fillers in the polymer matrix

Polymer matrix	Hybrid filler	Preparation technique	Findings	Applications	Ref.
Glass/Epoxy	Multiwall carbon nanotubes (MWCNTs), nano-silica (NS), and nano-iron oxide (NFe)	Sonication	Significant improvement to the polymer cross-linking web	Automotive, aviation, and defense industries	[14]
Epoxy	Alumina nanoparticles, nylon-6 fibers, glass fibers	Solution blend technique	Reduces the wear loss of epoxy hybrid nanocomposites	Biomedical domains	[15]
Bisglycidyl dimethacrylate (Bis-GMA)/triethylene glycol dimethacrylate (TEGDMA)	Silica/poly(methylmethacrylate) (PMMA)	Emulsion polymerization, sonication	A higher content of PMMA in the hybrid particles results in a lower modulus owing to the lower amount of crosslinking in the matrix	Dental restoration	[16]
GNP paper	CNF	Vacuum-assisted self-assembly	CNF efficiently links the neighboring GNP sheets, thus reinforcing the structural integrity of the GNP papers	Gas barrier	[17]
PVA (polyvinyl alcohol)	Cellulose nanocrystals (CNCs) and graphene oxide (GO)	Sonication	The synergistic effect of hybrid nanofillers improved dispersion, leading to nanocomposites with superior performance compared to those with single nanofillers	Food packaging	[18]

Through a synergistic interaction between the high surface area of nanocellulose and the remarkable strength of graphene, hybridizing nanocellulose with GNPs in a green epoxy matrix greatly enhances load transfer, crack resistance, and thermal conductivity [26]. The hybrid composite becomes suitable for sophisticated structural and functional applications while preserving biodegradability and sustainability due to this combination, which also strengthens the matrix at the nanoscale and improves barrier qualities, durability, and multifunctionality.

While a considerable volume of research has been conducted previously regarding nanoparticles and epoxy composites, encompassing an extensive range of scientific explorations and analyses. Nonetheless, there exists a gap in research aimed at investigating the implications of incorporating minimal concentrations of nanoparticles with bio-derived green epoxy in composite fabrication.

This research seeks to investigate the influence of low loading levels on CNF/GNP-reinforced green epoxy nanocomposites. The present investigation incorporates low-loading fillers of GNPs to address the mechanical deficiencies observed. The methodology employed for the fabrication of the nanocomposites was solution blending, followed by a thorough examination of their properties. In terms of mechanical evaluation, tensile, flexural, impact, and hardness tests were carried out. The composition of the nanocomposites was interpreted through the analysis of the Fourier-transform infrared (FTIR) spectrum. Furthermore, thermogravimetric analysis (TGA) was conducted to assess the thermal properties. In addition, field emission scanning electron microscopy (FE-SEM) was utilized to analyze the morphological attributes.

2 Experimental

2.1 Materials

The commercially available green epoxy resin, designated as SR Bio Poxy 28 (Part A), was provided by Mecha Solve Engineering (Kuala Lumpur, Malaysia) with its corresponding hardener SD 3304 (Part B). The density of the green epoxy resin was determined to be 1.17 ± 0.01, while its viscosity at a temperature of 30°C was measured at 4,500 mPa s, and the gel time at the same temperature was 3.45 h. GNP was sourced from GO Advanced Solutions Sdn. Bhd. (Malaysia). GNP presented as a black/gray powder, exhibiting a specific surface area in the range of 20–40 m^2/g, a carbon content exceeding 99%, an apparent density

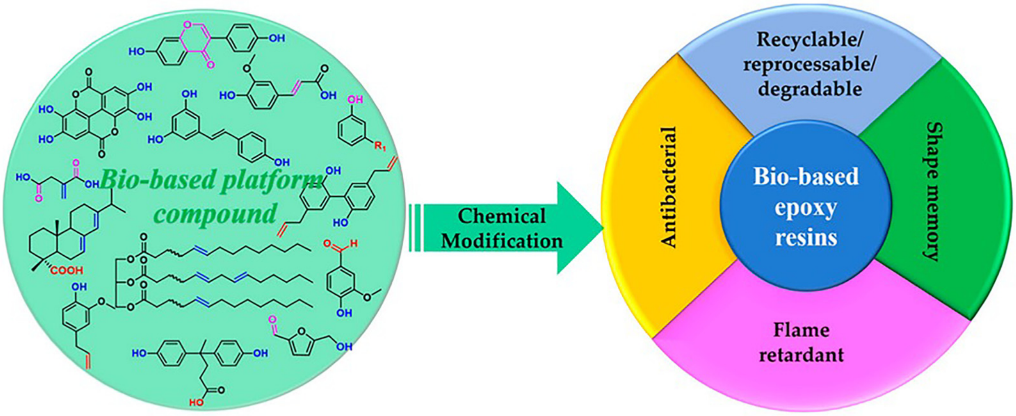

Figure 1: Transformation from green platform compounds to green epoxy resins [22].

between 0.06 and 0.09 g/mL, a water content below 2 wt%, and residual impurities less than 1 wt%. The spray-dried CNF derived from oil palm was obtained from Zoepnano Sdn. Bhd., Putra Science Park, Serdang, Selangor, Malaysia. The physical characteristics of the CNF were described as white and opaque. The nanocellulose content was found to be 99%, the pH level was neutral, the diameter was less than 50 nm, dynamic light scattering (DLS) measurements ranged from 40 to 25 nm, and the onset temperature was approximately 300°C, with the complete deposition temperature reaching around 600°C. Acetone served as a solvent agent, facilitating the dispersion process.

