

Mechanical and Viscoelastic Properties of Nanoclay Filled Bamboo/Glass Fibre Reinforced Unsaturated Polyester Hybrid Composites

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The developed novel Nanoclay (NC) filled bamboo fibre (BF)/glass fibre (GF) reinforced unsaturated polyester (USP) hybrid composites were subjected to static and dynamic mechanical studies. Effect of NC on mechanical properties of the neat and NC filled BF/GF hybrid composites were tested in accordance with the ASTM standards. Dynamic mechanical analysis was performed on the composites to evaluate their viscoelastic properties like, storage modulus (E'), loss modulus (E'') and damping factor ($\tan\delta$). Outcomes of the mechanical results revealed that, NC filled BF/GF-USP hybrid composites showed superior mechanical performance over the neat hybrid composites particularly 5 wt.% NC filled BF/GF hybrid composite (HC4) outperformed other composites. From the dynamic mechanical it was found that the addition of NC filler in the hybrid composites enhanced the E' , E'' and glass transition temperature (T_g). Fractured specimens of the hybrid composites were examined using Field emission electron microscope (FESEM). Matrix fracture, micro-cracks, fibre pullouts, stretching of fibres and delamination of the fibre layers were significantly noted in the fractography analysis. Therefore, from the obtained results, NC filled BF/GF USP hybrid nanocomposites can be employed for automotive applications.

Keywords: Nanoclay, Bamboo fibre, Glass, fibre, Nanocomposites, Hybrid composites, Stacking sequence, FESEM analysis.

1. Introduction

Developments in the area of materials engineering have introduced intriguing and most eco-friendly materials, called as “natural fibre composites”. Glass fibre reinforced composites hybridizing with natural fibres can imparts significant improvement in the physico-mechanical, and thermal properties since it reflects the properties of its own elements. Owing to its unique features like, biodegradability, low weight, better resistance to wear, inexpensive, and abundance, natural fibres are employed for various applications^{1,2}. Most famously used natural fibres as reinforcement materials are, flax, jute, kenaf, sisal, bamboo, hemp, banana and etc, which has a huge resource in India¹⁻⁵. Integrating of the natural fibres with glass fibre reinforced composites can enhance the mechanical properties of the composites by which it can be related as a potential component for a specific application. In this context, present study helps to give an insight into the effect of hybridization of natural fibre and glass fibre on strength improvement for specific application.

Among the natural fibres available, bamboo fibres have unique advantages, viz., fast growth rate, cheaper, consumes low energy for processing, light weight rich

in sources, and biodegradable in nature⁶. China is known for having rich sources of bamboo as it contains one third of world's bamboo forest⁷. Since bamboo fibres are categorized as useful non-timber forest resources owing to its socio-economic benefits, deforestation of the timber-based trees could be avoided and thereby encouraging to ensure harmonious coexistence and health of ecosystems for present and future generations^{7,8}. Bamboo fibres have better mechanical strength than other cellulose fibre and because of its promising attribute, it is considered as reinforcement member for polymer composites. Though bamboo composite's specific strength is superior over the glass fibre composites, still its applications are limited due to the presence of hydroxyl group which degrades the interfacial bonding between fibre and matrix⁹. Outcome of the many research papers reveals that chemical treatment of fibre improves the interfacial bonding and there by the properties of the composites. Hybridization of glass-bamboo fibre has given the positive impact on mechanical, thermal and wear performance of polymer hybrid composites. Raghavendra Rao et al.¹⁰ revealed that flexural and compression strength of the Bamboo-glass fibre epoxy hybrid composite increase with the glass fibre addition.

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Superior properties were exhibited by alkali treated bamboo-glass epoxy composites compared to untreated counterparts. Latha et al.¹¹ reported that alternative stacking of bamboo fibre with glass fibre has enhanced tensile and flexural strength of the epoxy hybrid composites. Retnam et al.¹² found that hybrid glass-bamboo fibre with $\pm 45^\circ$ orientation performed better $0^\circ/90^\circ$ hybrid composites. They also confirmed that introduction of the bamboo fibre in the glass fibre polyester composite reduced the overall cost of the laminates without compromising their properties. Replacement of few layers of glass with natural bamboo woven fabric in glass/propylene composites is feasible for developing excellent light weight hybrid composites was reported¹³. Hybridization of glass fibre in mat form with bamboo fibre in epoxy and polyester composites has given superior strength than glass fibre reinforcement in strand form was studied¹⁴. Furthermore, physico-mechanical properties of the composites can be enhanced by the addition of inorganic nano-filler particles in the matrix, thereby it not only improves the mechanical properties but also reduces the cost¹⁵. Literatures suggested that addition on nanoclay improved the thermal resistance and mechanical properties of the composites. Muralishwara et al.¹⁶ confirmed that adding 1.5 wt.% of nanoclay in the epoxy matrix has improved tensile and flexural properties of the pineapple leaf reinforced epoxy composites. Fractography analysis clearly confirms the agglomeration of the nanoclay at 3 wt.% and causing reduction in mechanical properties of the hybrid composites. Similar observations were recorded by Mylsamy et al.¹⁷ for nanoclay filled cocinnia indica reinforced epoxy composites. Better mechanical strength was noted when the chemically modified nanoclay addition in the epoxy composites in the range of 1-3wt.%. Poor interfacial bonding owing to agglomeration on nanoclay particles at 4 wt.% decreased the mechanical properties of the composites. Bulut et al.¹⁸ reported that different weight fractions of organoclay in the epoxy has influenced specific mechanical properties of the basalt fibre reinforced epoxy composites. Alamri and Low¹⁹ in their investigation described that flexural properties of the epoxy composites were found increasing with increase in nanoclay content and moreover water resistance property of the recycled fibre paper/epoxy nanocomposite was also improved.

