STM JOURNALS

Journal of
Polymer & Composites

https://journals.stmjournals.com/jopc

ISSN: 2321-2810 (Online)
ISSN: 2321-8525 (Print)
Volume 14, Issue 1, 2026
DOI (Journal): 10.37591/JoPC

Research JoPC

Tailoring Toughness of Pineapple Leaf Fiber-Reinforced Epoxy Composites Using Halloysite Nanotubes

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Abstract

This comprehensive study investigates the effects of halloysite nanotubes (HNTs) on the impact strength and fracture toughness of pineapple leaf fabric-reinforced epoxy (PALF/Ep) composites with an emphasis on sustainable material development. ASTM D256 and ASTM D5045 standards were followed in the fabrication of the composites. HNT weight percentages were systematically changed from 0 to 3wt% through using solution mixing and manual lay-up techniques, and vacuum bagging was used to obtain the best fiber-matrix impregnation. Results from these experimental studies demonstrate significant improvements in mechanical performance. According to single-edge notched bend tests, fracture toughness increased from 2.91 ± 0.3 MPa·m1/2 in the unfilled PALF/Ep to a maximum of 4.92 ± 0.2 MPa·m1/2 at 2wt% HNT loading. Izod impact testing revealed that impact strength reached a maximum of 433.9 J/m at this loading, an impressive 95.2% improvement above the unfilled PALF/Ep of 222.3 J/m. Due to nanotube agglomeration, which promoted stress concentrations and poor dispersion, performance somewhat decreased at 3wt% HNT loading. However, HNTs work well as nanofillers, increasing the PALF/Ep composite's impact and fracture toughness. These results advance bio-based engineering solutions by supporting HNT-modified PALF/Ep for high-performance green composites in structural areas including wind turbine blades and automotive panels.

Keywords: Fracture toughness, halloysite nanotubes, impact strength, nanocomposite toughening mechanisms, PALF/Ep composites

INTRODUCTION

Natural-fiber-reinforced polymer-matrix composites (NF-RPMCs) truly offer lightweight, cost-effective, and highly sustainable alternatives to various synthetic fibers. In contrast to synthetic fiber composites, NF-RPMCs have several well-known disadvantages, such as variable fiber quality, high moisture absorption, and most significantly for structural applications relatively weak fiber-matrix interfacial bonding, which frequently results in brittle matrix-dominated failure, limited impact resistance, and modest fracture toughness [1, 2].

Pineapple leaf fiber (PALF), with its high strength and biodegradability, is a promising reinforcement, yet its composites often suffer from limited toughness [3]. In the study by Roy [4], hybrid epoxy (Ep) composites reinforced with carbon fiber and pineapple leaf fiber (PALF) were examined with and without nano TiO_2 fillers. Superior performance was demonstrated by nano-reinforced composites, according to mechanical and thermal characterizations. Increased interfacial bonding led to a significant improvement in tensile, flexural, and impact strengths. The strength score of the hybrid composite filled with nano TiO_2 was 23.4, while that of neat Ep was 4.5.

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Received Date: October 25, 2025
Accepted Date: October 29, 2025
Published Date: February 14, 2026

Citation: Krishna, Hema H, Amoghavarsha, Bharath P B, Suresha B. Tailoring Toughness of Pineapple Leaf Fiber-Reinforced Epoxy Composites Using Halloysite Nanotubes. Journal of Polymer & Composites. 2026; 14 (1): 261–272p.