2.2 Methods

The GNP/CNF-reinforced green epoxy hybrid green nanocomposites were synthesized through a solution blending methodology encompassing four fundamental steps: the dissolution of nanoparticles in a solvent utilizing sonication (Digital Pro$^+$, Malaysia), the incorporation of the polymer solution, subsequent mixing, and ultimately, the removal of the solvent *via* evaporation (Faithful Instrument, China). The combined GNP/CNF was prepared in varying weight percentages, such as (0.1, 0.25, and 0.5) wt%, which were precisely measured using a digital balance (AND GF-1000, Japan) with a readability of 0.01 mg. These materials were then dispersed in acetone within a glass beaker and subjected to sonication with an ice bath employed to maintain a lower water temperature; after sonication, the suspension was permitted to remain undisturbed for a period to observe sedimentation characteristics.

Figure 2 illustrates the primary procedures involved in the synthesis of GNP/CNF-reinforced green epoxy hybrid nanocomposites. Green epoxy was subsequently incorporated in acetone and GNP/CNF, followed by sonication for a duration of 2 h. Acetone was subsequently eliminated through the utilization of a laboratory hot plate, with the mixture maintained at a temperature of 75°C for a period of 2.5 h. Continuous agitation was performed to ensure homogenous mixing of the green epoxy with GNP/CNF, facilitated by a mechanical stirrer (IKA RW 20, Malaysia) operating at 500 rpm. A curing hardener, designated as part B of the epoxy, was combined in a ratio of 8:2 and manually blended for 5 min. Steel mold with dimensions of (200 mm × 200 mm × 3.0 mm) was pre-prepared by applying mold release wax (Meigura's mirror glase) in three successive applications at 30 min intervals. The resulting mixture was then introduced into the steel mold and subjected to pre-curing for a duration of 16 h at ambient temperature. The mold was post-cured for 7 h at 90°C in an oven (Lichen Technology, China). After the sheet was taken out of the mold, samples were made for testing using a vertical bandsaw (Makita LB1200F, Japan).

2.3 Characterization

2.3.1 Chemical analysis

The functional groups present in GNP/CNF-reinforced green epoxy hybrid green nanocomposites were analyzed utilizing a Thermo-Fisher Nicolet iS10 spectrophotometer through FTIR spectroscopy, conducted within the frequency range of 400–4,000 cm^{-1}. To acquire the spectra for the FTIR analysis, 16 scans were performed at a velocity of 0.47 cm/s, which aimed to accurately explain the functional groups associated with the CNF/GNP component and to delineate the characteristics of the hybrid nanocomposites.

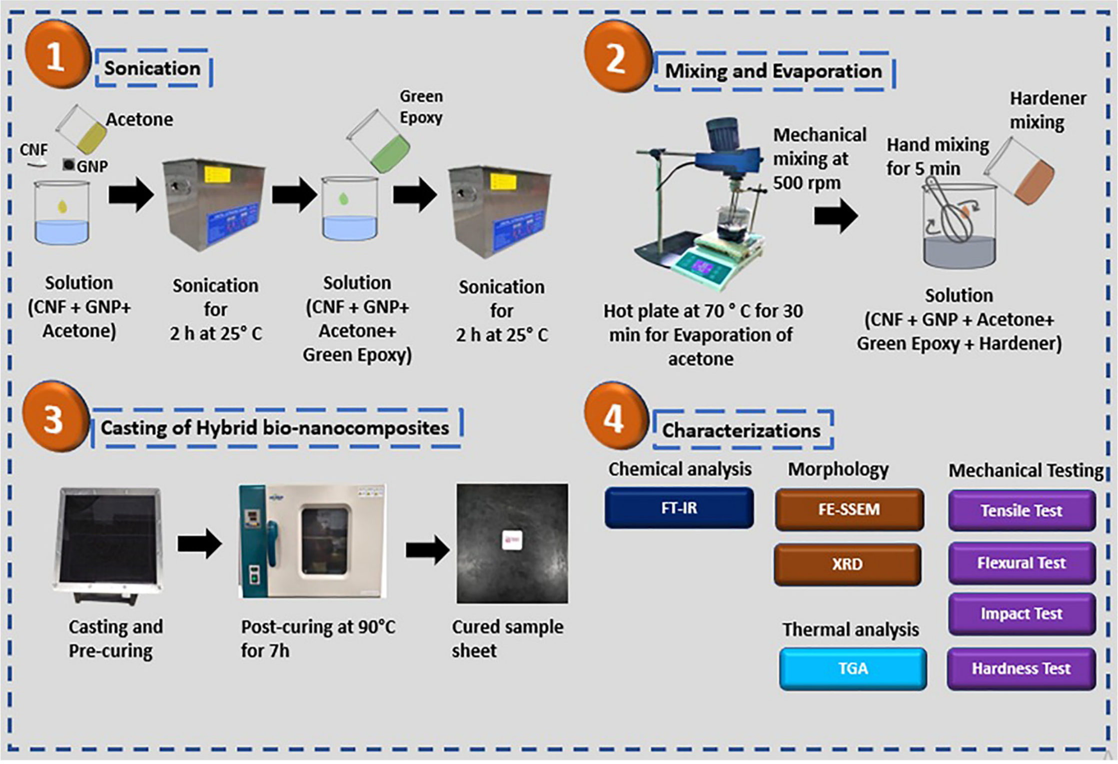

Figure 2: Methodology for the preparation of GNP/CNF-reinforced green epoxy hybrid nanocomposites.