Composite material's performance mainly relies on the factors like, unpredicted dynamic loadings, friction between the components, type of terrain and roads, and working temperature and vibration. Often such scenarios can be experienced by the materials employed in automotive applications. Thus, it is imperative to tailor the composite materials to apt for such applications. The confirmation of the material for meeting such a requirement can fulfilled by studying their viscoelastic properties via dynamic mechanical analysis which eventually gives insight into the interfacial properties of the composite materials. Furthermore, it has become a very important technique in ascertaining the interfacial properties of composite system by subjecting to dynamic loadings with varying temperature and frequency. Chee et al.²⁰ studied the effect of hybridization of fibres and inclusion of nano-particulates on the mechanical and DMA properties of the bamboo mat/kenaf mat reinforced epoxy hybrid composites. Experimental results showed that, tensile,

flexural and impact properties of the increases with increase in the nanoclay inclusion. FESEM images of the fractured samples reveals about microcracking, delamination and higher voids due to agglomeration of the particles when the following nanoclay were added montmorillonite (MMT), and halloysite nanotube (HNT). Arulmurugan et al.²¹ found that surface interaction between nanoclay and chemically modified jute fibre had improved static and DMA properties of the jute/epoxy hybrid composite. The hardness and modulus of elasticity of the nanoclay filled polyester composites was investigated and it was found that 5 wt.% of MMT clay in the polyester matrix improved the hardness by 26.5% and modulus of elasticity by 29.52%. Viscoelastic properties of the hybrid composites synthesized by reinforcing two different fibre reinforcement and filler particulates were investigated²². It was found that filler addition imparted the positive impact on the DMA behavior and glass transition temperature. Cole-Cole plot was used to understand the heterogeneity of the composite.

Literature infers that no works have been reported till now on bamboo/glass fibre reinforced unsaturated polyester hybrid composites filled with nanoclay. Hence, present research work aims to explore the potential use of biodegradable bamboo fibre and nanoclay (Cloisite93A) for enhancing the static and dynamic mechanical properties of the USP hybrid composites. FESEM analysis on the fractured specimens are also performed.

2. Materials and Methods

2.1. Materials

Unsaturated polyester (USP) resin (1.2 g/cc) with its following catalyst and accelerator, methyl ethyl ketone peroxides (MEKP) and cobalt naphthenate, were purchased from Vasavibala Resins Private Limited, Chennai, India. Bamboo fibres and bi-directional glass fibres were used as the reinforcement members for preparing the hybrid composites and were purchased from the local vendors, Coimbatore, India. To strengthen the matrix, Nanoclay (98% of purity) fillers were introduced in the hybrid composites and was supplied by the Sigma Aldrich, Bengaluru, India. The properties of the glass fibre, bamboo fibre and NC filler are presented in the Table 1.

2.2. Fabrication of the composites

A known quantity of the USP resin was mixed with 1.5% MEKP catalyst as provided by the suppliers. To which, NC filler was mixed and thoroughly stirred by means of magnetic stirrer to ensure uniform distribution of the particles in the matrix. Further this mixture, was added with 1.5% cobalt naphthenate known as accelerator to initiate the curing process. Fibres were placed on the mould (240 mm×240mm×4mm) following a mixture of USP resin with NC filler was poured. With the help of roller and metallic plate, mixture was uniformly spread on the fibres. Stacking sequence of fibres were GF-BF-BF-GF (GF- glass fibre; BF-bamboo fibre). Once the four layers of the fibres were attained with required weight fraction of the matrix and fillers, entire system was subjected for compression mould curing under room temperature (24 hours).

Cured hybrid laminates were taken off and cut as per the ASTM standards for further testing their characteristics. Figure 1 illustrates the systematic procedure of preparing

the BF/GF USP hybrid laminates. The weight fraction of the BF/GF-USP hybrid composites and their symbol (designation) is shown in the Table 2.

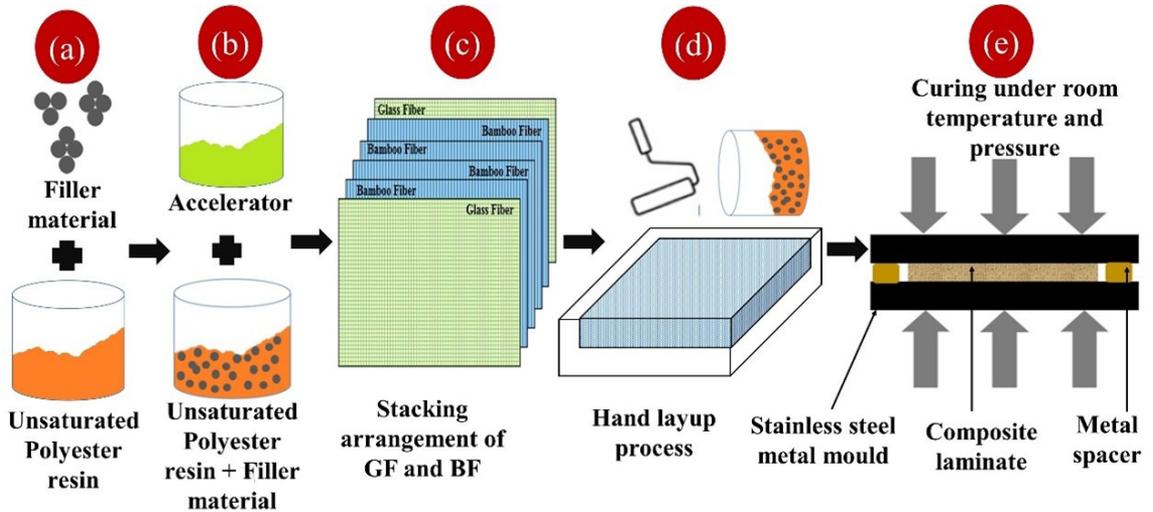


Figure 1. Stepwise procedure for fabricating the BF/GF-USP hybrid composites.

Table 1. Physical properties of the raw materials.

Material	Grade	Characteristics
Bamboo fibre (woven form)	-	Cellulose: 73%
		Hemicellulose: 12.49%
		Lignin: 10.2%
		Density: 0.91 g/cc
		Diameter of the fibre: 10-40 μ m.
Glass fibre-woven mat	E-type	Areal density: 220 g/m ²
		Density: 2.2 g/cc
		Areal density: 100 g/m ²
		Young's modulus: 75 GPa
		Elongation at break: 4.7%
Nanoclay	Cloisite93A	Bulk density-0.1659 g/cc
		Average size -1-5 μ m
		Colour-light grey and non-soluble in water
Unsaturated polyester resin	VB4503	Density: 1.2 g/cc
		Viscosity: 500-600 cps
		Gel time: 15-20 mins

Table 2. Composition of the BF/GF-USP hybrid composites.