Numerous studies have investigated using nanofillers, such as carbon nanotubes (CNTs) [5], graphene nanoplatelets (GNPs) [6], nanocellulose [7], halloysite nanotubes (HNTs) [8], nanoclay (oMMT) [9], and nanosilica (nS) [10], to overcome these mechanical shortcomings. The use of CNTs as nanofillers in polymer composites was reviewed by Nurazzi et al[5]. because of their remarkable electrical, thermal, and strength characteristics. The role of CNTs in improving mechanical performance for a variety of applications was highlighted in the study. The potential of CNTs hybridized composites reinforced with natural fibers, such as kenaf, bamboo, and oil palm fibers, was also highlighted. The first thorough analysis of GNPs-incorporated natural lignocellulosic fiber polymer composites and their expanding technological significance was provided by Da Luz et al[6].. They claimed that improved interfacial adhesion brought about by GNPs amphiphilic nature resulted in improved mechanical, thermal, electrical, and ballistic properties. Hybrid filler addition and graphene oxide/reduced graphene oxide surface functionalization are two key integration strategies that make advanced products for defense, biomedical, and energy applications possible. To improve mechanical, thermal, and barrier properties, Thomas et al.'s study [7] developed a polyvinyl alcohol (PVA)-polyethylene oxide (PEO) (PVA-PEO) nanocomposite reinforced with hybrid fillers of HNTs and nanocellulose. The optimized composition doubled the Young's modulus, increased thermal stability, and increased tensile strength by 20%. Additionally, its oxygen barrier performance improved by 67%, which qualified it for use in advanced packaging applications. Alkali-treated PALF reinforced Ep composites with different amounts of HNTs were studied by Suresha and Hemanth [8]. The tensile strength increased by 16%, the Young's modulus increased by 22%, and the flexural properties improved by roughly 23–30% with the addition of 3wt% HNTs. For advanced engineering applications, the study found that this composition greatly improved mechanical performance and morphology. The effects of oMMT on Ep hybrid composites reinforced with bamboo and kenaf were examined by Chee et al [9]. In comparison to other composites, the study showed that those containing 1wt% oMMT exhibited superior tensile, flexural, and impact strengths. Because of their improved viscoelastic qualities and increased fiber-matrix adhesion, oMMT-filled composites are perfect for load-bearing, lightweight structural applications. Composites of PALF and sisal fiber reinforced with nS exhibit the highest mechanical strength and the least number of wear at 4wt% nS. For applications in construction, packaging, and automobiles, the optimized C-type hybrid showed exceptional durability and environmentally beneficial potential [10].

The appropriate dispersion of nanofillers can improve fracture toughness and impact energy absorption by (a) increasing matrix stiffness and toughness, (b) altering the fiber–matrix interphase to promote stress transfer, and (c) blunting, deflecting, or bridging propagating fractures. Mousavi et al[11]. examined developments in employing GNPs, CNTs, and their hybrids to increase the toughness of Ep resins. Chemical functionalization enhances nanofiller dispersion and fracture resistance, according to the study. Insights into toughening processes that toughen Ep composites without sacrificing their desired characteristics were revealed. Zeinedini [12] established a model to examine the impact of GNPs agglomeration on nanocomposites' fracture toughness. They included variables for filler alignment, random orientation, and crack pinning mechanisms using the J-integral approach. The fracture toughness variations caused by filler dispersion were accurately predicted by the model, which demonstrated strong agreement with experimental data. The effect of CNTs and GNPs on the fracture toughness of Ep composites was investigated by Shirodkar et al[13].. The toughness and fracture energy of CNT-reinforced Ep with 0.5% filler increased the most, whereas GNP and hybrid CNT/GNP systems also improved properties, albeit less significantly. The observed improvements in toughness were explained by mechanisms, such as crack bridging and deflection that were identified through microscopy.

Significant increases in impact tolerance and interlaminar/Mode-I fracture toughness of NF-RPMCs following small additions of nanofiller have been reported in reviews and experimental studies conducted over the past five years [14–18]. Natural fiber reinforced polymer composites have become a promising material because of their favorable mechanical performance, low weight, and sustainability. In this work, the fracture toughness and surface morphology properties of epoxy composites reinforced with alkali-treated banana fibers and nano-MgO fillers are examined [14]. Epoxy nanocomposites reinforced with carbon black nanofillers made from biomass sources were studied by Dungani et al[15]..

While excessive 10% loading decreased toughness, optimal filler loading between 1% and 5% improved mechanical and physical properties. Microscopic examination showed that mechanisms that improve toughness include crack bridging, void formation, and crack deflection. To balance toughness, strength, and stiffness, Sun et al[16]. developed polyethylene terephthalate (PET)/ethylene-butyl acrylate-glycidyl methacrylate copolymer polyester blend composites reinforced with carbon nanofibers (CNFs) and carbon nanotubes. Mechanical properties were significantly enhanced by dual-network structures made of CNTs at interfaces and CNFs inside the PET matrix. Network formation and nanofiller dispersion control allowed the optimized composite to achieve impressive increases in impact strength, modulus, and tensile strength. The potential for improving the fracture toughness of fiber-reinforced composites through nanoscale reinforcements, especially CNFs, is encouraging. The mode I fracture behavior of unidirectional carbon fiber epoxy composites is examined in this work in relation to CNF morphology, dispersion, and surface functionalization [17]. The dense cross-linked structure of epoxy resins causes brittleness despite their exceptional mechanical strength. The superior efficiency of nanofillers, which produce comparable strength improvements at much lower loadings, is highlighted in this review that contrasts the reinforcing effects of nano and non-nano fillers [18]. The enhancement achieved through nanofiller addition largely depends on its type, concentration, and uniform dispersion within the polymer matrix. Processing methods also play a crucial role in determining the overall performance improvement of the composite [19, 20].

