

Intrinsic Response of surface modified Calotropis Gigantea Fiber and Nelumbo nucifera filler hybrid composite

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Research Article

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Posted Date: March 5th, 2024

DOI: <https://doi.org/10.21203/rs.3.rs-3890643/v1>

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Version of Record: A version of this preprint was published at Waste and Biomass Valorization on August 17th, 2024. See the published version at <https://doi.org/10.1007/s12649-024-02687-x>.

Intrinsic response of surface modified *Calotropis Gigantea* Fiber and *Nelumbo nucifera* filler hybrid composite

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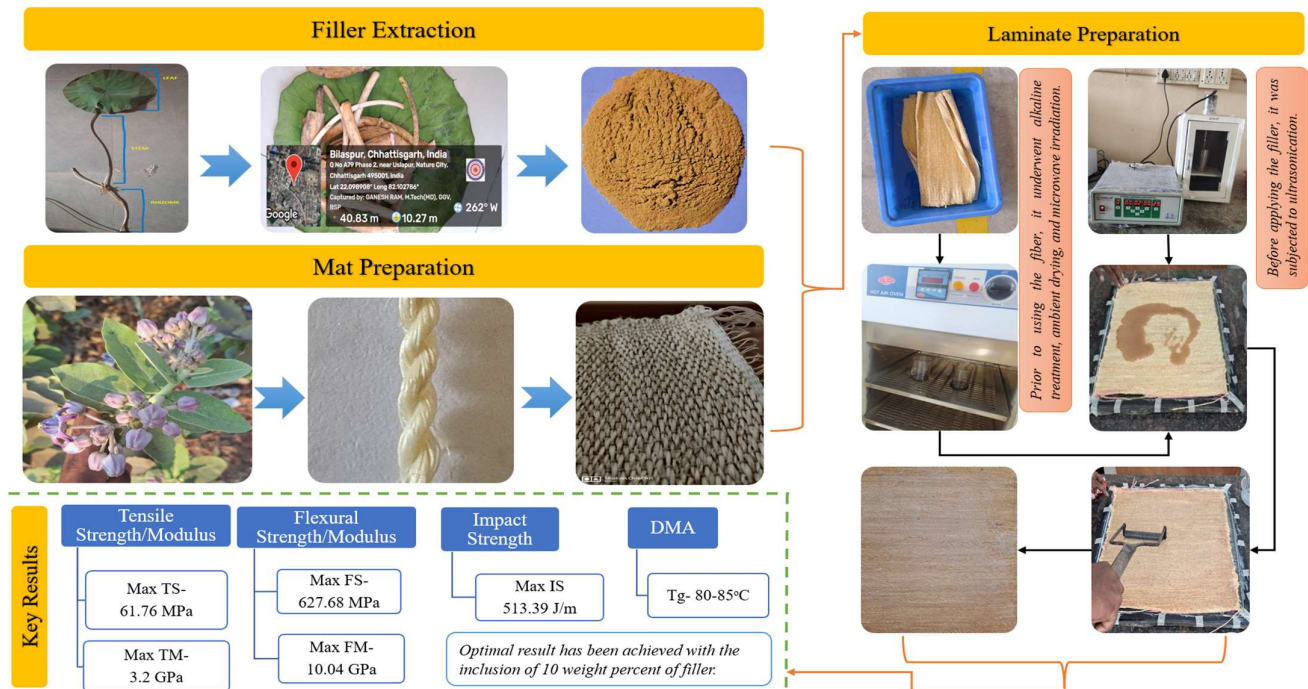
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Abstract— The present study used *Calotropis Gigantea* fiber, which has minuscule significance in vegetation value, and waste *Nelumbo Nucifera* filler for the production of polymer composites. The *Calotropis gigantea* yarn fiber is twisted into Two-Ply Yarn and subsequently woven into mats, the mat underwent both NaOH treatment and microwave irradiation in order to enhance their compatibility with epoxy. This work aims to examine the mechanical and dynamic properties of a hybrid composite material consisting of a Chemically and Microwave Treated *Calotropis Gigantea* (CMTCG) fiber combined with *Nelumbo nucifera* (NN) infills. The objective of this study is to investigate the possible improvement of the mechanical and dynamic characteristics of polymer composites by developing composite samples that include different weight proportions of NN fillers in CMTCG-reinforced Epoxy composites. The assessment of the mechanical properties of several combinations of CMTCG with NN infills was conducted utilizing established testing methodologies, including tensile, flexural, impact, and Dynamic Mechanical Analysis (DMA) to assess the viscoelastic nature of the composites. In addition to this, a Cole-Cole curve is plotted by utilizing the findings of the DMA to analyze the effect of particle impregnation and the anticipation of the heterogeneity of the composite material at different loadings of fillers. The current investigation revealed that a fiber loading of 10 weight percentages represented the optimal threshold, and any loading value resulted in a noticeable decline in all characteristics.

Graphic Abstract



Keywords—*Calotropis gigantea*, *Nelumbo nucifera* filler, Two-ply yarn, Dynamic Mechanical Analysis (DMA), Cole-Cole Plot.

Statement of Novelty

The *Calotropis gigantea* (milkweed) plant has less vegetation importance. The *Calotropis gigantea* plant and *Nelumbo nucifera* are found in several parts of the world. No previous study has been attempted on the synergistic combination of chemical and microwave-processed twisted two-ply *Calotropis gigantea* fiber and *Nelumbo nucifera*-infused hybrid polymer composite. This study examines the mechanical properties, such as tensile, flexural, and impact strength, and analyzes the dynamic response characteristics of the developed hybrid composite.

1. Introduction

In light of the implementation of environmental legislations and the growing demands from consumers, manufacturing industries are being driven to explore alternative materials that are both renewable and sustainable. The current legislative landscape and increasing demands are creating a strong impetus for the exploration of innovative materials that can effectively substitute commonly utilized non-renewable reinforcing materials such as glass and carbon [1]. Polymer composites, which are made using a range of reinforcing elements comprising natural fibers, have drawn increasing attention in the utilization of natural fiber-reinforced polymer composite materials, encompassing both industrial applications and fundamental research in recent decades [2]. Natural fibers have garnered significant interest due to their numerous advantages compared to conventional fibers. These advantages include low density, cost-effectiveness, environmental friendliness, biodegradability, and high specific mechanical performance. These composites find applications in various industries such as automobiles, the aerospace sector, commuter train coaches, the military, construction and building industries, packaging, and consumer products. Composite materials are utilized in the production of devices for storage, electrical devices, furnishings for homes, the transportation sector, and toys [3]. Mohammed et al. [4] conducted a comprehensive analysis of the utilization of natural fiber-reinforced composites across diverse industrial sectors.

The successful exploitation of various surface alteration methods for natural fibers has been achieved by researchers. These methods include physical and chemical techniques, as well as their combination. The primary objective of these modifications is to enhance the adhesion between the natural fibers and the continuous matrix phase in composite materials. Various chemical treatments can be used to modify the surface properties of fibers. These treatments include alkali, silane, acetylation, benzylation, peroxide, maleated coupling agents, isocyanate, sodium chlorite, and etc. [5]. The alkali treatment stands out among the various chemical treatments due to its economic advantages, primarily its low cost and ease of execution. The alkali treatment is a process that entails the application of a potent alkali solution to the cellulose fibers. This treatment induces fiber swelling and enhances their surface functionality. This swelling response does not cause cellulose breakdown but rather enhances fiber surface area [1]. According to research, alkali-treated natural fiber possesses a greater surface area [6] and displays enhanced mechanical properties as a result of a better compatibility with matrices [7].

