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Hygrothermal and Mechanical Performance of Carbon Nanotube–Natural Fiber Hybrid Composites: A Comprehensive Theoretical and Critical Study

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Abstract: Background: Hybrid composites that combine carbonaceous nanofillers such as carbon nanotubes (CNTs) with natural fibers have attracted significant interest for structural and multifunctional applications because they promise the stiffness and multifunctionality of nanocarbon reinforcements alongside the low density, sustainability, and toughness benefits of natural fibers (Balasubramanian and Burghard, 2005; Santulli, 2019). However, the behavior of these hybrid systems under combined environmental stressors — particularly moisture uptake and hygrothermal cycling — and the corresponding effects on viscoelasticity, creep, interfacial stress transfer, and damage evolution remain incompletely understood. Moisture interacts with polymer matrices, fiber surfaces, and nanoscale interfaces in ways that can produce reversible and irreversible changes in mechanical performance (Ferguson and Qu, 2006; Zhang and Wang, 2006; Athijayamani et al., 2009).

Objectives: This article synthesizes the theoretical frameworks and empirical findings relevant to the hygrothermal response and mechanical performance of CNT–natural fiber hybrid composites. It aims to (1) elucidate the mechanisms by which moisture and temperature influence matrix, fiber, and interface

behavior; (2) integrate knowledge on time-dependent phenomena such as creep and viscoelastic damping in the presence of nanocarbon fillers and natural fibers; (3) identify principal modeling approaches and experimental strategies to assess durability; and (4) propose a structured research methodology and interpretive framework for future high-fidelity experimental and computational studies.

Methods: The work undertakes an exhaustive theoretical elaboration, drawing on nanoscale functionalization studies for CNTs, experimental investigations into hygrothermal effects on carbon-fiber and natural-fiber composites, and research on creep and damping in polymer and nanocomposite systems (Balasubramanian and Burghard, 2005; Yizhuo et al., 2014; Jia et al., 2011; Tehrani et al., 2011). Mechanistic paths are delineated using continuum and multiscale reasoning — from molecular interactions at functionalized CNT surfaces to macroscopic composite constitutive responses — and critical analysis is applied to reconcile apparently conflicting empirical observations. The Methods section describes rigorous, purely textual experimental protocols and modeling pathways that would form a robust research program.

Results: A composite of theoretically derived outcomes indicates that: (a) chemically functionalized CNTs alter local hydrophilicity/hydrophobicity and interfacial energy landscapes, influencing water diffusion pathways and stress transfer (Balasubramanian and Burghard, 2005); (b) moisture-induced plasticization and hydrolysis in typical thermoset and thermoplastic matrices lead to a complex interplay of reversible softening and irreversible matrix degradation that scales with exposure time, temperature, and fiber/matrix chemistry (Ferguson and Qu, 2006; Jen and Huang, 2013); (c) natural fibers impart additional moisture sensitivity through capillarity and bound water within cellulosic microstructures, leading to swelling-driven internal stresses that can degrade fiber–matrix bonding (Athijayamani et al., 2009; Sarkar et al., 2010); and (d) CNTs can either mitigate or exacerbate hygrothermal damage depending on functionalization, dispersion, and interfacial coupling strategies (Zhang and Wang, 2006; Tehrani et al., 2011). Time-dependent behaviors such as creep and damping are highly sensitive to moisture content and interfacial integrity; nano-reinforcement

tends to raise stiffness and retard creep at low to moderate moisture contents but may accelerate localized stress concentrations and crack nucleation under severe hygrothermal exposure (Jia et al., 2011; Yao et al., 2013).

Conclusions: The hybridization of CNTs and natural fibers offers pathways to design composites with tunable hygro-mechanical performance, but realizing reliable, durable materials demands integrated approaches that combine chemical engineering of interfaces, precise control of microstructure, and multiscale predictive modeling. Continued experimental efforts using standardized hygrothermal protocols and advanced nanoscale characterization, coupled with constitutive models that include moisture-dependent viscoelasticity and interfacial debonding, are essential to translate laboratory promise into engineering practice (Mulenga et al., 2021; Alhijazi et al., 2020). This article provides a detailed conceptual and methodological roadmap for such research.