The plain-weave design of natural-fiber mats (such as woven jute, woven flax, and woven kenaf) offers handling advantages and ideal in-plane isotropy, but it also exhibits common laminate failure modes (matrix cracking, fiber pull-out, and delamination) under impact and fracture loading. To improve fiber–matrix bonding, Prasad et al. coated flax fibers with nano-TiO_2 following silane treatment, enhancing flax fiber composites. Dynamic mechanical analysis, SEM, and FTIR analyses verified that the ideal coatings (0.4–0.6wt%) greatly enhanced interlaminar fracture toughness and storage modulus [21]. The mechanical improvement of kenaf mat/Ep composites by hybridization with glass and carbon fibers was examined by Malik et al[22].. When compared to both pure and glass-based hybrids, the kenaf mat/carbon/Ep hybrid demonstrated superior improvements in tensile, flexural, shear, and fracture toughness properties. By reinforcing the matrix-fiber interface and boosting energy dissipation during crack formation, the incorporation of nanofillers into the matrix (or as interleaves) or hybridization with nanomaterials targets those very failure modes [23, 24]. Shrivastava and Singh [25] reviewed interlaminar fracture toughness in glass and carbon fiber-reinforced composites under Mode-I and mode-II conditions. They investigated processing parameters, matrix modification, and fiber treatment as well as other elements that affect fracture behavior. CNT incorporation methods, such as grafting and interleaving, which greatly improve fracture toughness and delamination resistance, were highlighted in the study. The effects of nano-$CaCO_3$ on the mechanical and thermal behavior of epoxy composites reinforced with pineapple fiber were investigated by Mahadevaswamy and Suresha [26]. At 1–3wt% filler loading, the composites demonstrated the best gains in tensile, flexural, impact strength, and fracture toughness in addition to improved thermal stability. Measurable gains in impact behavior and fracture toughness with various nano-additives have been confirmed by recent experimental work on woven natural fiber mats [27, 28].

Table 1. A summary of recent literature on fracture toughness of nanofiller-filled natural fiber-reinforced epoxy composites.

Nanofiller filled composites	**Key Findings on Fracture Toughness**	**Ref.**
Epoxy with boron nitride, nS	Up to 2 wt% BN gave 101% increase in fracture toughness; SiO_2 provided sustained performance, reducing crack growth.	[29]
Carbon fiber-epoxy with GNPs, Cellulose nanofibers	Functionalized GNPs achieved up to 142% fracture toughness improvement; good interfacial bonding essential.	[30]
Natural Fibers with ZnO Nanofillers	ZnO nanofillers enhanced fracture toughness and mechanical performance of natural fiber-epoxy composites.	[31]
Natural Fibers with various Nanofillers	Synthesis methods and dispersion of nanofillers critically affect mechanical and fracture toughness properties.	[32]

The improvements in fracture toughness for natural fiber epoxy composites filled with nanofiller are compiled in Table 1. Fracture toughness is significantly increased by boron nitride (BN) and nS nanofillers at ideal loadings, with BN yielding the largest gains. Further enhancements can be made with functionalized graphene nanoplatelets and cellulose nanofibers, emphasizing the significance of interfacial bonding. Although dispersion and synthesis technique are crucial for maximizing performance, ZnO, and other nanofillers also greatly increase fracture toughness.

The impact resistance and fracture toughness of NF-RPMCs can be considerably improved by nanofillers. However, careful control of dispersion, viscosity, and interfacial interactions is required to provide consistent performance and scalability [33]. In their discussion of recent developments in the use of carbonaceous nanofillers to increase the fracture toughness of epoxy-based composites, Mousavi et al[34]. point out that dispersion and interface chemistry are essential for increasing mechanical properties. Cozza et al[35]. found that localized shear bands triggered by stress concentrations at the particle-matrix interface were the main toughening mechanisms in Ep matrix material containing silica nanoparticles.