2.3.2 Morphological analysis

The microstructure analysis of the broken surface of hybrid green nanocomposites, which was the outcome of a tensile test, was done using a 5.0 kV field-emission scanning electron microscope (JEOL JSM-7600F FESEM, Japan). The vacuum suction source runs the FESEM at discrete magnifications ranging from 5,000 to 50,000. To improve the observation of fragmented surface morphology, the sample was coated with platinum using a sputter (Model: K575X, UK) for a duration of 50 s.

2.3.3 X-ray diffraction (XRD)

The dispersion state of the nanoparticles in each epoxy nanocomposite sample was analyzed using the XRD technique. XRD testing was performed using Rigaku SmartLab (2), MiniFlex 600 (Rigaku Corporation, Japan) at 25°C, 40 kV, and 30 mA. The samples (20 mm × 20 mm × 3 mm) were scanned from $2\theta = 5°$ to 60°.

2.3.4 Thermogravimetric analysis

Thermogravimetric analysis (TGA Mettler Toledo, Malaysia) serves to ascertain the temperature at which thermal degradation occurs, denoted as T_d. A sample mass of 10 mg was employed during this investigation. The sample holder is constructed from alumina and ceramics, possessing a volume of 70 μL. The experimental procedure is executed by directing 50 mL of N_2 gas through the apparatus while maintaining a temperature increment of 10°C/min. The temperature was maintained within the range of 25°C.

2.4 Mechanical testing

Tensile, flexural, and impact strength, along with Rockwell hardness, were measured for the mechanical properties of the nanocomposites. Every test was conducted on five samples, and the standard deviation and average value were calculated.

2.4.1 Tensile test

The tensile testing of hybrid nanocomposites was conducted by the ASTM D638–10 standard [27] utilizing a Universal Testing Machine (UTM) INSTRON 3366 (Instron, USA) under ambient temperature conditions. The dimensions of the specimens were maintained at (150 mm × 25 mm × 3.0 mm), adhering to the specified standards. During the execution of the tensile test, the crosshead velocity was set at 2.0 mm/min.

2.4.2 Flexural test

Samples of hybrid nanocomposites were tested on an INSTRON 3366 (Instron, USA) using a three-point loading method by ASTM D 790 standard [28]. According to the standard, the specimen's dimensions were (127 mm × 12.7 mm × 3.0 mm). During the flexural test, the span length was 50 mm, and the crosshead velocity was 12 mm/min. A test was run to determine the modulus of elasticity.

2.4.3 Rockwell hardness test

To measure the hardness of the hybrid nanocomposites, the Rockwell F hardness test was used on a digital hardness tester. The testing samples are 25 mm × 25 mm in accordance with ASTM D 785 [29]. A 1/16-inch-diameter steel ball was employed for 5 s at 490 N.

2.4.4 Impact test

The amount of energy absorbed prior to the rupture of the hybrid nanocomposites was measured using the Charpy test. Impact strength was computed using the device (Gunt Hamberg, Germany). Polymer nanocomposites were tested for impact strength using the ASTM D256 standard [30]. The samples were notched prior to the test and had dimensions of 65 mm × 15 mm × 3 mm.

3 Results and discussion

3.1 Chemical analysis

The FTIR spectral analysis was employed to identify the functional groups present within the hybrid composite and to explain the interactions among GNPs, CNF, and green epoxy molecules, as illustrated in Figure 2. The FTIR spectra reveal absorption peaks corresponding to both the pure green epoxy polymer, GNP, and CNF. The FTIR spectra of the pure polymer exhibit O–H stretching vibrations occurring in the range of 3,000–3,600 cm^{-1}, indicating the presence of hydroxyl groups as evidenced by the FTIR data, which contributes to the existence of a free hydroxyl group (moisture).

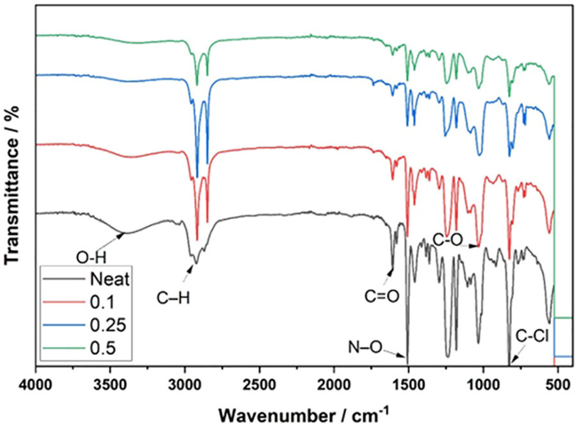

Figure 3: FTIR spectra of different neat and CNF/GNP loadings.