Symbol	Stacking sequence	Weight (gm)			Thickness (mm)	
		W_f				W_m
		W_G	W_B	W_{NC}		W_m
HC1	GF+BF+BF+GF+0% NC	120	40	0	290	4
HC2	GF+BF+BF+GF+1% NC	120	40	06	290	4
HC3	GF+BF+BF+GF+3% NC	120	40	15	290	4
HC4	GF+BF+BF+GF+5% NC	120	40	25	290	4
HC5	GF+BF+BF+GF+7% NC	120	40	34	290	4

GF- Glass fibre, BF-Bamboo fibre, NC-Nanoclay

2.3. Mechanical testing

All the specimens were cut in accordance with the ASTM standards using abrasive water jet cutting machine. Tension, flexural (three-point bending), Izod impact and hardness of the specimens were tested with respect to ASTM D638, ASTM D790, ASTM D256 and ASTM D785 respectively. Tension and flexural tests were conducted in computerized universal testing machine operated at the cross-head speed of 2.5 mm/min. Izod impact test was performed to measure the maximum energy absorbed by the specimen. Brinell Hardness Number (BHN) was determined by the load and area of depth of the impression, which has been found using following relation $BHN = (2 \times \text{applied load}) / (\text{area of indentation})$. Field emission electron microscope (Make: Carl Zeiss Sigma-300, Schottky FEG) was utilized to study the surface morphology of the NC filler and mode of failure caused in tested specimens.

2.4. Dynamic mechanical analysis (DMA)

The viscoelastic properties of neat and NC filled BF/GF-USP hybrid composites were studied using a dynamic mechanical analyser (SII Nanotechnology Japan-DMS 6100). The specimens were tested under three-point bending mode for the temperature range 25 °C to 150 °C operated with a heating rate and frequency, 5 °C min⁻¹ and 10 Hz respectively. Viscoelastic properties of the materials such as storage modulus (E'), loss modulus (E''), glass transition temperature (T_g), and damping factor ($\tan\delta$) were examined.

3. Results and Discussion

3.1. Mechanical properties of the neat and NC filled BF/GF-USP hybrid composites

3.1.1. Tensile strength

Tensile strength of the unfilled and NC filled BF/GF-USP hybrid composites is shown in the Figure 2. It is evident from the Figure 2 that, incorporation of the NC filler in the BF/GF-USP hybrid composites imparted positive impact on the tensile properties. HC1 composite (unfilled) exhibited 38.12 MPa. As it is clear that tensile strength of the hybrid composite increases with increase in the addition of NC filler. When the NC filler content increases tensile strength increases, however beyond the 5 wt.%, trend declines. Of the prepared HC laminates, HC4 filled with 5 wt.% of NC filler exhibited highest tensile strength of 76.88 MPa. This may be attributed to the dispersed NC particulates in the unsaturated polyester matrix phase might have blocked the propagation and failure²³. Besides, strong interfacial region too could have caused for the improvement in the tensile property of the hybrid composites as uniformly distributed NC particulates facilitated smooth stress transfer from matrix to reinforcement member. Figure 2 clearly indicated that tensile strength of the HC2 and HC3 composites is 53.45 MPa and 61.38 MPa, respectively. For the HC5 composite containing higher weight fraction of NC filler (7 wt.%) is 64.23 MPa, nearly 16.25% lesser than HC4 composite. Though the tensile strength of HC5 is higher than HC2 and HC3, the decrement beyond 5 wt.% filler inclusion may be attributed

to the deterioration of the interfacial bonding of fibre-matrix region resulted from the agglomeration of the NC particles. Similar observations have been reported in the literature^{24,25}.

3.1.2. Flexural strength

The impact of NC particles on the flexural strength of the unfilled and NC filled BF/GF-USP hybrid composites is illustrated in the Figure 3. Flexural strength of the BF/GF-USP hybrid composites increases with increase in the addition of NC particles. The trend noted for the flexural strength of the NC filled BF/GF-USP hybrid composites is unlike its tensile property. It is evident from the Figure 3 that, flexural strength of the NC filled hybrid composite outperformed neat hybrid composites. Superior bending strength of 92.86 MPa is exhibited by HC5 composites followed by HC4, HC3, and HC2 composites. However, no significant difference is observed between HC2 and HC3 hybrid composites, as their strength is 66.21 MPa and 68.75 MPa, respectively. Bending strength of the hybrid composites relies on its tougher reinforcement member used. In this study, while stacking reinforcement members, glass fibres are placed in the skin layer of the composites which is far tougher than bamboo fibres. Thus, good stiffness material as like glass fibre result in good flexural strength^{23,26}. From the figure it can be noted that, flexural strength of the HC5 composites showed 66.66% and 22.38% improvement over HC1 and HC4 composites, respectively, which shows that NC filler has significant impact on the bending property of the BF/GF-USP hybrid composites.

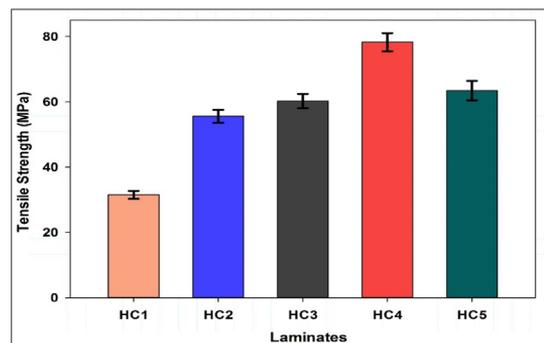


Figure 2. Tensile strength of the various BF/GF-USP hybrid composites.

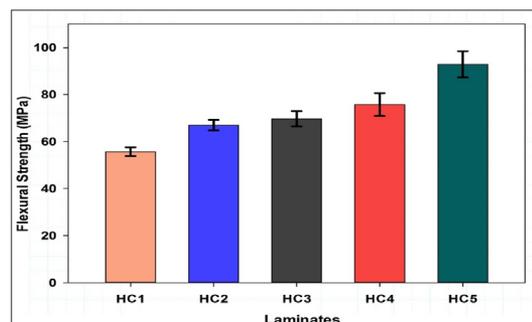


Figure 3. Flexural strength of the various BF/GF-USP hybrid composites.