Keywords: Carbon nanotubes; natural fibers; hygrothermal effects; creep; interfacial stress; viscoelastic damping

Introduction

The pursuit of composite materials that marry high specific stiffness, environmental sustainability, and multifunctionality has catalyzed intense academic and industrial activity over the past two decades. Two strands of development stand out. First, carbon nanotubes (CNTs) have been studied extensively for their extraordinary mechanical, electrical, and thermal properties as nanoscale reinforcements in polymer matrices, with chemical functionalization enabling improved dispersion and interfacial bonding (Balasubramanian and Burghard, 2005). Second, natural fibers — cellulose-based reinforcements obtained from plants — offer low density, renewability, and favorable toughness attributes which make them attractive as a sustainable alternative to synthetic fibers in many load-bearing and semi-structural applications (Santulli, 2019; Yamini et al., 2023). The hybrid composite concept, wherein CNTs and natural fibers are combined within a polymer matrix, aims to harness synergies: CNTs enhance stiffness, conductivity, and local stress transfer; natural fibers contribute bulk reinforcement

and energy-absorption capabilities while lowering embodied carbon (Mahesh et al., 2020; Thomason and Rudeiros-Fernández, 2018).

Despite the compelling design rationale, hybrid systems introduce complex multi-physics durability challenges. Moisture ingress — by vapor uptake, capillarity, or liquid exposure — modifies matrices, fiber cell walls, fiber surfaces, and nanoscale interfaces in ways that can reduce stiffness, lower strength, induce swelling, and alter time-dependent behaviors like creep and viscoelastic damping (Ferguson and Qu, 2006; Zhang and Wang, 2006). The presence of CNTs further complicates the picture: functionalization can change CNT wettability and bonding (Balasubramanian and Burghard, 2005), but CNT aggregates or poorly bonded nanotubes can act as stress concentrators, promoting microcracking under hygrothermal cycling (Tehrani et al., 2011). For natural fibers, hygroscopicity is intrinsic; cellulosic microfibrils and hemicellulose absorb water and swell, generating internal stress fields and weakening fiber–matrix adhesion (Athijayamani et al., 2009; Sarkar et al., 2010). Thus, predicting long-term performance of CNT–natural fiber hybrid composites requires a detailed understanding of interacting mechanisms across scales.

Two major knowledge gaps motivate this work. First, while many studies characterize moisture effects in carbon-fiber composites or in natural-fiber systems individually, fewer works address hybrid systems that combine CNTs with natural fibers, particularly under simultaneous thermal loading and long-duration exposures (Zhang and Wang, 2006; Yizhuo et al., 2014). Second, time-dependent mechanical responses — creep, recovery, and dynamic damping — are frequently studied in dry conditions, leaving open how moisture alters viscoelastic spectra, relaxation times, and damage evolution in composites containing both nano- and micro-scale reinforcements (Jia et al., 2011; Tehrani et al., 2011). Filling these gaps is essential for deploying such materials where environmental exposure is unavoidable, for instance in automotive, marine, and building applications (Mulenga et al., 2021; Alhijazi et al., 2020).

This article presents a deep, theory-driven synthesis of the state of knowledge on hygrothermally influenced mechanical behavior in CNT–natural fiber hybrid composites, integrating nanoscale interfacial chemistry, microstructural moisture transport, and macroscopic

viscoelastic and damage mechanics. The intended audience includes materials researchers, composite analysts, and engineers designing environmentally robust hybrid composites. The remainder of the introduction outlines the scope and organization: subsequent sections describe a detailed text-based methodology for experimental and modeling studies, present descriptive analytical results and mechanistic conclusions informed by the referenced literature, discuss limitations and future research directions, and conclude with key takeaways for materials design.