Microwave irradiation represents an alternative method for surface modification of natural fiber. This method uses microwave frequency electromagnetic radiation to treat the fibers. The application of this process can initiate a range of physical and chemical modifications on the surfaces of the fibers, resulting in enhanced properties. The utilization of microwave irradiation presents a highly promising approach for the surface transformation of natural fibers [8]. The implications of the effect of microwave irradiation on empty fruit bunch (EFB) fiber were investigated in a study conducted by B. Bakari et al [9]. The tensile strength experienced a significant increase from a value of 142.40MPa to a value of 187.25MPa after subjecting the fiber to microwave irradiation for a duration of 10 minutes. Extensive exploration was

conducted on numerous novel natural fibers, with the aim of facilitating their application as reinforcements for polymer composites. Additionally, efforts were made to develop materials that are environmentally sustainable. In this context, the authors attempted to investigate the viability of including *Calotropis* bark fiber mat as a novel material for reinforcement composites. *Calotropis gigantea* (CG), a member of the Apocynaceae family, is commonly known as 'Arka' and 'Mudar' in Sanskrit and English, respectively [10]. The choice of CG fiber in this study is based on its abundant availability in India and its barely noticeable importance in vegetation [11]. Despite the fact that CG fiber has considerable reinforcing potential, only limited studies are available on CG fiber-reinforced composites, and even then, filler experiments [12, 13] are seldom documented. Impregnation of fillers has emerged as a widely adopted technique for augmenting the thermomechanical properties of composite materials. The fillers are collected from plant extracts and biological waste materials and blended into matrices to significantly ameliorate their overall performance. Problems with fiber position, delamination, length of fiber shortcomings, and intricate production may all be mitigated with the use of filler incorporation [14–16]. The form, size, and percentage of filler, including the extraction process and treatment methods used on fillers, all have a substantial impact on the efficacy of binding inside interfaces and the characteristics of composite materials [17, 18]. Dynamic Mechanical Analysis (DMA) is an extensively used approach to ascertaining the temperature-dependent variations in the viscoelastic characteristics of composite materials [19]. This research paper aims to investigate the fabrication of polymer composites using alkali-treated and microwaved *Calotropis gigantea* (CMTCG) fiber mat and incorporating *Nelumbo Nucifera* filler. Various quantities of *Nelumbo nucifera* filler were introduced into epoxy resin to alter the properties of the composites produced. These modified composites were then compared to pure CMTCG fiber-reinforced polymer composites.

2. Material and Methods

This study focuses on the utilization of CG bast fiber for the purpose of strengthening fiber composites. The specific quantities of epoxy and hardener, expressed in weight percentage, are determined as necessary components for achieving successful bonding of the fibers. This study also involves the incorporation of *Nelumbo nucifera* filler in the production of hybrid composites.

2.1 *Calotropis gigantea* (CG) fiber

Calotropis gigantea, commonly known as giant milkweed, is a prevalent weed species found in various Asian countries including India, China, Malaysia, Pakistan, Thailand, and Indonesia. The plant *Calotropis gigantea* possesses toxic properties that can lead to various adverse effects, including vomiting, diarrhea, dilated pupils, and fatal outcomes [20]. The neat CG fiber was sourced from the southern part of Tamil Nadu, India. The fiber is a highly refined and versatile material that can be effectively employed for various practical applications. The collected fiber was effectively spun using a cotton spinning machine and subsequently woven into mats. The *Calotropis gigantea* plant is depicted in Figure 1(a), while Figure 1(d) displays the fiber and prepared mat.

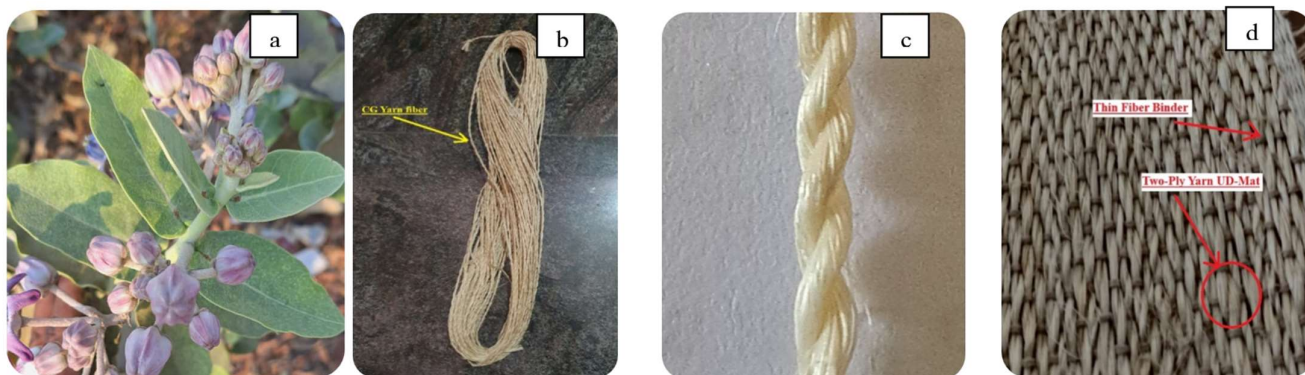


Fig.1 (a) *Calotropis gigantea* (Mudar, milkweed) plant (b) CG fiber yarn (c) Twisted-two-Ply yarn (d) CG UD fiber mat

2.2 *Nelumbo Nucifera* (NN) filler

The filler is derived from the waste *Nelumbo nucifera* rhizome, which was extracted from a pond in nearby Bilaspur, Chhattisgarh, India, positioned at Latitudes 22.193702° and longitudes 82.109578°, depicted in Figure 2(a). The extracted plant material has undergone a water-cleaning process to remove extraneous muddy materials and was then exposed to sunlight for 5 days to dry. Afterward, the mechanical retting process is carried out to extract fine powder by utilizing a 500-watt-powered domestic grinding machine. The filler material was stored in a cool and dry environment within a tightly sealed plastic container to prevent from air and moisture.

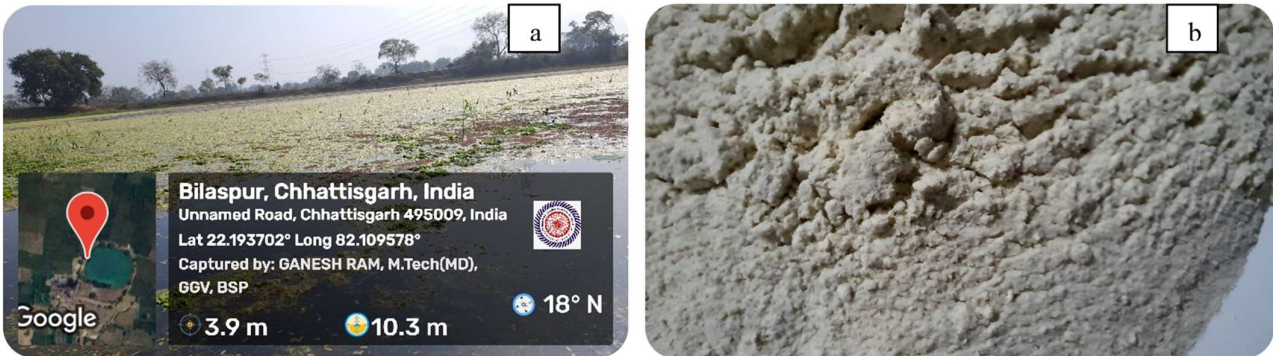


Fig.2 Preparation of NN filler (a) plants derived from submerged fields (b) NN filler

2.3 *Matrix material*

The use of epoxy in composites provides exceptional moisture barrier properties. The material exhibits strong bonding characteristics with fibers during the fabrication process of fiber-reinforced composites. Hardeners are known to undergo reactions with epoxy resins, resulting in notable effects on the final characteristics of the cured epoxy resin system such as gel time, and viscosity [21]. The epoxy resin bisphenol A (LY556) and the hardener triethylenetetramine (HY951), were purchased from REACHEM Laboratory Chemicals Private Limited, Chennai, Tamilnadu, India. The prescribed ratio of the epoxy resin and hardener for curing was chosen as 10:1 by weight. The epoxy system was chosen due to its appropriateness for the production of industrial composites.