The spectral peaks observed at 2,916 and 2,848 cm^{-1} are associated with the valence vibrations of the –CH bonds within the epoxy ring and the stretching vibrations of the $-CH_2$ groups present in aromatic rings, respectively, which are categorized under alkanes, as the characteristic range for this group spans from 3,000 to 2,840 cm^{-1}, as depicted in Figure 3. The symmetrical stretching vibrations of the $-CH_3$ functional group are discerned at the peak near 2,868 cm^{-1}. The infrared spectra of pure epoxy demonstrate distinct peaks at 1,608, 1,508, and 1,454 cm^{-1}, which correspond to the C–C stretching vibrations of aromatic rings. The peak observed at 1,237 cm^{-1} is indicative of the C–O stretching vibrations characteristic of alkyl aryl ethers. The pronounced vibrational frequencies identified at 1,107 and 1,180 cm^{-1} are ascribed to the robust C–O stretching associated with tertiary alcohols [31]. The spectral peaks observed at 3,445 cm^{-1} are indicative of the stretching vibrations associated with N–H functional groups. With the incremental loading of GNPs and CNF, the peaks corresponding to NH and OH groups (which are overlapping) exhibited a shift toward higher wavenumbers, concurrently with an increase in the peak intensities of hydrogen-bonded carbonyl groups. This visible peak can be attributed to the presence of hydroxyl and carbonyl functional groups located at the periphery of the GNP [32]. The spectrum analysis revealed that the functional groups present within the green epoxy composites exert an influence on the remaining nanocomposites, as evidenced by the absence of newly formed peaks within the nanocomposites [33]. The significant peak detected at approximately 1,500 cm^{-1} is likely a consequence of the incorporation of carboxylic groups onto the primary alcoholic groups present at the surface of CNF, which are consistently observed across all epoxy nanocomposites. This observation substantiates the effective incorporation of these components into the green epoxy matrix [34].

While observing FTIR spectra closely, it can be seen that all the peak values were nearly identical. This implies that there was no new chemical bonding between the polymer and loadings [35].

3.2 Morphological analysis

The micrographs of fractured surfaces of hybrid nanocomposites obtained by FE-SEM [36] are displayed in Figure 4. The neat epoxy image is displayed in Figure 4a, whereas the remaining micrographs (Figure 4(b)–(d)) depict the hybrid nanocomposite materials with varying loadings of CNF/GNP. The morphology of the fractured surface in Figure 4(a) demonstrated the neat epoxy composite's brittle traits. This was due to the material's weak resistance to crack induction and transmission under load, which resulted in low fracture toughness [37]. When 0.1 wt% CNF and 0.1 wt% GNP were loaded as the hybrid nanofillers, there was no restacking of GNP or agglomeration of CNF in the micrographs of the green epoxy composites. Consequently, void forms were eliminated since the evenly distributed CNF restricts the stacking of GNP (Figure 4(b)). An increase in CNF loads also increases the interconnectivity of nanofillers in the matrix. This can be due to the process by which loading quantity modifies the connection of the nanoparticles, leading to improved mechanical characteristics. The fracture surface of 0.1 wt% cracks took additional complicated paths on the well-dispersed fillers on the composites [38]. Thus, no river line marking was perceived, which makes the surface coarser. Hence, the loading of CNF impacts the dispersing ability of GNP into the green epoxy polymer matrix, which has been justified by the synergistic effect of CNF and GNP.

The micrographs of the hybrid green epoxy sample reinforced with 0.25 and 0.5 wt% CNF/GNP demonstrate that GNP restacks into the epoxy polymer matrix due to van der Waals forces, which are caused by the plane-to-plane contact of adjacent GNP [39] (voids are indicated by the arrow in Figure 4(c)). This could be because the use of 0.25 and 0.5% GNP as a reinforcing agent with the same quantity of CNF loading is high. Therefore, the micrographs of both the loadings of green epoxy composites