Methodology

Given the textual nature of this article, the Methods section provides a comprehensive blueprint — conceptual, experimental, and theoretical — that a researcher would follow to investigate hygrothermal and mechanical behaviors in CNT–natural fiber hybrid composites. The methodology is intentionally exhaustive, specifying materials choices, surface chemistry strategies, specimen preparation, standardized hygrothermal conditioning regimens, mechanical testing protocols, nanoscale and microscale characterization techniques, and multiscale modeling frameworks. Each element is motivated by findings in the cited literature and designed to isolate and quantify the critical mechanisms identified in the literature review.

Materials selection and functionalization strategy. The hybrid composite system comprises three components: a polymer matrix (either a thermoset epoxy or thermoplastic such as polypropylene), natural fibers (e.g., sisal, jute, flax), and multiwall or single-wall carbon nanotubes (CNTs). The matrix selection reflects common engineering practice: epoxy matrices are widely used in high-performance composites and serve as a benchmark for hygrothermal degradation studies, while polypropylene allows examination of thermoplastics where melt processing and different moisture-matrix interactions predominate (Ferguson and Qu, 2006; Jia et al., 2011). Natural fiber selection should prioritize fibers with documented mechanical properties and moisture sensitivities, such as sisal or flax, which have been widely studied in hybrid contexts (Athijayamani et al., 2009; Yamini et al., 2023).

CNT functionalization is a pivotal variable. Covalent and non-covalent functionalization routes are both relevant: covalent grafting of functional groups (e.g., carboxyl, hydroxyl, amine) modifies CNT surface energy and enables chemical coupling to epoxy networks or compatibilizers in thermoplastics (Balasubramanian and Burghard, 2005). Non-covalent approaches (e.g., polymer wrapping or surfactant stabilization) preserve CNT intrinsic properties but rely on physical adsorption that can be less robust under hygrothermal stress. The methodological plan requires preparing three CNT variants: pristine, covalently functionalized, and polymer-wrapped, to compare how interfacial chemistry influences moisture transport and mechanical durability (Balasubramanian and Burghard, 2005; Tehrani et al., 2011).

Specimen fabrication and microstructure control. Achieving reproducible microstructures is critical. For epoxy-based systems, a carefully controlled hand lay-up or resin infusion followed by vacuum curing minimizes void content. CNT dispersion must be optimized: ultrasonication, three-roll milling, or high-shear mixing of CNTs in the resin — with and without dispersing agents — should be systematically varied to generate samples with low, medium, and high degrees of CNT aggregation. Natural fibers should be used either as randomly oriented mats or aligned bundles to represent different application scenarios (Athijayamani et al., 2009; Santulli, 2019). For thermoplastic matrices, melt compounding with compatibilizers and twin-screw extrusion is recommended, ensuring uniform CNT distribution and fiber premixing.

Standardized hygrothermal conditioning. To interrogate moisture effects, specimens must be placed under controlled relative humidity (RH) and temperature (T) conditions. A set of regimen tiers is proposed: (1) low-level exposure (30–50% RH at 23°C) to simulate ambient conditions; (2) moderate exposure (65–85% RH at 40–60°C) to simulate warm, humid service environments; and (3) aggressive exposure (immersion in water at elevated temperatures, e.g., 60–80°C) to probe severe hygrothermal stress and accelerated aging. Cyclic exposures — alternating humid and dry cycles, or thermal cycling with constant humidity — should be included to observe hysteresis, permanent changes upon re-drying, and fatigue-like behaviors (Ferguson and Qu, 2006; Jen and Huang, 2013).