The NC filler has potential to enhance the fracture resistance. In the FRP composites, polymer matrix and fillers are accountable for transferring load to rigid fibres through shear forces at the junction and thus it demands a strong relation between matrices, fillers, and fibres. Improvement in the flexural strength of the NC filled BF/GF-USP hybrid composites attributed to the strong adherences of fibre, filler and matrices. Inclusion of NC filler in BF/GF-USP hybrid composites has improved toughness owing to interfacial interaction between NC filler –fibres and epoxy matrix is stronger²⁷.

3.1.3. Impact strength

Figure 4 depicts the impact energy of the unfilled and NC filled BF/GF-USP hybrid composites. It is clear from the plot that impact energy of the hybrid composite improved with NC filler addition and linearly increases with weight fraction up to 5 wt.% and beyond declines. HC4 composite recorded the highest impact energy 1.691 J, followed by HC5, HC3 and HC2 composites. When compared with the neat hybrid composites, 60.89% higher impact energy is displayed by the HC5 composites which clearly shows the significant effect of the NC filler in the hybrid composites. Enhancement of the impact energy in the composites might have caused due to the strong interfacial adhesion of fibre-filler-matrix which assisted smooth distribution of load from matrix to fibre member. Similar behaviour is recorded by the other researchers^{2,6,27}.

3.1.4. Surface hardness property

Figure 5 depicts the BHN surface hardness value of the neat and NC filled BF/GF-USP hybrid composites. BHN increasing with increase of wt.% of NC particulates and highest value is recorded for the HC4 composites (28 BHN), containing 5 wt.% of NC filler as a secondary reinforcement member in the composites. Basically, hardness property is related with the plastic deformation, inclusion of the NC particulates in the hybrid composites could have enhanced the resistance to the plastic deformation and hence hardness is improved with the increase in the wt.% of NC filler. Moreover, uniform distribution of the NC filler reduces voids in the composites, which might have also been aided in increasing the hardness of the composites. These BF/GF-USP hybrid composites were prepared keeping glass fibre as a skin layer; whereas bamboo fibres in the core. This might have also resulted in the better hardness value of the composites. Similar observation was reported by the Dhanunjayarao et al.²³, where E-glass fibre used as a skin layer in the glass/jute hybrid composites due to which hardness of the composite has improved.

3.2. Viscoelastic properties of the neat and NC filled BF/GF-USP hybrid composites

3.2.1. Storage modulus (E') property

Storage modulus (E') plays the vital role in understanding the load bearing capacity of the composite materials as it reflects the elastic component of the viscous elastic material. Apart from conveying material toughness and grade of crosslinking, it also gives insight into the fibre-matrix

interfacial attachment. Solid region of the E' curve reveals the bonding capacity of the composite material during the increase of temperature. Increasing the ratio of crystalline to amorphous region conveys improvement in the stiffness of the fibre and declining flexibility. Higher crystalline composite material provides more strength but less elongation. However, hardness and density of the material increases with increase in degree of crystalline. The developed BF/GF USP hybrid laminates are amorphous material and their elastic region was identified using the E' curve as shown in the Figure 6. From the curves it is evident that HC4 composite found to be more stiffness and has high elastic attributes compared with other neat and NC filled BF/GF-USP hybrid composites.

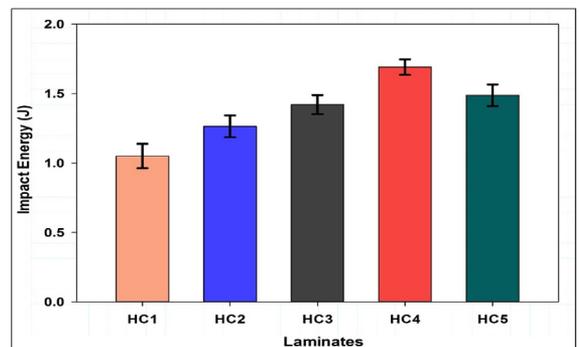


Figure 4. Impact energy of the various BF/GF-USP hybrid composites.

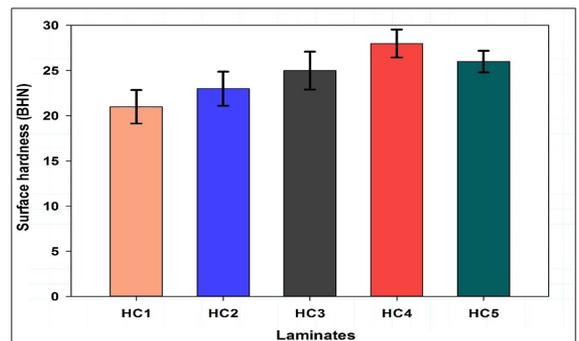


Figure 5. Hardness of the various BF/GF-USP hybrid composites.

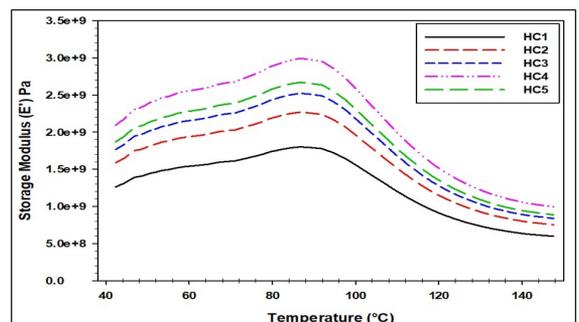


Figure 6. Storage modulus (E') of the various BF/GF-USP hybrid composites.

The maximum storage modulus of 2990 MPa was noted at the temperature 86.64°C and minimum 2011 MPa at 109.67°C. While initiating the experiment at 41.19°C, storage modulus of the HC4 composite was 2133 MPa. When compared with neat BF/GF USP hybrid composites, HC4 has 60.23% higher stiffness which clearly indicates the positive impact of the NC filler in the hybrid composite. However, HC5 composite which has 7 wt.% of NC filler showed lower storage modulus property than HC4, but higher than rest of their counterparts. This may be owing to the agglomeration of the particles at higher weight fraction restricts the stress transmission from matrix to fibres resulting from poor interfacial bonding of matrix-filler-fibre.