Although PALF has promised mechanical and biodegradable characteristics, PALF/Ep composites frequently have limited toughness and brittle matrix-dominated failure. While it has been demonstrated that nanofillers including CNTs, GNPs, and nS improve fracture toughness in natural fiber-reinforced composites, nothing is known about how HNTs affect PALF/Ep systems. Moreover, micro-voids, micro-crazing, and shear-banding are examples of toughening mechanisms that have not been thoroughly studied in relation to HNT loading, dispersion, and fiber–matrix interfacial interactions. Consequently, the purpose of this study is to clarify the related microstructural mechanisms and modify the toughness of PALF/Ep composites by carefully including HNTs. Fabricating PALF/Ep hybrid composites with different HNT concentrations, assessing their impact resistance and fracture toughness, and figuring out the ideal nanofiller loading for best results are the goals. It is anticipated that the results will offer mechanistic understanding of toughening caused by nanofillers, paving the way for the creation of lightweight, high-performing, environmentally friendly composites that may be used in industrial and structural settings.

MATERIALS AND METHODS

Materials

Pineapple leaf fabric/Epoxy (PALF/Ep) composites were made using Ep resin, which is recognized for its strength, adaptability, low shrinkage, and insulation. Epoxy is a thermosetting resin widely used as a matrix in composite materials due to its excellent mechanical strength and strong adhesive properties. Its robust chemical resistance and ease of processing make it ideal for engineering and industrial applications. In the present work, Epoxy L12 (Lapox 12) with K6 hardener (Atul India Ltd., Gujarat) was used in this investigation. K6 hardener is a low-viscosity, aliphatic polyamine curing agent used for epoxy resins, enabling rapid cure and short pot life at ambient temperatures. Its properties make it ideal for applications, such as adhesives, castings, and laminates requiring efficient room-temperature processing. Table 2 provides a summary of the resin and hardening agent's main characteristics.

Table 2. Properties of epoxy (Lapox 12) and hardener (K6).

Properties	**Epoxy (L12)**	**Hardener (K6)**
Viscosity at 25 °C (cP)	1000–1200	10–20
Density at 25 °C (g/cm³)	1.16–1.20	0.94–0.95
Young’s modulus (GPa)	3.792	---
Tensile strength (MPa)	82.74	---
Pot life	6-9 h at 25°C	---
Glass transition temperature (°C)	120-140	
CTE (K^{-1})	$45\text{-}55 \times 10^{-6}$	

A tough and lightweight natural reinforcement is pineapple leaf fabric (PALF). It is rigid and strong due to its high cellulose content. The fibers form a strong bond with the epoxy matrix because of their rough surface. Strong bonding enhances overall mechanical performance and stress transfer. In addition to being environmentally beneficial, PALF is a good substitute for synthetic fibers. Alkali-treated (5wt% NaOH solution) pineapple leaf fiber (PALF; 64–67% cellulose) in plain weave form, purchased from Gogreen Products, Chennai, was utilized as reinforcement. It offered low density, biodegradability, and strong matrix bonding along with improved tensile strength (500–700 MPa), stiffness (25–40 GPa), and elongation at break of 1.5–2.5% [36].

Halloysite nanotubes (HNTs) are naturally occurring aluminosilicate nanomaterials with a tubular structure. They enhance the mechanical strength, toughness, and thermal stability of polymer composites by improving stress transfer and energy absorption. The source of HNTs was Sigma Aldrich in Bengaluru, India. The HNTs have a cylindrical shape, measuring between 1 and 3 μm in length and on average of 30 to 70 nm in diameter. To improve the mechanical and thermal properties of polymer composites, they can be used as nanofillers because of their stated density of 2.53 g/cm3 and specific surface area of 64 m²/g.

Fabrication

Pineapple leaf fiber-reinforced epoxy (PALF/Ep) composites filled with halloysite nanotubes (HNTs) were fabricated using hand layup and ultrasonication techniques to achieve uniform nanofiller dispersion. Ultrasonication was employed specifically to prevent HNT agglomeration and ensure enhanced mechanical properties. This process results in composites with improved tensile, flexural, and impact strengths due to optimal filler-matrix interaction. To ensure even dispersion without overheating, epoxy, and HNTs were ultrasonically sonicated for 1 h at 22 kHz in an ice bath. After being trimmed to size, PALF mats were laminated and impregnated with a 10:1 ratio of ultrasonicated Ep/HNT containing K6 hardener. Four PALF mat layers were stacked, post-cured for 1 h at 80°C, then cured for 24 h at 0.5 MPa. A thread-cutting machine was used to produce test specimens from the cured laminates. The four series of PALF/Ep hybrid composites that were manufactured are listed in Table 3 and schematically illustrated in Figure 1.