Moisture uptake quantification and diffusion analysis. Researchers should measure mass gain as a function of exposure time to determine diffusion kinetics. Experimental data should be compared to classical Fickian diffusion models and extended diffusion formulations that account for dual-mode sorption and hygroscopic swelling in natural fibers. The methodology calls for fitting sorption curves to estimate effective diffusion coefficients, equilibrium moisture content, and transient diffusion regimes. Particular attention must be paid to non-Fickian behaviors that arise from coupled mechanical swelling and time-dependent matrix relaxation (Zhang and Wang, 2006; Athijayamani et al., 2009).

Mechanical testing under controlled moisture content. Mechanical characterization must be performed on specimens equilibrated to specified moisture states and after re-drying. Quasi-static tests include tensile, flexural, and interlaminar shear strength (ILSS) tests to assess stiffness, strength, and interfacial integrity. Dynamic mechanical analysis (DMA) should be performed to obtain storage modulus, loss modulus, and tan delta across a frequency and temperature sweep in dry, humid, and wet states, providing insight into viscoelastic damping and glass transition shifts (Jia et al., 2011; Tehrani et al., 2011). Time-dependent testing for creep and recovery — constant stress or constant strain hold tests over extended durations — should quantify rate-dependent deformation in different hygrothermal states (Jia et al., 2011; Yao et al., 2013).

Multiscale characterization of interfacial and microscale damage. A battery of microscale and nanoscale characterization techniques is required to identify damage modes and interfacial changes induced by moisture. Scanning electron microscopy (SEM) of fracture surfaces, atomic force microscopy (AFM), and nanoindentation can reveal fiber surface degradation, matrix softening, and local modulus variations. Spectroscopic methods (e.g., Fourier-transform infrared spectroscopy, FTIR) detect chemical changes such as hydrolysis or oxidation. For CNTs and their functional groups, Raman spectroscopy and X-ray photoelectron spectroscopy (XPS) provide valuable chemical state information. These techniques help correlate macroscopic mechanical property changes to microstructural and

chemical alterations (Balasubramanian and Burghard, 2005; Yizhuo et al., 2014; Tehrani et al., 2011).

Modeling approaches: constitutive laws and damage mechanics. The methodology prescribes developing a hierarchy of constitutive models starting from moisture-dependent linear viscoelasticity to nonlinear, damage-inclusive frameworks. At the microscale, models must represent moisture-dependent matrix modulus, fiber swelling strains, and interfacial bond degradation. At the mesoscale, representative volume element (RVE) analysis with explicit CNT and fiber morphologies can be used to extract effective properties and simulate stress concentrations around fibers and nanotube aggregates. At the macroscale, continuum damage mechanics models with internal variables for moisture content, interfacial damage fraction, and creep strain permit simulation of long-term performance (Mulenga et al., 2021; Alhijazi et al., 2020). The methodology emphasizes coupling diffusion equations with mechanical response — i.e., poroelastic or chemo-mechanical coupling — to capture swelling-induced stresses and non-Fickian transport.

Experimental design and statistical considerations. The methodology must ensure that sufficient sample numbers are used to capture variability intrinsic to natural fibers and CNT dispersions. A factorial experimental design that varies CNT functionalization, CNT loading, fiber type and volume fraction, matrix chemistry, and hygrothermal exposure level is advocated. Statistical analysis (ANOVA, regression) will identify dominant factors and interactions affecting durability metrics such as stiffness retention, strength retention, and time-to-failure under creep.

Ethical and sustainability considerations. While the focus is technical, the methodology addresses lifecycle and sustainability implications: sourcing of natural fibers, environmental impact of CNT production, and end-of-life options for composites. These aspects inform material selection and highlight trade-offs between performance and sustainability (Santulli, 2019; Yamini et al., 2023).

By following this structured methodology, researchers can systematically probe the interplay among chemical functionalization, moisture transport, microstructural damage, and time-dependent mechanical responses

that define the service life of CNT–natural fiber hybrid composites.

Results

Because this article is a synthetic, text-based research treatise rather than a report of new experimental data, the Results section aggregates and elaborates on mechanistic findings, integrating observations and conclusions from the cited literature into a coherent set of outcomes that a rigorous experimental program would produce if the methodological blueprint were executed. Each descriptive result is supported by the literature and by logical inference grounded in multiscale mechanics.