Storage modulus curve reveals the USP hybrid composite's phase transition zones namely solid-state region, glass transition region and rubbery region. Solid state phase has occurred from the starting temperature 40°C to 80°C; following glass transition between 80°C to 110°C and after this, rubbery region. From the Figure 6 it is clearly notes that, E' values of the hybrid composites drop as it passes through glass transition region. In the rubbery region, HC4 composites outperformed other hybrid composites by exhibiting higher E' (2011 MPa). This may be attributed to the strong interfacial bonding of matrix and filler allowing stress transmission from the matrix phase to fibres. All the USP hybrid composite experiences lower E' with increasing temperature owing to the loss of stiffness of the fibre at higher temperature. As soon as the glass transition temperature region approaches, free volume of the composite grows and mobility of the polymer chain intensifies. Modulus of the USP hybrid composite decreases dramatically after glass transition region and following no apparent changes in E' is noticed as material has entered rubbery region. Similar observations were reported by the other researchers.

3.2.2. Loss modulus (E'') property

Composite material's viscous behavior and heat dissipation during the increasing of temperature is examined using loss modulus. The loss modulus (E'') curve of the neat and NC filler BF/GF USP hybrid composite is shown in the Figure 7. It is important to note that, all the specimens have shown the similar trend as indicated in the graph, from starting increasing to until reaches maximum and before settling down to lower E'' value in rubbery region. This clearly indicates the polymer chain in the BF/GF hybrid composite gets intensified during exposure to thermal energy and thereby temperature acts as a catalyst for the free mobility of the polymer chain. Of the prepared hybrid composites, HC4 exhibited high peak loss modulus than other composites which shows the effect of the NC filler presence. BF/GF USP hybrid composite with NC filler exhibited higher E'' than neat one this may because NC filler might have assisted in enhancing the internal friction that endorses energy dissipation in the hybrid laminates. Hybridization of the glass fibre and bamboo fibre also affects the loss modulus property of the composites. Portella et al.²⁸ reported that E'' value of the cotton polyester composites found to be superior when it was reinforced with glass fibre than unhybridized composite. Because in hybrid composite internal friction is quite higher as a result of hybridization

of the different fibres which eventually promotes the higher energy dissipation in the composite system

3.2.3. Damping factor ($Tan\delta$)

Damping factor denoted as $Tan\delta$ and is the ratio of storage modulus to the loss modulus which conveys the damping capacity of the composite material. $Tan\delta$ reveals the material behavior during rubbery region. High value of damping ratio imply that the material is non-elastic as it has dissipated lot of energy and on other hand, material is more elastic if the value is lower. In the Figure 8, the value of damping factors is the glass transition temperature (T_g) of the composite which represent the interphase bonding of the composites. The value of the damping factor and its corresponding T_g values are shown in the Table 3. The $Tan\delta$ peak values found lower for HC4 and HC5 composites which indicates the composites have strong interface adhesion than rest of the composites. This might have occurred by the restriction of the polymer chain owing to the rigidity of the hybrid composites and thereby causing lesser to dissipate.

3.3. FESEM analysis

For the tensile fractured specimens, stretching and breaking of GF and BF are noted with matrix damages as shown in the Figure 9. BF pull outs are predominantly observed as these fibres are used as core reinforcement member, however skin layer used GF are found strongly adhering with matrix phase than BF, may be due to its minimum areal density.

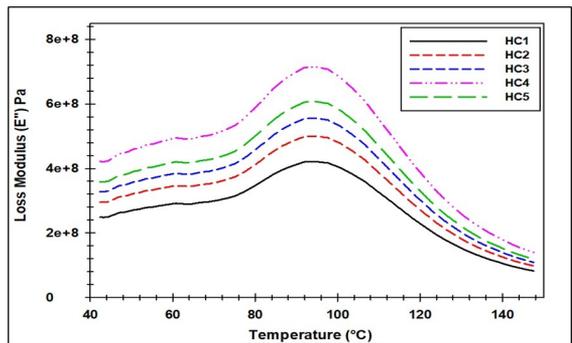


Figure 7. Loss modulus (E'') of the various BF/GF-USP hybrid composites.

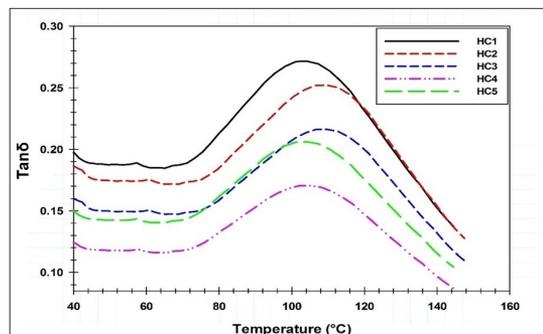
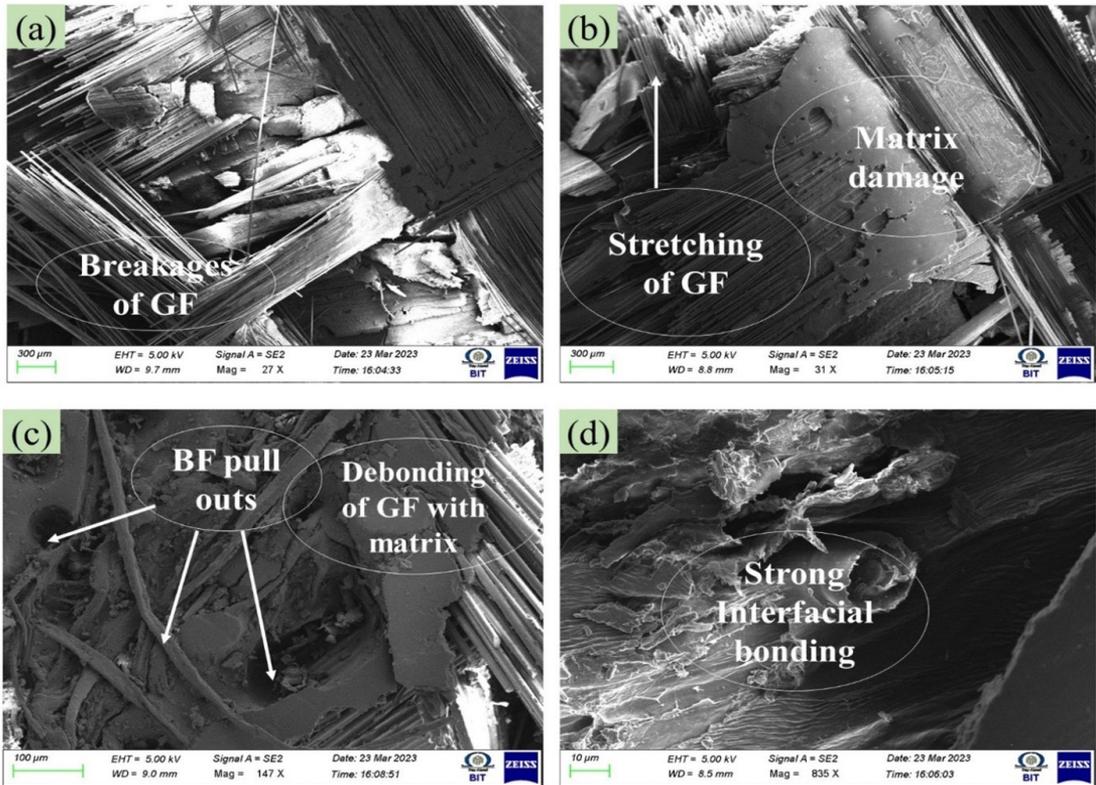