2.4 *Sodium hydroxide and distilled water*

The Alkali treatment of *Calotropis gigantea* fiber involves the utilization of sodium hydroxide and distilled water. The aforementioned treatments facilitate the elimination of impurities and the improvement of fiber properties, rendering it appropriate for utilization in polymeric composites. Distilled water, and Sodium Hydroxide (NaOH -M.W.40.00) Pellets were kindly supplied from REACHEM Laboratory Chemicals Private Limited, Chennai, Tamilnadu, India.

2.5 *Twisted Two-Ply Yarn of CG fiber*

Single yarns undergo Z-directional twisting to prepare Two-ply yarns as depicted in Figure 1(c), followed by weaving into mats, prior to their use as reinforcement in composite manufacturing and this mat fabrication was carried out by **Vedhanayaki Fabs**, a well-known mat manufacturer located in Bhavani, Tamilnadu, India. The process of intertwining two yarns results in a yarn that has enhanced strength and improved tensile characteristics compared to a single yarn [22, 23]. The interlacing of the unidirectional two-ply yarns (Arranged next to one another in order to create a UD mat and a thin, single layer of fibers used as a binder that holds a mat together as shown in Figure 1(d) enhances their structural strength and facilitates more efficient dispersion of the fibers throughout the composite material [20].

2.6 Chemical and Microwave treatment of the *Calotropis gigantea* fiber mat (CMTCG)

The CG fiber underwent alkaline treatment and microwave irradiation prior to application as a reinforcing agent in a polymer composite. In order to perform the alkaline treatment process on the CG fiber, it is essentially immersed in a 5% W/V alkali aqueous solution for a duration of 24 hours. Subsequently, the soaked CG fiber is subjected to ambient air for a period of 24 hours to facilitate drying. Following this, the fiber is exposed to microwave radiation at a temperature of 80 degrees Celsius for one hour. The sequential steps are illustrated in Figure 3. During the alkali treatment, the hydroxyl group in the fibers undergoes a process of degradation, wherein they interact with the hydrogen of the alkali (H-OH) and subsequently exit the fiber structure. The residual reactive molecules establish cell-O-Na groups by linking the cellulose chain structures [5]. As a result, the hydrophilic hydroxyl groups undergo reduction, leading to an enhancement in the moisture resistance property of the fibers. These treatments also promote the removal of partially amorphous components such as waxes, lignin, and hemicellulose, altering the surface to a rough texture. Furthermore, it has the capability to decrease the diameter of the fiber, resulting in an increase in the aspect ratio, which is defined as the ratio of length to diameter. This results in an increased effective surface area of the fiber, promoting better compatibility with the matrix. The treatment significantly enhances the mechanical and thermal properties of the composite [5, 13].



Fig.3 Activities conducted during chemical treatment and microwave irradiation (a) NaOH -aqueous solution (b) fiber soaked in aqueous solution (c) open air-drying (d) microwaved for one hour

3. Experimental Procedure

3.1 Fabrication of laminate

To prepare the composites, NN fillers were combined with resin at various weight percentages (0, 5, 10, and 20 wt%). The mixture was subjected to manual stirring using a glass rod, followed by ultrasonication for a duration of 30 minutes using an ultrasonication probe instrument to achieve a homogeneous dispersion of the NN particles. The surface was coated with wax on the surface of the glass mold. The purpose of using wax is to ensure a smooth release of the composite material. The CMTCG fiber mat were positioned onto the mold, and subsequently, the sonicated mixture of epoxy and NN filler of different weight percentages was spread on the surfaced CMTCG fiber mat. After that, the cylindrical roller of 15 mm diameter was rolled on the surface, as illustrated in Figure 4. For the uniform distribution of the epoxy filler mixture on the CMTCG fiber mat, this cylinder roller was employed to roll the mixture. The aforementioned procedure is iterated for the second layer of the laminate. In order to mitigate the occurrence of air pockets, a load was exerted on the top portion of the glass mold. The sample was permitted to undergo a curing process for a duration of 24 hours. After making of NN filler impregnated CMTCG fiber reinforced hybrid polymer composite laminate structures, the samples were extracted and subsequently utilized for testing purposes. The combination of CMTCG fiber mat, epoxy, and filler for the investigation, specimen samples is fabricated according to the scheme outlined in Table 1.

Table 1 Designation of the prepared laminates

S.No.	Sample Designation	Description
1	XG	Epoxy with CMTCG fiber mat
2	XG5N	Epoxy with CMTCG fiber mat and 5 wt% of NN infill
3	XG10N	Epoxy with CMTCG fiber mat and 10 wt% of NN infill
4	XG20N	Epoxy with CMTCG fiber mat and 15 wt% of NN infill



Fig.4 Activities carried out during laminate preparation (a) Glass mold (b) Incorporation of NN filler-epoxy mixture at a varying weight percentage (0, 5, 10, and 20 wt%) (c) Disseminating the mixture (d) A roller utilized during hand layup process (e) prepared laminate

3.2 Specimen Preparation

The composite laminate boards were sliced into standard test specimens. The cutting process involved using motorized saw tools and notch cutter tools. All samples were prepared in accordance with ASTM specifications as illustrated in Table 2. The test specimens were stored in a controlled environment for a duration of 48 hours prior to commencing the test. The tests were conducted in a standard laboratory environment.

3.3 Mechanical testing

The test specimens underwent a series of tests in accordance with the standards set by ASTM. The test specimens conforming to ASTM standards are presented in Table 2.

Table 2 Specimen Test Standards

S.No.	Test Type	Standard	Dimensions
1	Tensile	ASTM D3039-76	250mm x 25mm x 2.5
2	Flexural	ASTM D790	154mm x 13mm x 3mm
3	Impact (Izod)	ASTM D256	63.5mm x 12.7mm x 3.2mm
4	DMA	ASTM D4065	50mm x 10.8mm x 4.8mm

3.3.1 Tensile Test

Tensile testing is widely regarded as one of the primary mechanical characteristics investigations that can be performed on a material. The tensile strength as well as the elongation properties of the material can be determined when it is subjected to tension. The tensile test is typically performed following the guidelines outlined in ASTM D3039. The testing specimen possesses dimensions of 250 mm in length and 25 mm in width. The item was securely clamped within the jaws of a digital universal testing machine (UTM). The composite's tensile strength and modulus were measured based on the stress-strain diagram.

3.3.2 Flexural Test

Flexural testing is a widely used technique for determining the flexible or bending properties of a specific material. The flexural test is performed following the guidelines outlined in ASTM D790. The dimensions of the test samples used in this study are 154 mm x 13 mm x 3 mm. A load of 10 kilonewtons (kN) was applied, while the cross-head speed was set to 1 millimeter per minute (mm/min).

3.3.3 Impact Test

The evaluation of a composite material's ability to withstand sudden fluctuating loads or impacts is typically performed by conducting an impact test on a specimen. The ASTM D256 standard is commonly employed to perform impact measurements on composite specimens.