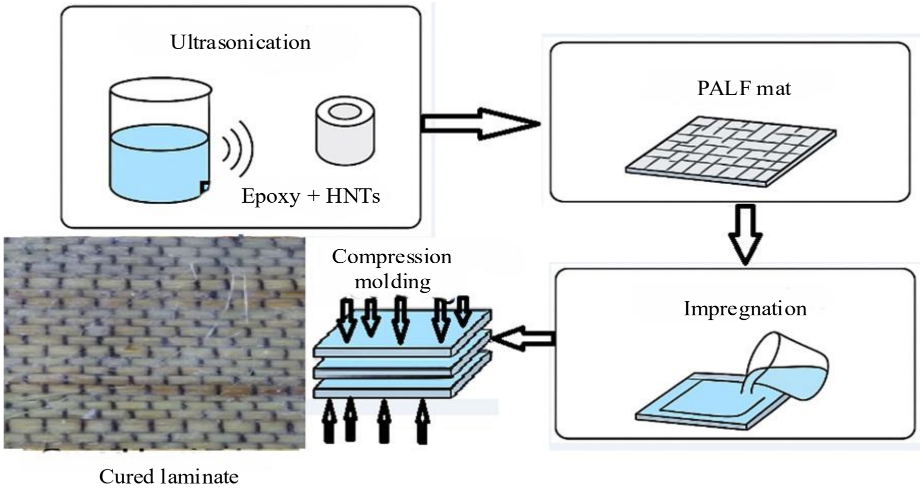

Figure 1. Schematic representation of the fabrication process for HNTs-reinforced PALF/Ep composites.

Table 3. Series of the fabricated composites studied in this work.

Composites	**PALF mat (wt%)**	**Epoxy (wt%)**	**HNT (wt%)**
PALF/Ep	30	70	---
PALF/Ep + 1 wt%	30	69	1
PALF/Ep + 2 wt%	30	69	2
PALF/Ep + 3 wt%	30	69	3

Testing

The Izod impact test was conducted as per ASTM D256 [37] to assess the impact strength of PALF/Ep composites both with and without halloysite nanotubes (HNTs). Standard notched specimens were prepared and subjected to a pendulum impact in controlled conditions. The energy absorbed at fracture was measured, providing comparative toughness values for each composite formulation. Results indicated the reinforcing effect of HNTs on the composite's impact resistance. This method involves clamping a rectangular-cross-sectioned specimen with a notch vertically in the testing apparatus, so that the notch faces the pendulum. A swinging pendulum hammer strikes the specimen, shattering it instantly. The device measures and displays in joules the amount of energy absorbed by the specimen during fracture in the Izod impact test. This value directly reflects the material's toughness and resistance to sudden impact. The resistance of the material to abrupt impact loading is reflected in this absorbed energy. To maintain uniformity and exclude external influences on the fiber reinforcement and polymer matrix, all specimens were conditioned and evaluated at a regulated temperature of 23 °C.

As per ASTM D5045 [38], Single Edge Notch Bending (SENB) specimens were used to assess the fracture toughness of PALF/Ep composites. To replicate Mode-I crack growth, notched rectangular samples were loaded in three-point bending. The critical stress intensity factor (KIC), a measure of the material's resistance to fracture propagation, is determined by the test. Standard formulas that take into consideration the specimen's geometry, notch length, and span are used to gather and analyze load and displacement data. Environmental controls that guarantee reproducibility and accurate comparison between HNT-filled and unfilled composites include testing at 23 °C. Understanding fracture mechanisms and maximizing composite durability for engineering applications are made easier with the help of the SENB method.

Table 4. Impact strength of HNTs-reinforced PALF/Ep composites.