Figure 8. Damping factor of the various BF/GF-USP hybrid composites.

Table 3. Viscoelastic properties and glass transition temperature (T_g) of the composites.

Viscoelastic properties	Neat and NC filler BF/GF USP hybrid composites				
	HC1	HC2	HC3	HC4	HC5
E' (MPa)	1803	2269	2523	2992	2671
E'' (MPa)	421	500	555	607	714
Damping factor ($\tan\delta$)	0.252	0.249	0.223	0.157	0.181
T_g ($^{\circ}\text{C}$)	103.5	109.32	110.11	104.5	101.11

**Figure 9.** FESEM images of the tensile fractured specimens of (a, c) HC1 and (b, d) HC4 composites.

Despite, debonding of matrix-GF and severe matrix fracture before the failure is noted in the Figure 9c for HC1 composite due to the phase change transformation in the matrix as a result of hybridisation of fibre. In the HC5 composite at 7 wt. % of NC filler addition, stretching of GF and fracture, pull outs of BF are not significant as observed in the case of HC1 composites, this may be due to the strong interfacial bonding of matrix-fibre which have restricted the stretching of skin layer GF, and avoided it breakage, thus increasing in tensile strength of the NC filled hybrid composite is found.

FESEM images of the hybrid composites were captured to analysis the interfacial properties, internal cracks and internal morphology of the fractured surface. Bending of the glass fibres and bending with fracture in bamboo fibres were noteworthy to observe due to the flexural load is shown in the Figure 10a, which clearly indicates that HC1 composites possess brittleness property. During the flexural testing, one surface of the composite specimen experiences strong tensile

force and another surface by compression force. Because of such mechanism, major failure of the composite embarks with the matrix cracking following intense fibre fracture and pull outs. As the HC1 composites are brittle in nature, it endures more matrix fracture and fibre pull outs than NC filled composite as indicated in the Figure 10c. Addition of the NC filler in the hybrid BF/GF hybrid composite reduced the stretching of glass fibres and cracks in the bamboo fibres and moreover, matrix cracking is merely seen in the NC filled composite even at higher magnification as indicated in the Figure 10b, which clearly implies improvement in the flexural strength of the composites due to the incorporation of NC filler.

Impact strength of the unfilled and NC filled BF/GF-USP hybrid composite reflects the toughness of the material which depends on the constituents, fibre-matrix interface, and geometry of the testing. It is clearly indicated from the study that, impact strength of the HC4 composite exhibited higher

impact strength than other composites. Though difference among the NC filled composites is very marginal, it clearly indicates the improvement in the toughness of the material after the addition of NC filler, which may be attributed to the strong absorption of impact energy by the NC filler and transfers the same by hindering the crack propagation as indicated in the Figure 11d, where no cracks were found.

Moreover, HC5 composites have suffered less fibre pull-outs, fibre breakages, and matrix damages than other composites clearly convey the strong bonding between fibre and NC filler which is evident from the Figure 11c, d. Delamination of GF with matrix phase found higher than BF owing to poor adhesion is observed from the Figure 11a, which is later reduced after the inclusion of NC filler.

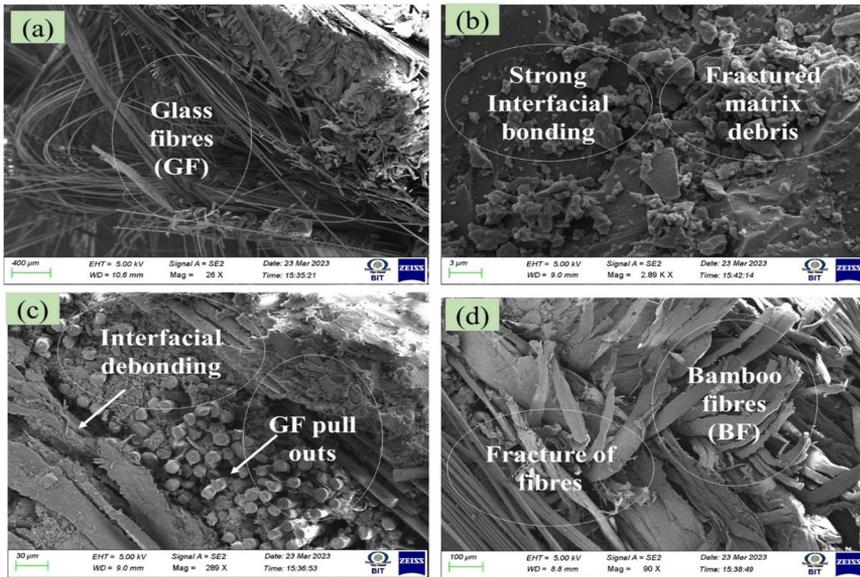


Figure 10. FESEM images of the flexural fractured specimens of (a, c) HC1 and (b, d) HC4 composites.