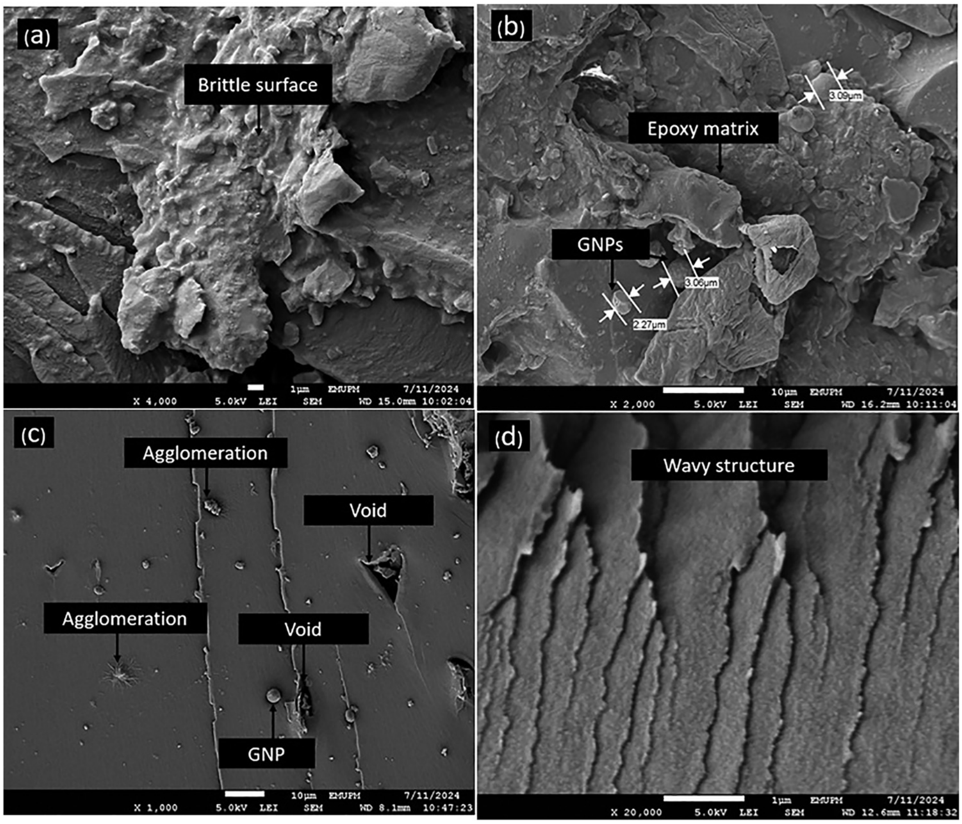

Figure 4: FE-SEM images depicting the fractured surfaces of both neat and nanocomposite materials resulting from tensile testing at varying loadings of GNP/CNF: (a) the neat epoxy composite, (b) a composition containing 0.1 wt% CNF/GNP, (c) a composition with 0.25 wt% CNF/GNP, and (d) a composition exhibiting 0.5 wt% CNF/GNP.

show the formation of agglomerates of CNF and GNP, resulting in stress concentration [40]. These agglomerations generate voids and cavities between the GNP and the green epoxy polymer matrix as well as limit the flow of matrix into the agglomerated GNP, which leads to the formation of waves and other deformations (Figure 4(d)). Poor mechanical and thermal qualities are the outcome of the filler's poor adherence and compatibility with the matrix at high loadings.

3.3 Structural analysis

XRD was used to characterize the amorphous structures of neat and GNP/CNF hybrid nanocomposites. The van der Waals forces of contact between the layers are weakened by the sonication of GNP and CNF, which permits green epoxy molecules to intercalate. Similarly, nanoparticles with an intense tendency to aggregate in epoxy were intercalated with the aid of sonication and magnetic stirring [41]. It is assumed that in nanocomposites, the extremely thin nature of GNP allows them to easily slide and move within the larger spaces between the epoxy polymer chains, essentially "filling in" the gaps due to their small thickness compared to the overall epoxy matrix structure [42]. Figure 5 shows an XRD graph for a neat sample and varying loadings of GNP/CNF-reinforced composites. There is no evident diffraction of GNP seen in the figure in the nanocomposite at GNP contents of 0.1 and 0.25 wt%, which is believed to be associated with a small amount of consistently dispersed GNP embedded in epoxy matrices [43]. A small peak positioned at 26.3° can be observed on the XRD pattern of 0.5 wt% CNF/GNP nanocomposites when the GNP content is 0.5 wt%.

Figure 5 also shows that during sonication, the nanosheets in the green epoxy matrix separated. Two broader peaks with lower intensities were visible in the nanocomposite results. The two nanoparticle interactions with epoxy molecules, which permit the molecules to migrate between the layers, are responsible for these peaks. The two unique peaks can be explained by the presence of individual nanoparticles in different places across the system, while lower peak intensities indicate a major disruption in the nanoparticle arrangements. As a result, XRD data results indicate that each nanoparticle is sufficiently dispersed throughout the epoxy system. The vertical dashed line near $2\theta \approx 26.5°$ indicates the (002) plane of graphene. The calculated interlayer spacing (d_{002}) is approximately 0.336 nm, which is typical for few-layer or graphitic structures.

As CNF/GNP content increases (from H0.1 to H0.5), the peak intensity decreases and broadens and shows disruption in crystalline order, lower stacking regularity of graphene layers, and better dispersion or exfoliation of graphene in the epoxy matrix. All nanocomposites XRD graphs show the relative intensity of wider peaks that indicate how the matching nanoparticles are distributed throughout the epoxy system [33].

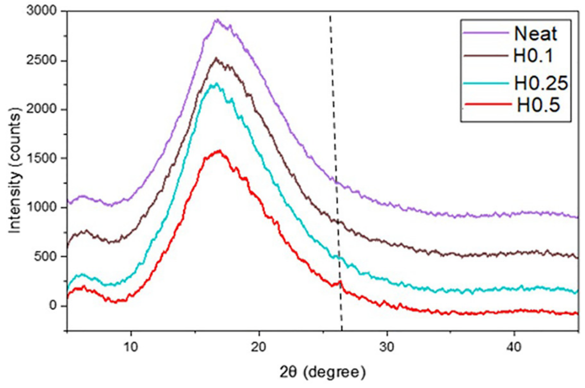

Figure 5: XRD patterns of neat and GNP/CNF hybrid nanocomposites.