Composites	**HNT loading(wt%)**	**Impact strength(J/m)**	**Increase in Impact strength (%)**
PALF/Ep	0	222.3 ± 4.2	---
PALF/Ep + 1 wt%	1	418.2 ± 2.2	88.1
PALF/Ep + 2 wt%	2	433.9 ± 3.1	95.2
PALF/Ep + 3 wt%	3	421.5 ± 5.2	89.6

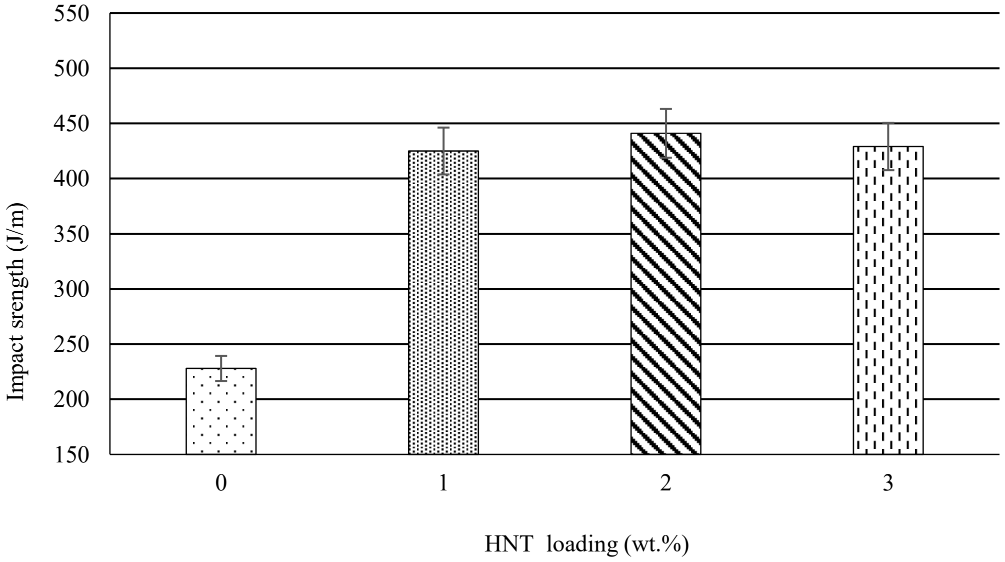

Figure 2. Enhanced impact strength of PALF/Ep with HNTs.

RESULTS AND DISCUSSION

Results of HNTs on Impact Strength of PALF/Ep Composites

According to the Izod impact test results (Table 4) and Figure 2, the addition of halloysite nanotubes (HNTs) significantly enhanced the impact strength of PALF/Ep composites when compared to the unfilled composite. When 1wt% HNTs were added, the impact strength of the unfilled composite, which was 222.3 J/m, nearly quadrupled to 418.2 J/m, representing an improvement of nearly 88.1%. An additional 2wt% HNT loading resulted in the highest impact strength, 433.9 J/m, which was 95.2% better than unfilled ones. This implies that the nanotubes significantly affect energy absorption and crack deflection under impact loading. While it still improved by 89.6% over the control, the impact strength significantly fell to 421.5 J/m at 3wt% loading.

Discussion of HNTs on Impact Strength of PALF/Ep Composites

The PALF/Ep composite's impact strength increased significantly from 222.3 J/m (0wt% HNTs) to a peak of 433.9 J/m at 2wt% HNTs. It then slightly decreased to 421.5 J/m at 3wt%, indicating optimal reinforcement at 2wt% due to uniform nanotube dispersion; higher loading lead to agglomeration and decreased performance [39]. Similar trends have been documented by Krishnaiah et al[40]., and Rasana et al[41]., who found that HNTs improve energy absorption and impact strength through mechanisms like energy dissipation and crack deflection, with performance decreasing after an ideal filler concentration. While Ashok et al[42]. reported similar improvements in luffa fiber/Ep systems reinforced with nano lead oxide, On the other hand, filler agglomeration caused by excessive fine particle additions, as nano tamarind shell ash in water hyacinth fiber/Ep composites, decreased impact strength [43]. In the present work, the homogeneous dispersion of nanotubes throughout the PALF/Ep composite is responsible for the notable improvement in impact strength at low HNT levels (1–2wt%). By stabilizing microcracks and encouraging energy dissipation through processes such nanotube bridging, pull-out, and crack pinning, these evenly distributed nanotubes improve energy absorption [44]. Strong interfacial bonds between HNTs and the polymer matrix enable effective stress transfer and crack-arresting action, according to Idumah et al[39].. In a similar vein, Stern et al[45]. emphasized that energy-dissipating interactions between nanoparticles and the fracture front enhance toughness. Additionally, Aguiar et al[46]. and Churruca et al[47]. revealed that HNTs improve toughness, boost micro-crack resistance, and refine the composite microstructure. Similar increases in impact strength were noted by Franciszczak et al[48]. for polymer nanocomposites with modest HNT loadings. With just 3wt% HNTs, the PALF/Ep composite in this study increased impact strength by 1.89 times, from 222.3 J/m to 421.5 J/m. This finding demonstrates that substantial toughening, akin to rubber-toughened systems, can be accomplished at low filler concentrations. In line with recent findings [39–48], the current work supports these findings by showing that uniformly distributed nanotubes stabilize microcracks and improve energy absorption through mechanisms like nanotube bridging, pull-out, and crack pinning.