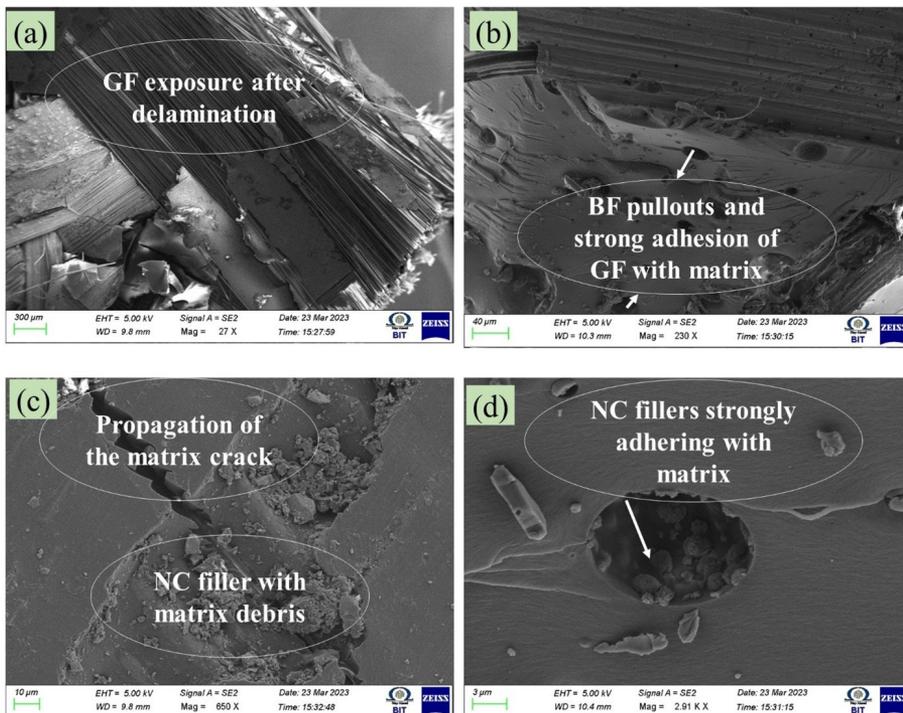


Figure 11. FESEM images of the impact fractured specimens of (a, b) HC1 and (c, d) HC4 composites.

4. Conclusions

BF/GF USP hybrid composites filled with different weight fraction of NC filler were successfully fabricated. Impact of NC filler on mechanical and dynamic mechanical properties of the hybrid composites were studied and from which following inferences were drawn:

1. Impregnation of the NC filler in the BF/GF-USP hybrid composites enhanced the mechanical performance of the composites. Of the prepared hybrid-nanocomposites, HC4 (5 wt.% NC filler) exhibited superior tensile (76.88 MPa), impact (1.69 J) and hardness (28 BHN) properties than other composites. Whereas in case of flexural property, HC5 composite containing (7 wt.% NC filler) dominated other laminates. However, between HC4 and HC5 composites, only 22.38% improvement in flexural strength was recorded.
2. Dynamic mechanical analysis results showed that, E' , E'' , and damping factor of the NC filled BF/GF-USP hybrid composites showed better performance than the neat composites. This may be because of the hybridization of the fibres with an inorganic filler promoted for the highest dissipation of energy in the composites resulting from the intense internal friction by the hybridization effect.
3. Highest storage modulus (E'), and loss modulus (E'') was exhibited by the HC4 composites. Damping factor ($\tan\delta$) found decreased with increase in the NC filler content and better damping property (0.157) was displayed by the HC4 composite followed by HC5, HC3, HC2, and HC1 composites. However, glass transition temperature (T_g) of the neat hybrid BF/GF-USP composite was found to be 103.5°C and increased to 110.11°C due to the addition of the (3 wt.% NC filler) as exhibited by the HC3 composites. This may be attributed to the requirement of high thermal energy for inducing uniform motion of molecules.
4. Examination of the mechanically fractured specimens clearly revealed their mode of failure endured during the testings. Common observations like, microcracking, matrix fracture, fibres pullouts, fibre stretching especially skin layer glass fibre, and poor interfacial adhesion were significantly identified. However, in hybrid nanocomposites filled with 5 wt.% NC filler (HC4 composite) showed strong interfacial adhesion especially between NC filler and USP matrix. This may be the main reason for the HC4 composite to outperform other laminates in static and dynamic mechanical tests.