3.3.4 Dynamic Mechanical Analysis (DMA)

The elastic modulus of a material is a measure of its performance in a given real-world application. When subjected to heat and surpassing the glass transition temperature (T_g), the polymeric material undergoes a transformation from a rigid and glass-like state to a more flexible and rubbery state, this phenomenon is referred to as the viscoelastic properties of the material. This transition leads to a decrease in the material's modulus. The occurrence of a drop in modulus (stiffness) can potentially give rise to significant difficulties. DMA is a technique that offers valuable insights into the viscoelastic properties of both unfilled and filled polymers across various time scales. The interfacial interactions within filled polymers are crucial in determining their overall bulk characteristics and performance. In the context of Dynamic Mechanical Analysis (DMA), the elastic storage modulus (E') and the loss modulus (E'') are determined by analyzing the response of the material. The utilization of various moduli enables enhanced material characterization by facilitating the examination of its energy storage capacity (E') and energy dissipation capability (E'') due to the elastic and viscous behavior of the material, respectively, and the damping ratio, $\tan(\delta) (= \frac{E''}{E'})$. The estimation of the T_g (glass transition temperature) of the sample can be determined by analyzing the peak values of the loss modulus, or $\tan(\delta)$ [24–27].

3.3.5 Cole-Cole Plot

A Cole-Cole plot of Dynamic Mechanical Analysis (DMA) is a graphical representation of the elastic and viscous properties of a material as a function of frequency. The plot typically comprises a horizontal axis that represents the elastic modulus (E') and a vertical axis that represents the loss modulus (E''). The tangent, which is the ratio of the viscous modulus to the elastic modulus, serves as an indicator of damping ratio $\tan(\delta)$. The Cole-Cole plot provides valuable insight into a material's viscoelastic properties [28]. According to Y. Karaduman et al. [29], The utilization of Cole-Cole plots has been employed to ascertain the level of homogeneity present in composite materials. These diagrams analyze the structural modifications made to polymers due to the addition of reinforcements. The homogeneous polymer structures exhibit a well-defined semi-circular diagram encompassing from E'' to E' . The inclusion of fibers in the matrix results in the heterogeneity of the system, which is indicated by imperfect semicircular curves.

4. Result and Discussion

4.1 Tensile test

In the context of materials, tensile strength (TS) pertains to the maximum stress experienced during stress-strain studies at which the sample breaks. Table 3 and the Figure 5 presents a comprehensive depiction of the tensile strength (TS), elongation at break, and tensile modulus values for each combination. The data presented in Table 3 indicates that the Tensile strength of XG10N laminate exhibited the highest value of 61.76 MPa, which is a 57.19% increase compared to laminate XG without the addition of NN fillers. Furthermore, Young's modulus of 3.2 GPa of the XG10N sample also exhibited a 56.1% increase compared to XG laminate. This considerable gain in elastic modulus can be attributed to the superior stiffness of the CMTCC

fiber and NN fillers [30]. The inclusion of 10 wt% of NN fillers in the laminate results in a substantial improvement in both tensile strength and modulus when compared to other laminates. This indicates that the mechanical characteristics of the composite are enhanced with the addition of these fillers. The study suggested that it has significance for the advancement of nonstructural components and low load-bearing members such as cabinets and two-wheeler brake levers, etc., as it showcases the possibility of enhancing the strength and stiffness of *Calotropis gigantea* composites via the integration of these fillers. The experimental findings indicate that the maximum load capacity of the *Calotropis gigantea* composite material, when reinforced with 10 wt% NN, is measured to be 5050 N. This value represents a 6.8% increase compared to the load capacity of the pure CMTCG laminate without the inclusion of NN. However, the lower tensile strength and Young's Modulus is reported in the laminates XG5N, this may be ascribed to the potential disruption produced by the existence of NN particles, which may have disturbed the arrangement of CMTCG fibers and created flaws within the matrix, which may serve as stress concentrators. The mechanical properties of composite materials are often influenced by morphological factors, including the dispersion and distribution of fibers/fillers within the polymer matrix, as well as the compatibility among the polymer and reinforcements. A large amount of NN content, namely XG20N, reported a decreased value of tensile strength and modulus, these decreases may be attributed to the phenomenon of particle agglomeration. These agglomerates behaved as regions of stress concentration too, increasing the probability of catastrophic failure of specimen samples with a comparatively small value of tension [31, 32]. Furthermore, it is possible that the uptake of water by the cellulose fillers might have resulted in alterations to the geometry of the composites, perhaps leading to a decrease in their mechanical performance [33]. The current study yielded a maximum tensile strength of 61.76 MPa, surpassing the tensile strength of the jute polymer composite filled with padauk (43 MPa) [34], the luffa polymer composite with wood powder impregnation (23.9 MPa) [35], and approaching the tensile strength of the Indian mellow polymer composite with added wood powder (65 MPa) [36].

Table 3 Tensile Test Result of CG-NN/Epoxy Composites

S. No.	Sample	Tensile Strength (TS) (MPa)	Young's Modulus (GPa)	% Elongation (EB)
1	XG	39.29	2.05	2.792
2	XG5N	31.88	2.05	1.772
3	XG10N	61.76	3.2	2.092
4	XG20N	35.54	1.73	2.98

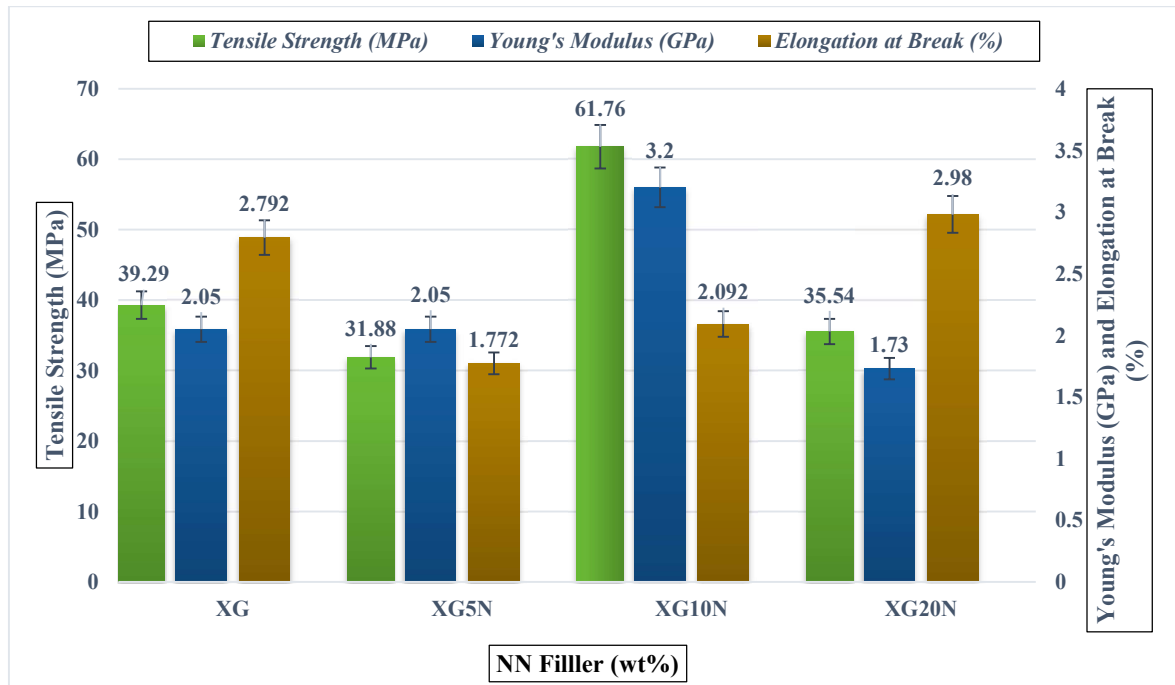


Fig.5 Tensile test result of the CMTCG reinforced with NN infill laminates

4.2 Flexural test

Flexural behavior is characterized by a mix of compressive and tensile behavior. The measurement of flexural strength, also known as bending strength, can be conducted by utilizing a 3-point flexural testing procedure. The flexural strength values of the composites that have been developed in various combinations are presented in the Figure 6. The test findings clearly indicate that the XG10N composites surpassed the performance of the other sample combinations and exhibited the highest ultimate flexural load of 4500N, flexural strength of 627.68MPa, and Flexural Modulus of 10.04GPa. The flexural strength shown in the current study exhibited a much greater magnitude compared to previous research. The researcher, J. Nagarjun et al. [37], observed a maximum flexural strength of 167.47 MPa for date seed powder at 3 wt% loadings, and in another study, T. P. Sathiskumar et al.[38], observed a flexural strength of 80 MPa for snake grass polyester composites. The selection of fibers and the technique of reinforcing are crucial factors in determining the strength of the material. The incorporation of NN filler into an alkaline-treated CG fiber-epoxy matrix system exhibited enhanced interfacial bonding. The factor contributing to the enhancement of flexural strength and modulus is the inclusion of fibers and fillers, which serve as an inherent toughening mechanism by virtue of the presence of NN fillers along the crack propagation path. Therefore, fillers are able to endure increased bending stress transfer, resulting in improved outcomes. The authors, A. H. Elango et. al., [7] demonstrated a comparable pattern in their study on Nelumbo nucifera-reinforced epoxy composites. They observed that higher filler percentages yielded improved outcomes.