1. Chemical functionalization of CNTs modifies water interaction pathways and interfacial stress transfer. Covalently functionalized CNTs present polar groups that can engage in hydrogen bonding with hydrophilic matrix components or with treated natural fiber surfaces. Balasubramanian and Burghard (2005) demonstrated that such chemical functionalization increases the effective interfacial area for stress transfer and enhances load transfer when chemical coupling is favorable (Balasubramanian and Burghard, 2005). However, polar functional groups also increase local hydrophilicity, potentially creating preferential sorption sites for water molecules. Consequently, functionalized CNTs can both improve mechanical coupling and create localized zones where moisture-induced plasticization or hydrolytic degradation initiates. This duality implies a design trade-off: optimized functionalization that balances mechanical coupling with controlled hydrophobicity or crosslinking density is essential for durability.
2. Moisture induces matrix plasticization, glass transition depression, and permanent modulus loss under re-drying. Studies in epoxy underfills and similar thermoset matrices show that absorbed moisture acts as a plasticizer, lowering the effective glass transition temperature and reducing modulus and strength (Ferguson and Qu, 2006; Jen and Huang, 2013). When specimens are re-dried, part of the mechanical degradation is

often not fully reversible: polymer chain scission, hydrolytic reactions at vulnerable chemical sites, and microcracking generated during swelling lead to permanent reductions in stiffness and strength (Ferguson and Qu, 2006). Jen and Huang (2013) highlighted that combined temperature and moisture exposures exacerbate these effects, accelerating chemical degradation pathways and increasing the irreversibility of property loss.

3. Natural fibers present complex moisture storage and swelling behaviors that drive interfacial debonding. Natural fibers contain amorphous hemicellulose and lignin phases and crystalline cellulose microfibrils. Bound water in hemicellulose and free water in microvoids contribute to multi-mode sorption and swelling (Athijayamani et al., 2009). Swelling generates internal stresses at the fiber–matrix interface, especially when fibers are constrained by a relatively stiff matrix. Over time and repeated moisture cycles, these stresses lead to fiber–matrix debonding, fiber–matrix gap formation, and eventual reductions in composite stiffness and load-bearing capacity (Athijayamani et al., 2009; Sarkar et al., 2010). The capillary action within fiber bundles can also accelerate liquid ingress in immersion scenarios, which is particularly pernicious in hybrid composites where CNTs create highly heterogeneous microstructures.

4. CNTs can reduce bulk creep at low to moderate moisture but may intensify local stress concentrations under high moisture content or poor dispersion. Nanoscale reinforcement with CNTs generally increases stiffness and can reduce creep rates in polymer matrices by constraining chain mobility and providing load-bearing pathways (Jia et al., 2011; Tehrani et al., 2011). For polyurethane and similar polymers, Yao et al. (2013) reported reduced creep with CNT addition under dry conditions. However, under significant moisture absorption, matrix plasticization can lower the effectiveness of CNT reinforcements, and CNT aggregates act as stress concentrators that promote microcracking and localized debonding under cyclic hygrothermal loading (Tehrani et al., 2011). Thus, dispersion quality and interfacial bonding are critical to ensuring that CNTs confer long-term creep resistance in humid environments.

5. Combined hygrothermal loading modifies interfacial stress transfer characteristics of CNT-reinforced systems. Zhang and Wang (2006) analyzed how moisture and temperature jointly affect stress transfer in CNT-reinforced composites, noting that both diffusion-driven swelling and temperature-dependent softening alter the interfacial shear transfer capacity (Zhang and Wang, 2006). Molecular-level changes in the resin — such as chain mobility and debonding potential — interact with mechanical stresses to produce time-dependent loss in interfacial efficiency. In hybrid systems, the presence of natural fibers introduces additional, moisture-driven deformation that redistributes stresses to CNT–matrix–fiber junctions, making the effective interfacial network highly heterogeneous.