5. References

1. Ibrahim NI, Shahar FS, Sultan MT, Shah AU, Safri SN, Mat Yazik MH. Overview of bioplastic introduction and its applications in product packaging. *Coatings*. 2021;11(11):1423. <http://dx.doi.org/10.3390/coatings11111423>.
2. Kumar SV, Singh H. Wear, morphological and thermal behavior of flax fibre reinforced epoxy composites. *Indian J Fibre Text Res*. 2023;48(3):326-35. <http://dx.doi.org/10.56042/ijftr.v48i3.6057>.
3. Mahmud S, Hasan KF, Jahid MA, Mohiuddin K, Zhang R, Zhu J. Comprehensive review on plant fiber-reinforced polymeric biocomposites. *J Mater Sci*. 2021;56(12):7231-64. <http://dx.doi.org/10.1007/s10853-021-05774-9>.
4. Hasan KF, Horváth PG, Alpár T. Potential natural fiber polymeric nanobiocomposites: a review. *Polymers*. 2020;12(5):1072. <http://dx.doi.org/10.3390/polym12051072>.
5. Joseph B, Sagarika VK, Sabu C, Kalarikkal N, Thomas S. Cellulose nanocomposites: fabrication and biomedical applications. *J Bioresour Bioprod*. 2020;5(4):223-37. <http://dx.doi.org/10.1016/j.jobab.2020.10.001>.
6. Jena H, Pandit MK, Pradhan AK. Effect of cenosphere on mechanical properties of bamboo-epoxy composites. *J Reinf Plast Compos*. 2013;32(11):794-801. <http://dx.doi.org/10.1177/0731684413476925>.
7. Natrayan L, Kumar P, Baskara Sethupathy S, Sekar S, Patil PP, Velmurugan G, et al. Effect of nano TiO₂ filler addition on mechanical properties of bamboo/polyester hybrid composites and parameters optimized using grey Taguchi method. *Adsorpt Sci Technol*. 2022;2022:6768900. <http://dx.doi.org/10.1155/2022/6768900>.
8. Wang G, Chen F. Development of bamboo fiber-based composites. In: Fan M, Fu F, editors. *Advanced high strength natural fibre composites in construction*. London: Woodhead Publishing; 2017. p. 235-55. <http://dx.doi.org/10.1016/B978-0-08-100411-1.00010-8>.
9. Chen F, Jiang Z, Wang G, Shi SQ, Liu X. Bamboo bundle corrugated laminated composites (BCLC). Part I. Three-dimensional stability in response to corrugating effect. *J Adhes*. 2013;89(3):225-38. <http://dx.doi.org/10.1080/00218464.2013.739044>.
10. Raghavendra Rao H, Varada Rajulu A, Ramachandra Reddy G, Hemachandra Reddy K. Flexural and compressive properties of bamboo and glass fiber-reinforced epoxy hybrid composites. *J Reinf Plast Compos*. 2010;29(10):1446-50. <http://dx.doi.org/10.1177/0731684409105077>.
11. Latha PS, Rao MV, Kumar VK, Raghavendra G, Ojha S, Inala R. Evaluation of mechanical and tribological properties of bamboo-glass hybrid fiber reinforced polymer composite. *J Ind Text*. 2016;46(1):3-18. <http://dx.doi.org/10.1177/1528083715569376>.
12. Retnam BS, Sivapragash M, Pradeep P. Effects of fibre orientation on mechanical properties of hybrid bamboo/glass fibre polymer composites. *Bull Mater Sci*. 2014;37(5):1059-64. <http://dx.doi.org/10.1007/s12034-014-0045-y>.
13. Zuhudi NZ, Lin RJ, Jayaraman K. Flammability, thermal and dynamic mechanical properties of bamboo-glass hybrid composites. *J Thermoplast Compos Mater*. 2016;29(9):1210-28. <http://dx.doi.org/10.1177/0892705714563118>.
14. Kushwaha PK, Kumar R. The studies on performance of epoxy and polyester-based composites reinforced with bamboo and glass fibers. *J Reinf Plast Compos*. 2010;29(13):1952-62. <http://dx.doi.org/10.1177/0731684409342006>.
15. Moustafa NM, Mohammed KA, Al-Ameen ES, Ogaili AAF, Al-Sabbagh MNM. Mechanical and tribological properties of walnut/polypropylene natural composites. *J Mech Eng Dev Res*. 2020;43(4):372-80.
16. Muralishwara K, Fernandes L, Kalkura R, Bangera S. Effect of surface modified montmorillonite nanoclay on tensile and flexural properties of pineapple leaf fiber reinforced epoxy composite. *Mater Res*. 2023;26:e20220437. <http://dx.doi.org/10.1590/1980-5373-mr-2022-0437>.
17. Mylsamy B, Palaniappan SK, Subramani SP, Pal SK, Aruchamy K. Impact of nanoclay on mechanical and structural properties of treated *Coccinia indica* fibre reinforced epoxy composites. *J Mater Res Technol*. 2019;8(6):6021-8. <http://dx.doi.org/10.1016/j.jmrt.2019.09.076>.
18. Bulut M, Bozkurt ÖY, Erklığ A, Yaykaşlı H, Özbek Ö. Mechanical and dynamic properties of basalt fiber-reinforced composites with nanoclay particles. *Arab J Sci Eng*. 2020;45(2):1017-33. <http://dx.doi.org/10.1007/s13369-019-04226-6>.

19. Alamri H, Low IM. Effect of water absorption on the mechanical properties of nanoclay filled recycled cellulose fibre reinforced epoxy hybrid nanocomposites. *Compos, Part A Appl Sci Manuf.* 2013;44:23-31. <http://dx.doi.org/10.1016/j.compositesa.2012.08.026>.
20. Chee SS, Jawaid M, Sultan M, Alothman OY, Abdullah LC. Effects of nanoclay on physical and dimensional stability of Bamboo/Kenaf/nanoclay reinforced epoxy hybrid nanocomposites. *J Mater Res Technol.* 2020;9(3):5871-80. <http://dx.doi.org/10.1016/j.jmrt.2020.03.114>.
21. Arulmurugan S, Venkateshwaran N. The effect of fiber reinforcement on fracture toughness assessment of nanoclay filled polymer composites. *Surf Rev Lett.* 2019;26(9):1950050. <http://dx.doi.org/10.1142/S0218625X19500501>.
22. Arulmurugan S, Venkateshwaran N. Effect of nanoclay addition and chemical treatment on static and dynamic mechanical analysis of jute fibre composites. *Polimeros.* 2019;29(4):e2019054. <http://dx.doi.org/10.1590/0104-1428.08619>.
23. Dhanunjayarao B, Sanivada UK, Swamy Naidu N, Fanguero R. Effect of graphite particulate on mechanical characterization of hybrid polymer composites. *J Ind Text.* 2022;51(2, Suppl):2594S-615S. <http://dx.doi.org/10.1177/15280837211010670>.
24. Dalbehera S, Acharya S. Effect of cenosphere addition on the mechanical properties of jute-glass fiber hybrid epoxy composites. *J Ind Text.* 2016;46(1):177-88. <http://dx.doi.org/10.1177/1528083715577936>.
25. Ramakrishnan S, Krishnamurthy K, Rajasekar R, Rajeshkumar G. An experimental study on the effect of nano-clay addition on mechanical and water absorption behaviour of jute fibre reinforced epoxy composites. *J Ind Text.* 2019;49(5):597-620. <http://dx.doi.org/10.1177/1528083718792915>.
26. Oladele I, Oladejo M, Adediran A, Makinde-Isola B, Owa A, Akinlabi E. Influence of designated properties on the characteristics of dombeya buettneri fiber/graphite hybrid reinforced polypropylene composites. *Sci Rep.* 2020;10(1):11105. <http://dx.doi.org/10.1038/s41598-020-68033-y>.
27. Karthick L, Rathinam R, Kalam SA, Loganathan GB, Sabeenian R, Joshi S, et al. Influence of nano-/microfiller addition on mechanical and morphological performance of kenaf/glass fibre-reinforced hybrid composites. *J Nanomater.* 2022;2022:1-10. <http://dx.doi.org/10.1155/2022/9778224>.
28. Portella EH, Romanzini D, Angrizani CC, Amico SC, Zattera AJ. Influence of stacking sequence on the mechanical and dynamic mechanical properties of cotton/glass fiber reinforced polyester composites. *Mater Res.* 2016;19(3):542-7. <http://dx.doi.org/10.1590/1980-5373-MR-2016-0058>.