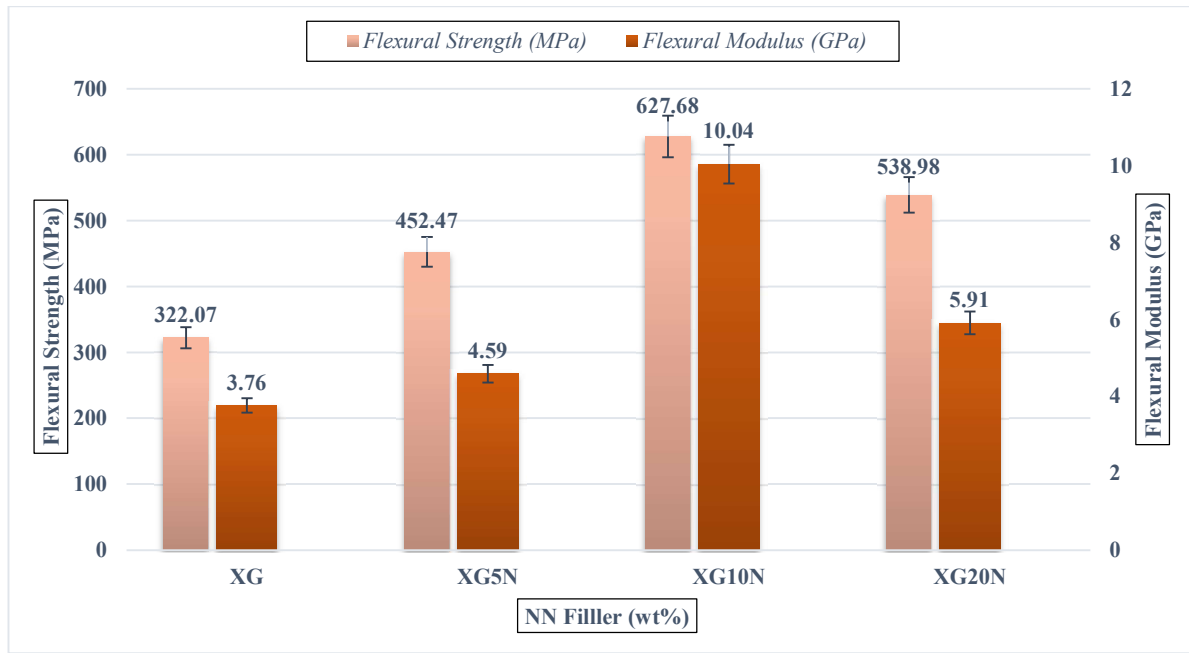


Fig.6 Flexural strength and Modulus of the CMTCG reinforced with NN infill laminates

4.3 Impact test

The test specimen's impact strength is a critical characteristic that influences its capacity to endure abrupt shocks or impacts in practical applications. The measurement of the specimen is often conducted by the application of a controlled impact force, followed by an analysis of the resulting deformation or fracture. The experimental findings indicate that the incorporation of NN filler up to a weight percentage of 10% resulted in an increase in the impact strength of the test. Notably, specimen XG10N exhibited a significantly higher impact strength of 513.39 J/m, indicating its ability to absorb more energy during impact testing. The observations of the current study are presented in the Figure 7. The impact strength observed in this study was much greater than that of the Date Palm filled polymer composite, which measured 99.45 J/m [39], and the Rice Husk-filled Polypropylene composite with added ethylene acrylic ester maleic anhydride compatibilizer, which measured 64.7 ± 2.6 J/m [40]. The impact strength is primarily controlled by factors such as toughness, and the characteristics of the interfacial zone. A decrease in impact strength was noticed with the addition of more than 10 wt% of NN particles. In this particular instance, it is plausible that the introduction of a filler at a concentration of more than 10 wt% led to a decrease in the strength of impact. Thereupon, the matrix became more rigid and prone to brittleness, diminishing the specimen's capacity to endure abrupt shocks or impacts [20]. Moreover, the inclusion of large amounts of filler particles in the composite material may result in a reduction in its ductility and flexibility. Consequently, the composite material exhibits a high level of rigidity, which may be attributed to the cross-linking properties of the polymer-filler matrix, as discussed in previous studies [41]. Making it more susceptible to brittleness and less capable of withstanding abrupt stresses or impacts.

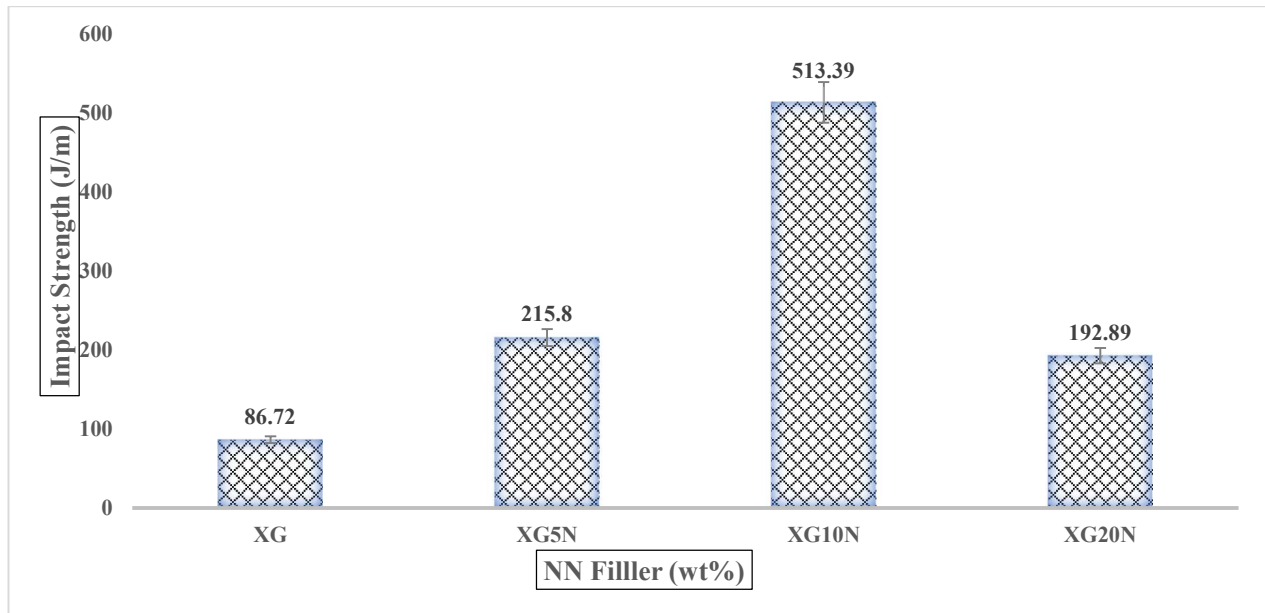


Fig.7 Impact test of the CMTCG reinforced with NN infill laminates

4.4 Dynamic Mechanical Analysis

The viscoelastic behavior, such as the storage modulus (E'), loss modulus (E''), and damping factor of the CMTCG reinforced composite with *Nelumbo nucifera* particle dispersion, the specimens underwent testing utilizing 3-point bend analyzer with a span length of 50mm. Figure 8-10 illustrates the viscoelastic comparison graphs demonstrating the effects NN fillers on the polymer composite specimens, at a frequency of 1 Hz and monitored them with respect to variations in the 3 °C/minute range from 30 to 180 degrees centigrade temperature. The storage modulus (E') and loss modulus (E'') correspond to the elastic modulus and energy dissipation during intermolecular friction of polymer chains respectively, under dynamic loading conditions. Moreover, the damping factor ($\tan \delta$) exhibits high sensitivity to structural transformations.