6. Nonlinear dynamic responses in laminated and complex geometries amplify hygrothermally induced destabilization. Shells and curved layered structures with imperfections are particularly sensitive to hygrothermally induced property changes; small reductions in stiffness or interlaminar strength can precipitate buckling or localized delamination under service loads (Nanda and Pradyumna, 2011; Shlykov et al., 2022). These effects are compounded in hybrid laminates where natural fiber plies and CNT-rich plies coexist, giving rise to mismatches in hygrothermal expansion and time-dependent deformation that can trigger damage modes absent in homogeneous laminates (Nanda and Pradyumna, 2011).

7. Macro- and micro-interfacial properties evolve differently under hygrothermal exposure. Yizhuo et al. (2014) demonstrated that macro-scale interfacial properties such as measured ILSS degrade with moisture exposure, while micro-interfacial properties — as probed by nanoscratch or microbond tests — can reveal more detailed debonding mechanisms and localized failure modes (Yizhuo et al., 2014). For hybrid

composites, the micro-interfacial response around CNTs and fiber surfaces governs early damage initiation, whereas macro-interfacial properties control global failure progression. Understanding both scales is requisite for predictive modeling.

8. Time-hardened degradation: cumulative exposure leads to synergetic deterioration beyond simple additive effects. Several studies indicate that the damage from combined moisture and temperature is not simply the sum of individual effects; instead, coupling leads to synergistic phenomena where moisture accelerates thermal oxidation or where thermal softening increases susceptibility to swelling-induced debonding (Jen and Huang, 2013; Ferguson and Qu, 2006). Consequently, durability assessments must consider such coupling explicitly rather than relying on separate fade curves for temperature and moisture.

9. Mechanical damping and vibrational characteristics are strongly moisture-dependent; hybridization shifts viscoelastic spectra. Dynamic mechanical analyses reveal that absorbed water reduces storage modulus and shifts relaxation peaks, altering both energy storage and dissipation characteristics. CNTs can increase storage modulus and change the damping peak pattern, while natural fibers, by virtue of damping through internal friction and microfibril slipping, influence loss moduli in a moisture-dependent fashion (Jia et al., 2011; Tehrani et al., 2011). For structural applications involving dynamic loads or acoustic considerations, these changes are engineering-significant.

10. Modeling frameworks that couple diffusion, swelling, viscoelasticity, and damage are necessary to capture the observed behaviors. Authors advocating multiscale and chemo-mechanical coupling approaches provide convergent evidence that only models incorporating moisture-dependent constitutive parameters, damage evolution laws for interfaces, and time-dependent viscoelastic response can reproduce the non-Fickian sorption, hysteresis, and permanent property losses observed experimentally (Mulenga et al., 2021; Alhijazi et al., 2020).

Collectively, these results point toward a nuanced design space where CNT functionalization, fiber treatment, matrix chemistry, and microstructural control co-determine the composite's hydro-mechanical performance. The following Discussion section expands on these inferences, evaluates limitations of current knowledge, and proposes specific future research avenues.

Discussion

This section interprets the descriptive results with an emphasis on mechanistic synthesis, trade-offs in materials design, limitations of current research, and recommendations for future work. The aim is to provide a critical roadmap for researchers and engineers seeking to deploy CNT–natural fiber hybrid composites in environments where moisture and temperature are relevant.

Mechanistic synthesis: balancing interfacial chemistry and moisture affinity. The dual role of CNT functionalization emerges as a central theme. Covalent functional groups enhance load transfer but increase local hydrophilicity, thereby creating potential initiation sites for moisture-induced degradation (Balasubramanian and Burghard, 2005; Tehrani et al., 2011). Conversely, non-covalently modified CNTs preserve intrinsic properties but may be less effective in translating nanoscale stiffness to the matrix under hydrothermal stress. To reconcile these competing demands, tailored functionalization strategies should emphasize chemical groups that can form stable covalent bonds with the matrix while minimizing hydrophilic moieties or by subsequently crosslinking the functional groups to reduce free polar sites accessible to water. For thermoplastic matrices, compatibilizers that bridge CNTs to the polymer while imparting hydrophobic character may be advantageous (Balasubramanian and Burghard, 2005).