Results of HNTs on Fracture Toughness of PALF/Ep Composites

The fracture toughness of PALF/Ep composites clearly improves with increasing halloysite nanotube (HNT) concentration, as seen in Figure 3 and Table 5. The unfilled composite exhibited a fracture toughness of 2.91 ± 0.3 MPa·$m^{1/2}$, which increased to 4.76 ± 0.4 MPa·$m^{1/2}$ at 1wt% and reached a maximum of 4.92 ± 0.2 MPa·$m^{1/2}$ at 2wt% HNTs, representing a 69.1% improvement. The uniform dispersion of nanotubes within the epoxy matrix facilitates efficient stress transfer and crack-arresting mechanisms, which are responsible for this improvement. At 3wt%, there was a modest drop to 4.48 ± 0.3 MPa·$m^{1/2}$, which was probably caused by nanotube aggregation impeding crack resistance. mechanical performance is reached at modest filler loadings, beyond which aggregation lowers dispersion quality and diminishes property improvements [49, 50]. These findings are consistent with prior reports on HNT-filled epoxy systems, which indicate that optimum.

Discussion of HNTS on Fracture Toughness of PALF/Ep Composite

Toughening mechanisms like pull-out, nanotube bridging, and micro-crack deflection are responsible for the observed increase in fracture toughness. Recent research works confirms that the best possible dispersion of HNTs in the epoxy matrix improves energy absorption and stress transfer during crack propagation, leading to an effective barrier against crack growth. In the work of Muralidhara et al[51]., HNTs greatly increase the toughness of epoxy composites reinforced with carbon fabric by enhancing load transfer and strengthening interfacial bonding. These features also help crack pinning, nanotube bridging, and resistance to matrix separation, which raises energy and durability under mechanical stress. While silicon carbide contributes to improved tensile and flexural properties but has a relatively moderate effect on toughness, graphite filler greatly increases the Mode-I fracture toughness of glass fabric reinforced epoxy composites by encouraging crack deflection and energy dissipation [52]. Mechanisms like the formation of a large deformation zone, crack pinning, and crack deflection, which improve energy dissipation and obstruct crack propagation, are largely responsible for the increased fracture toughness of graphene/epoxy nanocomposites [53]. Mechanisms like crack bridging, crack deflection, and plastic deformation of the epoxy matrix surrounding HNT clusters are primarily responsible for the increased toughness of HNT-modified/Ep nanocomposites. These mechanisms interact with cracks and prevent their propagation, resulting in notable increases in fracture toughness without sacrificing other mechanical properties [54]. According to Ulus et al[55]., adding 2wt% HNTs significantly improved toughness, raising the Mode-I interlaminar fracture toughness by 18% and the stress intensity factor by 43%. Strong interfacial bonding and crack-bridging mechanisms that improved resistance to delamination even after extended seawater aging were credited with this development. While adding inorganic rigid particles simultaneously improves mechanical strength and toughness through plastic deformation of the matrix surrounding the particles, crack pinning, deflection, and crack tip pinning, adding a second thermoplastic or rubber phase to epoxy resin significantly increases fracture toughness by dissipating energy through yielding of the second phase [56]. Fracture toughness and impact strength in natural fiber composites are greatly increased by enhancing interfacial adhesion through appropriate physical and chemical fiber treatments, according to Maharama and Sendur [57]. Stronger fiber-matrix bonding, which encourages effective stress transmission and prevents crack initiation and propagation, is the cause of this improvement. Successful toughening also depends on good dispersion and strong interfacial bonding, with graphene and carbon nanotubes providing high fracture toughness improvements without compromising other properties [58, 59].

Table 5. Fracture toughness of HNTs-reinforced PALF/Ep composites.