4.4.1 Storage Modulus (E')

In this study, the preliminary findings of all composite specimens indicate that the storage modulus (E') is greater because of the closely packed fibers, NN fillers, and epoxy, resulting in increased molecular chain immobility [14]. Figure 8 demonstrates that the storage modulus had the highest value of 6.9 GPa at room temperature for XG samples that did not have any fillers, and Figure 8 further illustrates a decline in storage modulus when NN particles are added to the epoxy composite reinforced with CMTCG fiber. This decrease in storage modulus can be attributed to the potential interference caused by the presence of NN particles, which may interrupt the orderly alignment of CMTCG fibers and epoxy, thereby compromising the elastic property of the composites and causing them to exhibit brittle behavior. As a result, the sample XG5N exhibited a reduced storage modulus of magnitude 3.95 GPa and initial viscoelastic characteristics. Composite sample XG20N illustrated a reduced storage modulus of 4.2 GPa compared to specimens XG (6.9 GPa) and XG10N (5.23 GPa) at room temperature. This may be due to a large amount of filler content, which may cause particle agglomeration, which may disrupt the uniform distribution of reinforcement throughout the matrix and weaken the incompatibility, compromising the strength. However, sample XG10N has shown better performance compared to other laminates filled with NN. Furthermore, the storage modulus of the specimens experiences a progressive decline as the temperature rises, which may be attributed to the reduction in fiber stiffness. The viscoelastic behavior of composites is seen in the glass transition zone, which occurs within a temperature range of 60 to

90 °C. During this phase, the storage modulus of all composite specimens experiences a significant decrease. The observed behavior may be ascribed to the increased molecular mobility of the epoxy chain [29, 42, 43].

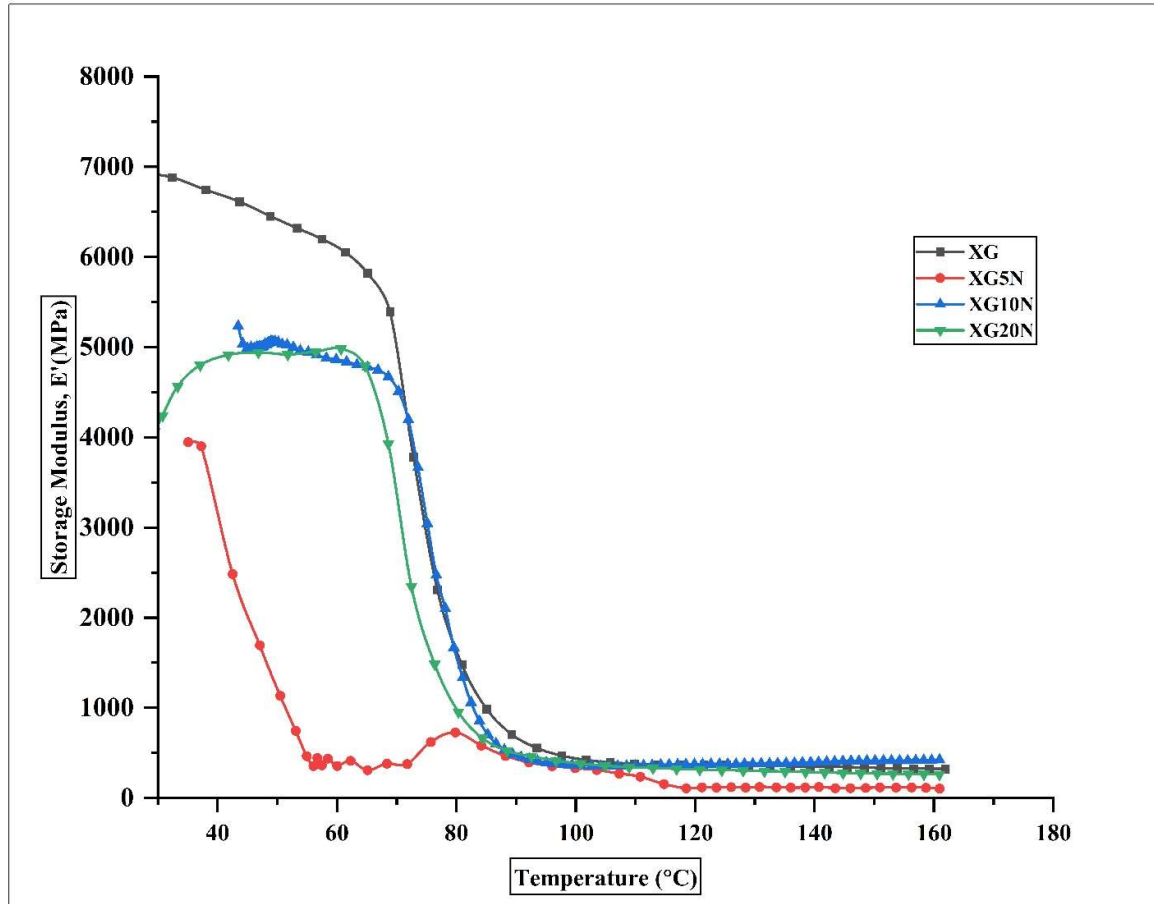


Fig.8 Storage Modulus (E') of the CMTCG reinforced with NN infill laminates

4.4.2 Loss Modulus (E'')

The loss modulus, often represented as E'' , refers to the material that undergoes irreversible energy dissipation in the form of heat under cyclic deformation and it is linked with concept of "internal friction" and exhibits sensitivity towards various molecular movements, transitions, relaxation operations, and morphology [43]. Figures 9 demonstrate the variation in loss modulus (E'') in hybrid composite specimens with respect to temperature, from room temperature to 180 °C at 1 Hz frequency. The loss modulus plots show a peak followed by a decline at elevated temperatures, indicating enhanced mobility of the polymer chains at higher temperatures. The incorporation of fibers into composites leads to an increase in the loss modulus. This can be attributed to the inherent elasticity of fibers compared to the viscoelastic behavior of polymers. As a result, the fibers exhibit enhanced energy absorption, with greater heat dissipation occurring at temperatures at the glass transition temperature (T_g) [44]. The specimen XG5N has the lowest values of loss modulus, measuring at 0.312 GPa. On the other hand, the specimen XG reveals a loss modulus of 0.834 GPa. The use of fillers in composites may result in even greater values of loss modulus when compared to pure virgin polymers [45–47]. Figure 9 provides further illustrations of the loss modulus of the XG10N sample, which was measured at 0.721 GPa (10 wt% of

NN particles), This value of loss modulus was found to be the highest among other NN incorporated samples during the current study.