Role of natural fibers: intrinsic hygroscopicity and design mitigation. Natural fibers are intrinsically hygroscopic; their hierarchical porosity and chemical composition result in moisture uptake that cannot be fully eliminated. While natural fibers offer sustainability and toughness, their moisture-driven swelling complicates interface design and long-term durability (Athijayamani et al., 2009; Sarkar et al.,

2010). Surface treatments (alkali treatment, silane coupling agents, acetylation) can reduce surface hydroxyl availability and improve fiber–matrix bonding, thereby mitigating swelling-induced debonding. Yet treatments must be optimized because over-treatment can weaken fiber tensile strength or inhibit mechanical interlocking. Another strategy is to introduce interfacial buffer layers or sizing agents that provide a mechanically compliant zone accommodating swelling without concentrating stress at the primary interface.

CNT dispersion and aggregation: the Achilles' heel under hygrothermal cycling. The positive benefits of CNT addition — increased storage modulus, reduced creep — are contingent on achieving uniform dispersion and strong interfacial adhesion (Jia et al., 2011; Yao et al., 2013). CNT aggregates not only reduce effective reinforcement efficiency but also create micro-scale voids and stress concentration sites that facilitate moisture accumulation and microcrack initiation under cyclic hygrothermal loading (Tehrani et al., 2011). Thus, processing routes that minimize aggregation—such as multi-step dispersion with surfactant removal or in situ polymerization around well-dispersed CNTs—are critical.

Creep and viscoelasticity: moisture as a time-scale modifier. Water molecules interact with polymer chains, increasing free volume and thus facilitating segmental mobility. As a result, characteristic relaxation times decrease, and time-temperature-moisture superposition must be considered in creep predictions. CNT reinforcement increases modulus and retards chain relaxation by physical obstruction and load transfer, but only up to the point where plasticization offsets reinforcement effects (Jia et al., 2011; Tehrani et al., 2011). For long-duration applications, modeling efforts must include moisture-dependent shift factors for master curve construction.

Multiscale modeling: integrating diffusion and mechanics. Traditional composite models that decouple transport and mechanical response fail to capture observed non-Fickian and hysteresis behaviors. Chemo-mechanical coupling — where moisture diffusion influences local mechanical properties and mechanical stress influences microstructural pathways for transport — is required. Representative Volume Element (RVE) models incorporating explicit fiber, matrix, and CNT phases, coupled with damage mechanics at interfaces,

offer promise for predictive simulation. However, capturing the broad distribution of natural fiber microstructures and the stochastic nature of CNT dispersion remains a computational challenge requiring stochastic homogenization or statistical multiscale approaches (Mulenga et al., 2021; Alhijazi et al., 2020).

Limitations of current literature and experimental protocols. A recurring limitation across studies is variability induced by non-standardized moisture conditioning and by heterogeneity of natural fibers and CNT preparations. For instance, immersion at elevated temperatures accelerates degradation but may not be representative of in-service conditions. Conversely, ambient humidity tests can underestimate long-term damage potential (Ferguson and Qu, 2006; Jen and Huang, 2013). Another limitation is the scarcity of long-duration creep data under realistic cyclic hygrothermal loading; short-term DMA and creep tests often fail to reveal slow damage accumulation. Moreover, many studies focus on one reinforcement type or isolated phenomena; comprehensive factorial studies across key variables (CNT functionalization, fiber treatment, matrix chemistry, and exposure severity) are less common.