3.4 Thermogravimetric analysis

It is apparent that the homogeneous dispersion of nanoparticles in hybrid nanocomposites and the quality of interface interactions between nanoparticles, along with the matrix, determine the thermal and mechanical properties of polymer composites. However, in the hybrid green epoxy nanocomposites, this was not the case, as shown in Figure 6. In the previous published research by Yusuf *et al.* [44],

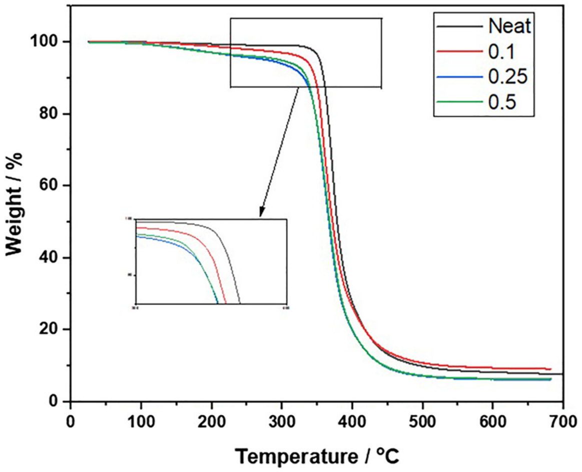

Figure 6: TGA curves for neat and nanocomposites with varying CNF/GNP loadings.

the thermal degradation temperature (T_d) of GNP-reinforced green epoxy nanocomposites was increased as compared to the neat sample. However, the results were not the same in the case of CNF/GNP hybrid green epoxy nanocomposites. A major possible reason is the addition of CNF. CNF caused the agglomeration of CNF, which led to limited compatibility of GNP and CNF particles in green epoxy. Due to the stronger links between the clumping nanoparticles and the green epoxy resin, particle-to-particle interactions become high. As a result, molecular mobility is not restricted, which lowers the decomposition temperature [38].

While neat epoxy composites were stable up to 334°C, hybrid green nanocomposites with 0.1 wt% CNF/GNP filler loading exhibit a lower temperature of 322°C and a decreasing trend with increasing filler loading (Figure 5). The neat epoxy composite's increased thermal stability can be attributed to the green epoxy resin's increased cross-linking [45]. However, out of three loaded samples, a sizable amount of thermal stability improvement occurred at 0.1 wt% CNF/GNP compared to different loadings that were caused by the evenly distributed GNP. Adding CNF along with GNP, which showed a decreasing trend for the degradation temperature, is because of the CNF's susceptibility to trapping moisture, which can accelerate thermal degradation and reduce the composite's thermal stability [46]. Overall, it can be concluded that the decrease in the thermal properties at 0.1, 0.25, and 0.5 wt% CNF/GNP loading, in contrast to the neat sample, may be caused by the addition of CNF [47].

The primary reasons for the decrease in thermal stability are the susceptibility of CNF to trap moisture. This moisture can accelerate thermal degradation, leading to a reduction in the overall thermal stability of the composite [48]. The presence of moisture can create pathways for thermal degradation to occur at lower temperatures, which negatively impacts the thermal performance of the material. The higher loading levels of CNF and GNP can lead to non-uniform dispersion and agglomeration within the composite. This agglomeration can create voids and weak points in the material, which may facilitate thermal degradation. As the filler loading increases, the effectiveness of the reinforcement diminishes, resulting in a decrease in thermal stability. While the neat epoxy composite exhibits increased thermal stability due to enhanced cross-linking, the addition of CNF and GNP at certain loading levels can disrupt this cross-linking network. The study explores that the hybrid green nanocomposites with 0.1 wt% CNF/GNP showed a lower thermal degradation temperature compared to neat epoxy, suggesting that the addition of these fillers may interfere with the optimal cross-linking required for maintaining thermal stability. As the loading of CNF/GNP increases, there is a corresponding decrease in thermal properties. Specifically, the thermal degradation temperature (T_d) of the composites decreases with higher filler loading, which suggests that the composite's ability to withstand heat diminishes as more filler is added [49].

3.5 Mechanical testing

The hybrid nanocomposite's tensile strength was influenced by a number of variables, including the filler's form, availability of functional groups, loading in the matrix, state of dispersion, aspect ratio, and interaction with the matrix [50]. A key component in the fabrication of polymer composites was the filler's homogeneous dispersion (GNP and CNF). The characteristics of the green nanocomposite were negatively impacted by the high loading of GNP and CNF. To prevent agglomeration and the formation of GNP and CNF lumps, which were the cause for the decline of the tensile strength, an effective dispersion state of the fillers in epoxy was required. The load transfer capability between the fillers and the polymer matrix is reduced because of these lumps acting as stress concentration spots, which also limit and diminish the interfacial bonding of GNP/CNF and epoxy [51].