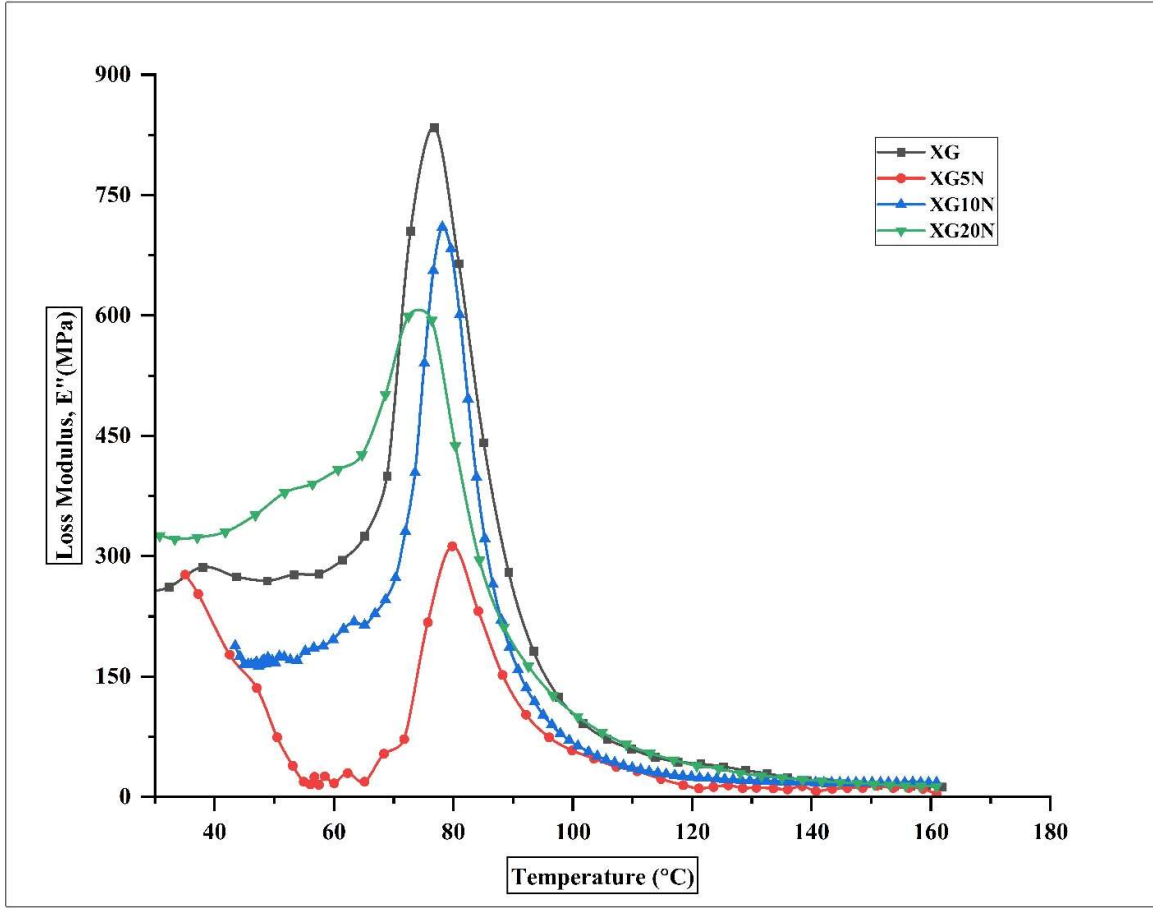


Fig.9 Loss Modulus (E'') of the CMTCG reinforced with NN infill laminates

4.4.3 Damping factor ($\tan \delta$)

The damping factor, often referred to as the loss factor ($\tan \delta$), is a dimensionless quantity that characterizes the energy dissipation capabilities of a viscoelastic material and is defined as the ratio of the loss modulus (E'') to the storage modulus (E'). Figure 10 illustrates the findings of the current study, demonstrating that hybrid composite samples exhibit the lower loss factor, this can be attributed to the presence of NN filler in the CG fiber epoxy composites, which acts as an effective barrier, thereby reducing the rate of energy loss in the epoxy molecule chain [48]. In the transitional zone characterized by elevated temperatures, the loss factor experiences a rise and attains its peak values at the glass transition temperature (T_g). Subsequently, in the rubbery region, the loss factor abruptly decreases due to the absence of any impediment to the flow. The current study demonstrates that the T_g falls within the range of 80°C to 85°C.

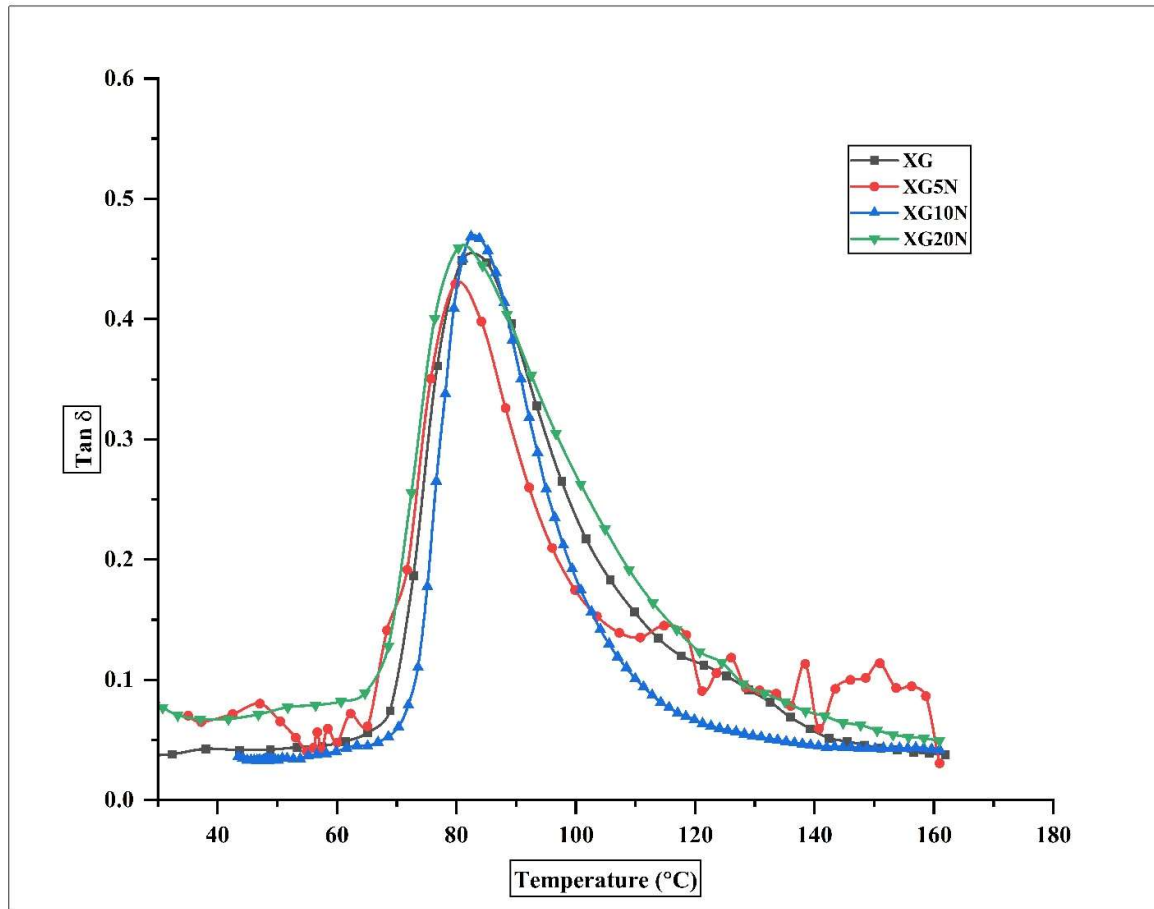


Fig.10 Damping factor (Tan δ) plot of the CMTCG reinforced with NN infill laminates