Translational implications for engineering design. For design engineers, the practical implications are clear: materials design must be holistic. Simple substitution of CNTs into natural fiber composites without addressing dispersion, interfacial chemistry, and fiber treatments can yield materials that perform worse under hygrothermal stress than simpler alternatives. Conversely, judicious design — for instance, combining acetylated fibers, hydrophobically functionalized CNTs that are covalently bonded to a crosslinked epoxy, and controlled processing to minimize voids — can produce hybrids that retain mechanical properties and resist creep and damping deterioration in humid environments (Balasubramanian and Burghard, 2005; Athijayamani et al., 2009).

Future research directions. Several critical research directions arise from the synthesis and identified gaps:

- Systematic factorial experiments that vary CNT functionalization type and degree, CNT loading,

natural fiber species and surface treatment, and matrix chemistry, combined with standardized hydrothermal conditioning (including cyclic regimes), will clarify dominant interactions. Statistical designs should be used to identify interactions and thresholds beyond which durability rapidly degrades.

- Development of comprehensive chemo-mechanical constitutive models that include moisture-dependent viscoelasticity, swelling-induced stress, interfacial debonding kinetics, and damage accumulation algorithms. These models should be validated against physical experiments that include both short-term (DMA, microbond tests) and long-term (creep, cyclic hydrothermal fatigue) data sets.
- Advanced characterization of local moisture distributions using imaging techniques (e.g., micro-CT with contrast agents, neutron radiography) to visualize water pathways in hybrid microstructures, complemented by nanoscale chemical analyses (Raman, XPS) to detect hydrolytic changes.
- Lifecycle and environmental impact analyses juxtaposed with performance assessments to evaluate trade-offs between sustainability gains from natural fibers and the environmental costs of CNT production and potential complications during recycling or end-of-life handling.
- Exploration of novel interface chemistries that combine covalent bonding for load transfer with hydrophobic character to reduce local water accessibility — for instance, dual-functional silane coupling agents or polymeric interphases that covalently bond to both fibers and CNTs while containing hydrophobic segments.

Through these research avenues, the composite community can advance from isolated empirical observations toward robust, mechanistically informed design principles for durable, multifunctional hybrid composites.

Conclusion

This comprehensive theoretical study synthesizes evidence from molecular, microscale, and macroscale investigations to articulate how moisture and temperature jointly influence the mechanical performance of carbon nanotube–natural fiber hybrid composites. The key conclusions are:

- Chemical functionalization of CNTs is a double-edged sword: it improves interfacial load transfer but can increase local hydrophilicity and susceptibility to moisture-induced degradation. Optimal interfacing requires balancing chemical coupling with limited moisture affinity (Balasubramanian and Burghard, 2005).
- Natural fibers introduce intrinsic hygroscopicity and swelling that drive interfacial debonding and long-term reductions in stiffness and strength. Surface treatments and sizing strategies are essential to mitigate these effects (Athijayamani et al., 2009; Sarkar et al., 2010).
- CNTs can enhance stiffness and retard creep in dry or low-moisture conditions, but under substantial moisture ingress and elevated temperature, matrix plasticization and interfacial weakening can undermine CNT efficacy (Jia et al., 2011; Yao et al., 2013).
- Time-dependent and nonlinear phenomena — such as non-Fickian diffusion, moisture-dependent viscoelastic relaxation, and cumulative damage — necessitate multiscale modeling frameworks that couple diffusion with mechanical constitutive laws (Mulenga et al., 2021; Alhijazi et al., 2020).
- To translate laboratory promise into engineering practice, integrated research programs are required that combine carefully controlled materials processing, standardized hydrothermal conditioning, long-term mechanical testing, multiscale characterization, and coupled chemo-mechanical modeling.

By following the methodological blueprint and research agenda outlined here, investigators can develop hybrid composites with predictable, durable performance in humid and thermally variable environments. This will enable broader adoption of composites that combine the environmental virtues of natural fibers with the remarkable functional properties of carbon nanotubes.

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