Figure 7(a) shows the behavior of CNF/GNP green epoxy hybrid nanocomposites and neat green epoxy. At 0.1 wt%, the maximum tensile strength of 81 MPa was observed. This is because the filler's loading of 0.1 wt% is very small and does not exceed the percolation threshold, indicating no excess loading. As a mechanical support inside the polymer, CNF offers a linked cellulosic network. The presence of amorphous domains along nanofibrils and the high aspect ratio of CNF contributed to its flexibility in forming the network [32].

When compared to the neat sample of 62 MPa, tensile strength improved to 23.46% at 0.1 wt% CNF addition. As the loading increased to 0.25 and 0.5 wt%, the value decreased from 35 to 25 MPa. A strong filler–epoxy interaction can efficiently produce fibrillar interphase areas. There might be a higher volume fraction of fibrils because of improved stress in the interphase area as compared to clean epoxy, which would improve the mechanical properties [52].

As fillers were loaded above 0.1 wt% loadings in the matrix, the tensile strength was further reduced; this was most likely due to the potential for accumulation to occur. Agglomerated areas and fillers' hydrophilic nature would operate as stress concentration points, causing failure or early break and lowering tensile strength [53].

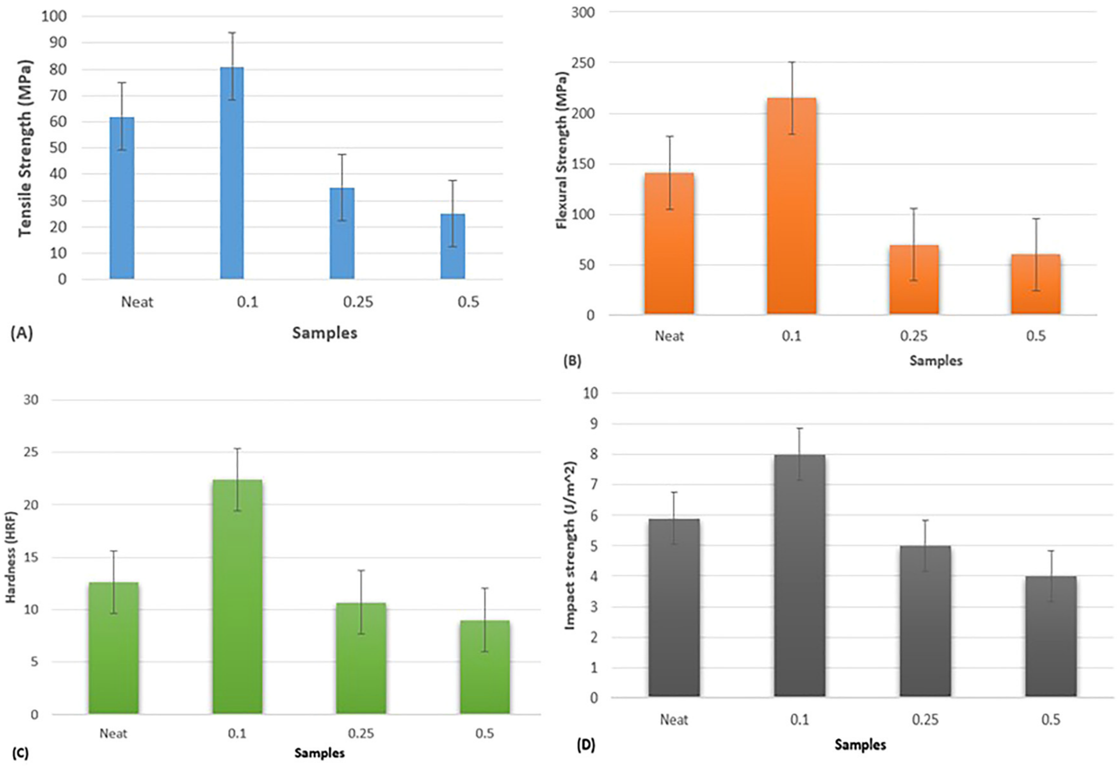

Figure 7: Mechanical testing of different CNF/GNP loadings: (a) tensile test, (b) flexural test, (c) hardness test, and (d) impact test.

Flexural strength was initially increased by the loading of GNP/CNF from 141 to 215 MPa in the matrix, but when the loading of GNP/CNF exceeded 0.1 wt%, a sharp decrement in flexural strength was recorded at 0.25 and 0.5 wt% from 70 to 60 MPa, as shown in Figure 7(b). The interphase that forms between the fiber and the matrix, which is anticipated to be stiffer than the epoxy, is responsible for the increase in flexural strength. This results from the uniform dispersion of hard GNP, which stiffens it [54]. At the same time, CNF also acted as an effective hardening mechanism, acting as bridges that increase the energy required for crack propagation [55]. However, a further increase in GNP/CNF loading led to a sharp reduction in flexural properties. It was due to the agglomeration of GNP/CNF in the hybrid composite. These clusters may serve as stress concentrators, hastening the propagation of interface-generated faults. These flaws may enlarge beyond the threshold crack size, leading to the failure of hybrid green nanocomposites [56].