4.4.4 Cole -Cole Curve

The use of Cole-Cole plots has been employed in the assessment of the homogeneity level of composite materials. The plots shown in this study investigated the structural modifications that took place in polymeric materials after the integration of fillers. The experimental data acquired between the storage modulus (E') and the loss modulus (E'') were used to construct the Cole-Cole plot, which served as a means of assessing the dispersion of NN fillers. A homogeneous polymeric system has a well-defined semi-circle diagram spanning from storage modulus (E') to loss modulus (E''). The Cole-Cole plot was used to analyze the alterations in molecular structure integrity that arise in the polymer composite as a result of the incorporation of *Calotropis gigantea* fibers and NN fillers as reinforcements. The current investigation observed a deviation from the semi-circular curve due to the presence of CMTCG fiber and NN filler in the epoxy for all composite samples, as illustrated in Figure 11. This observation suggested that the composite samples exhibited a significant degree of homogeneity except for XG5N. This may be attributed to the uniform dispersion of the fillers inside the polymer composite. However, there is a significant degree of divergence seen in sample XG20N, indicating a tendency towards heterogeneity and different relaxation process. Moreover, the composite sample of XG5N led to a high deviation from the semicircular curve and instead displayed a linear trend, hence causing the composite to exhibit heterogeneity [28, 29].

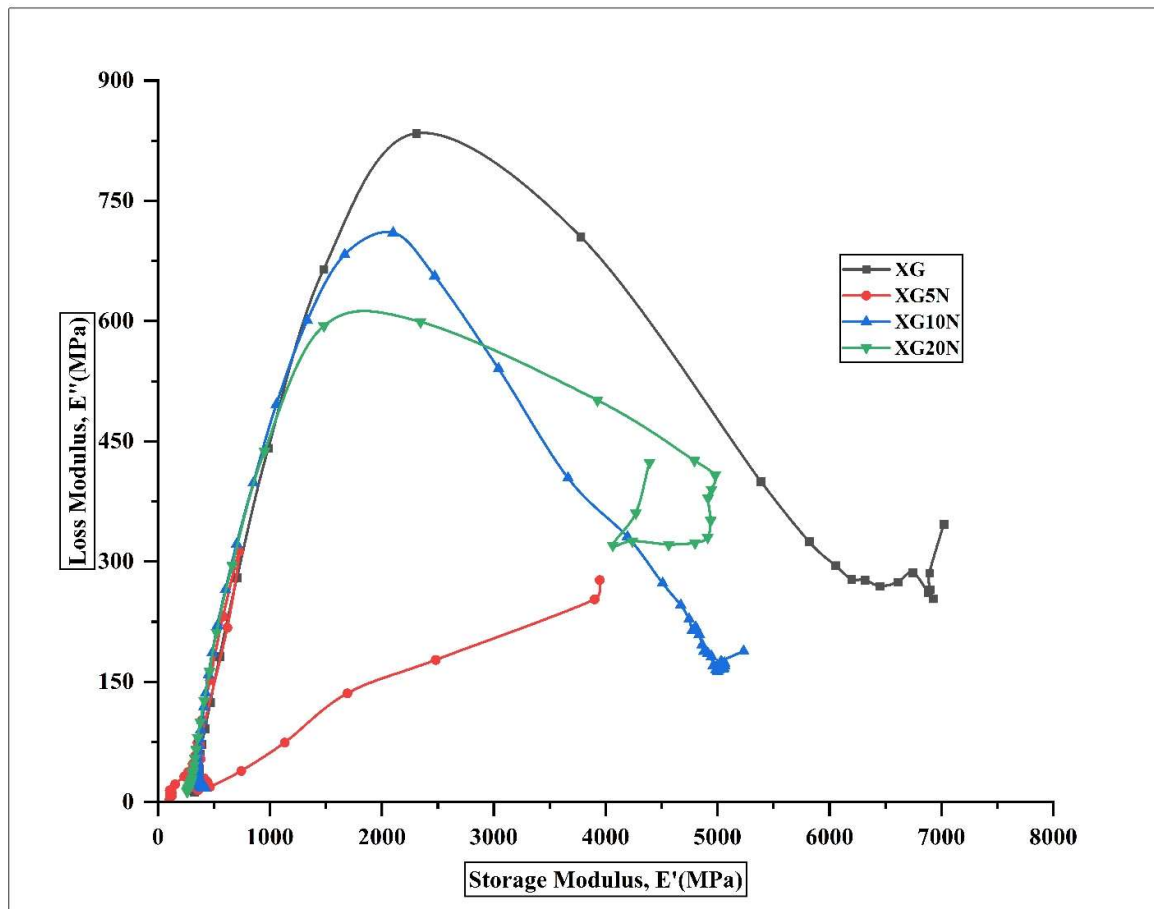


Fig.11 Cole -Cole plot of the CMTCG reinforced with NN infill laminates

5. Conclusion

The present work incorporates the CMTCG fiber and NN filler to prepare hybrid polymer natural composites. The reinforcement effectiveness of the NN filler was investigated by mechanical and DMA tests. The Experimental observations reveal that the filler impregnation weight proportion influences the composite's mechanical and viscoelastic characteristics. The mechanical performance and DMA of the composites were explored using a variety of samples with filler weight proportions ranging from 0, 5, 10, and 20 weight percentages.

Based on the analysis conducted, the following inferences have been drawn:

1. The composites' impact energy increased with the percentage of NN infill.
2. The addition of 10 wt% NN infill to composite materials results in increased impact strength of 513.39 J/m.
3. In comparison to laminate without NN filler, the tensile strength (TS) of laminate containing 10 wt% NN filler increases by 57.19% and highest tensile strength observed is 61.76MPa.
4. Due to delamination caused by insufficient interlaminar shear stress, the addition of NN particles beyond 10wt % to CMTCG fiber, laminate weakened its flexural strength.
5. DMA aided in the determination of the storage modulus values. The storage modulus of the 10 wt% of NN infills sample (XG10N) demonstrates superior performance compared with the other NN infills samples, such as XG5N and XG20N. The increase in temperature resulted in a decrease in the storage modulus (E') due to the disruption of cross-linking at higher temperatures caused by the mobility of the molecular chains[16].

6. The decrease in damping factor ($\tan \delta$) observed with filler inclusion in CG fiber-reinforced composites is caused by the limited mobility of molecular chains when filler particles are present. The glass transition temperature (T_g) of all the composites falls within the range of 60 to 90°C.
7. The relationship between storage modulus (E') and loss modulus (E'') of the NN filler incorporated CMTCG fiber reinforced composite was analyzed by Cole-Cole Plot. For the composite with 5% NN filler weight, the plot exhibited a straight-line graph, indicating the presence of heterogeneity in the composite.
8. The positive impact of incorporating the waste NN filler in CG fiber laminate has been observed (XG10N), resulting in improved performance. This enhancement is particularly beneficial in applications that involve impact and are affected by temperature.
9. The addition of 10 wt% particles exhibit optimum results in hybrid polymer composites in current study.

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Statements and Declarations

Funding

The authors declare that no funds, grants, or other financial support were received during the process of conducting the experimental study and preparation of this manuscript.

Competing Interests

The authors have a strong inclination towards analyzing the behavior of newer materials and their suitability for specific applications. Furthermore, this exploration study would pave the way for the learners to expand their research in hybrid composites.

Author Contributions

Ganesh Ram and Vinoth Kumar Kalidas - Research Gap identification, Experimentation, manuscript preparation, inferring the results.

T.V. Arjunan - Overall Supervision

Loganathan T G - Reviewing and Editing

All authors read and approved the final manuscript.

Data Availability

The raw experimental dataset generated during the current study is not publicly available due to confidentiality agreements with the participants involved. However, a summary of the findings and key results is reported in the manuscript, but it is available from the corresponding author upon reasonable request.