Composites	**HNT loading (wt%)**	**Fracture toughness (MPa m$^{1/2}$)**	**Fracture toughness improvement (%)**
PALF/Ep	0	2.91 ± 0.3	---
PALF/Ep + 1 wt%	1	4.76 ± 0.4	63.6
PALF/Ep\ + 2 wt%	2	4.92 ± 0.2	69.1
PALF/Ep + 3 wt%	3	4.48 ± 0.3	54.0

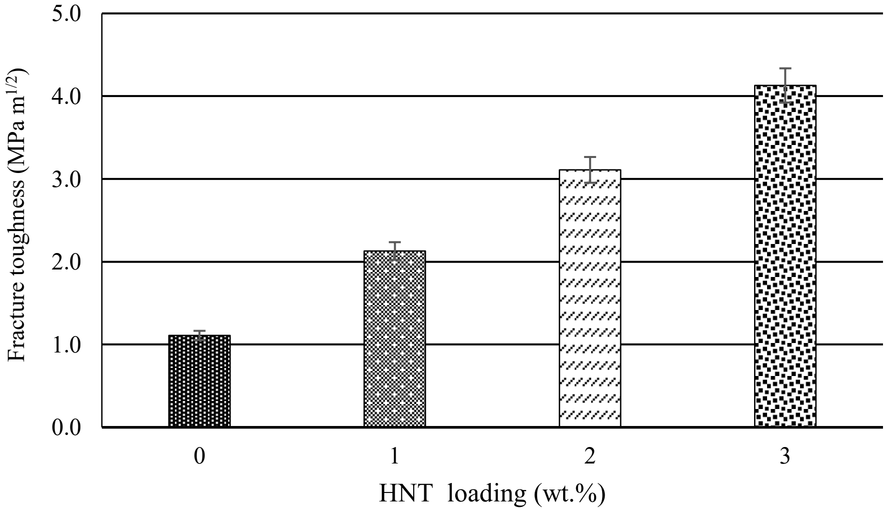

Figure 3. Enhanced Fracture Toughness of PALF/Ep with HNTs.

In this study, HNTs enhanced energy absorption and stress transfer during crack propagation, thereby significantly increasing the fracture toughness of PALF/Ep composites. The toughness was significantly increased because of the effective resistance to crack growth provided by the toughening mechanisms, which included crack bridging, nanotube pull-out, and micro-crack deflection. In line with recent research findings, this improvement is ascribed to the high interfacial bonding and excellent dispersion of HNTs within the epoxy matrix.

Finally, by using several synergistic toughening methods, the addition of HNTs greatly increased the fracture toughness of PALF/Ep composites in the current work. Effective resistance to crack initiation and propagation was provided by the well-dispersed HNTs, which improved energy absorption and stress transfer throughout the matrix. Important processes like pull-out, micro-crack deflection, and nanotube bridging helped create a sizable plastic deformation zone that dispersed fracture energy. Effective load transmission and crack tip blunting were further made possible by strong interfacial adhesion between HNTs and the epoxy matrix. Overall, the fracture toughness of the composites was significantly increased without sacrificing other mechanical properties due to the combined impacts of uniform dispersion, interfacial adhesion, and energy-dissipative mechanisms.

CONCLUSIONS

The improvement of mechanical properties in PALF/Ep composites by adding halloysite nanotubes (HNTs) was methodically examined in this work. The study offers important new information about the best filler loadings and toughening processes that enhance fracture toughness and affect performance. The main concluding remarks are as follows:

The impact strength of PALF/Ep composites was significantly increased by the addition of HNTs, with the greatest improvement of 95.2% noted at 2wt% loading because of homogenous nanotube dispersion. Under impact loading, toughening mechanisms like pull-out, crack pinning, and nanotube bridging improved energy absorption and offered strong resistance to crack propagation. Nanotube agglomeration caused minor reductions in impact strength at higher loadings (3wt%), demonstrating that maximum energy absorption is attained at moderate HNT loadings. The fracture toughness of PALF/Ep composites was greatly enhanced by the inclusion of HNTs, reaching a 69.1% increase at 2wt% because of uniform dispersion and strong interfacial bonding. Effective resistance to fracture initiation and propagation was given by toughening mechanisms like nanotube pull-out, bridging, and micro-crack deflection, which also improved energy absorption. Agglomeration decreased performance above the optimal HNT loading of 2wt%, indicating that a moderate filler content ensures the ideal ratio of strength to toughness. The future work will focus on optimizing HNT surface treatments and studying environmental durability, while exploring hybrid fillers and scaling up manufacturing processes. Additionally, investigating fatigue behavior, developing predictive models, and assessing sustainability will advance the application of PALF/Ep/HNT composites.

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