It was significant to note that the reduction in strength properties relied on the hybrid CNF/GNP composition, in which the higher the GNP amount, the lower the strength [57,58]. Due to this reason, the reduction in flexural properties was observed. The large surface area and stiffness of GNP have been linked to a predicted decrease in strength, particularly at higher loads, which increases the stiffness in the sample [59]. Another possibility is that GNP aggregation would serve as a steric barrier, obstructing the flow of the polymer into them and causing gaps and holes to emerge between the GNP and epoxy [60]. Hence, it can be concluded that, due to the significant void content and nonuniform dispersion of GNP/CNF, flexural characteristics decreased as the GNP loading increased. When force was applied, the densely packed layer of GNP generated enhanced internal delamination, resulting in low flexural strength [61].

The hardness increased as GNP/CNF was loaded into the matrix, as shown in the hardness of the neat sample was noted as 12.6, which increased to 22.4 when 0.1 wt% of CNF was loaded, and approximately 77% of the total increment was recorded. The maximum hardness value of 0.1 wt% loading of GNP/CNF was recorded because of uniform dispersion and increased surface area of GNP/CNF in the matrix (Figure 7(c)). Proper distribution of fibrils in the matrix produces this effect, which is due to stronger interfacial bonds between the CNF and polymer matrix, which tend to decrease void contents [62]. Additionally, the toughening mechanisms that lead to the gain in hardness are induced by the presence of GNP in the epoxy matrix. GNP functions as a hard nanofiller, which is evenly dispersed throughout the epoxy matrix, giving the nanocomposite materials their excellent hardness and favorably affecting other mechanical properties as well [63].

Nevertheless, as the loading is further increased, from 0.25 to 0.5 wt% of GNP/CNF, a decrease in hardness is noticed. Poor dispersion is the main cause of the hardness loss, which causes voids and porosity to form within the nanocomposite, which in turn causes a site of stress concentration and overlapped GNP slippage [64]. The addition of CNF lowers the mechanical properties because the intrinsic van der Waals forces between the polymer matrix and CNF are weaker, which lowers the hardness of the composite overall [65].

The impact strength of the composite material is characterized by the polymer's capacity to absorb energy when subjected to fracture propagation and dynamic impact loading within the matrix [66]. The impact strength value was lower for the neat sample than for the neat epoxy composites because the neat sample was brittle (Figure 7(d)). When GNP/CNF was added, the impact strength of nanocomposites increased, and the recorded impact strength was maximum at 0.1% GNP/CNF loading. The 0.1 wt% loading of GNP/CNF produced the highest impact strength value at 8 J, which was 35.5% greater than the neat sample. The uniform distribution of GNP/CNF throughout the matrix was identified as the cause of the impact test's high value. An important factor in raising the nanocomposite's fracture toughness is the interfacial contact. The nanocomposite's fracture toughness is further enhanced by the large surface [67]. Rapid fracture propagation starts when an impact occurs on a composite material. When the CNF/GNP and epoxy matrix have significant interfacial contacts, CNF in the composite absorbs impact energy and prevents fracture from spreading [38]. On the other hand, if the loading exceeds 0.1 wt% GNP/CNF, the matrix will not be distributed evenly. A larger weight percentage of GNP is linked to a decrease in fracture toughness due to the non-uniform dispersion of CNF/GNP in the matrix [68]. In a nutshell, the mechanical properties of nanocomposites are not exclusively dictated by the weight proportion of GNP/CNF. The dispersion technique, dispersant use, and functionalization can be further influential factors if applied. Utilizing an organic solvent is a vital factor in determining the improvement in mechanical properties.

4 Conclusions

This study examines the effects of low loading of CNF and GNP reinforcement within hybrid green epoxy nanocomposites, wherein the green epoxy resin comprises 28% carbon content sourced from biomass. The significant peak identified in the FTIR spectra at approximately 1,500 cm^{-1} may be ascribed to the effective incorporation into the green epoxy matrix. Various loading profiles were observed by TGA. The thermal degradation temperature (T_d) of the CNF/GNP curve with 0.1 wt% loading displayed a slight increase toward elevated temperatures, suggesting that the lower loading of CNF/GNP conferred enhanced thermal stability. An improvement in mechanical properties was observed at the 0.1 wt% CNF/GNP addition in comparison to the unmodified green epoxy. An increase in mechanical characteristics was observed at the 0.1 wt% CNF/GNP addition relative to the standard green epoxy sample. Conversely, elevated loadings of CNF/GNP resulted in a subsequent reduction in the measured values, a phenomenon corroborated by FE-SEM analysis and attributed to the presence of non-uniform dispersion, void formation, and the agglomeration of CNF/GNP. The specimen exhibiting the maximum tensile strength, which surpassed the control sample by 77% and demonstrated a 23.46% enhancement in hardness, was incorporated with 0.1 wt% CNF/GNP.

While this investigation reveals encouraging advancements in the domain of hybrid green nanocomposites, it is imperative to acknowledge that the scope of this research was confined to a limited spectrum of CNF/GNP loadings in green epoxy and that the primary focus was on the thermal, mechanical, and morphological properties. Future research could explore the broader applicability of these findings to various green polymer matrices and emphasize areas such as electrical properties, chemical resistance, environmental implications, and the potential industrial applications of green hybrid nanocomposites.

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