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Lignocellulosic Reinforcements in Polymer and Cementitious Matrices: Comprehensive Analysis of Mechanical Performance, Surface Interactions, and Application Potential

Ravi K. Menon
Department of Materials Science, University of Glasgow, United Kingdom

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Abstract

Background: Natural fibres and lignocellulosic materials have re-emerged as promising reinforcements in polymeric and cementitious composites due to their renewable origin, low density, and favorable mechanical-to-weight ratios (Kadolph, 2016; Sanjay et al., 2016). However, variability in fibre morphology, interfacial bonding, moisture sensitivity, and processing constraints hinder wider industrial adoption (Kumar et al., 2019; Peças et al., 2018). This study synthesizes extant empirical and theoretical knowledge to present an integrative perspective on the dynamic mechanical performance, surface morphology effects, and application-specific design considerations for natural fibre composites.

Methods: A critical synthesis methodology was adopted, combining comparative literature analysis with mechanistic interpretation anchored in the physico-chemical characteristics of lignocellulosic fibres. The analysis emphasises fibre linear density and strength standards (ASTM D1577, ASTM D3822), microstructural modification techniques, dynamic viscoelastic responses, and case studies spanning cementitious composites and soft body armour development (ASTM D1577, Shlykov et al., 2022; Mawkhlieng & Majumdar, 2019).

Results: The assembled body of evidence demonstrates that natural fibres such as sisal, kenaf, ramie, and recycled cellulose derivatives can effectively enhance stiffness and energy-absorption characteristics in both polymeric and cementitious matrices, provided that fibre treatment and surface engineering are optimised (Bahja et al., 2021; Abbas et al., 2022; Kusmono et al., 2022). Dynamic performance exhibits strong dependence on fibre fraction, orientation, and interfacial damping properties; hybridization strategies and nanoscale cellulose inclusion improve impact resistance and dispersion (Shlykov et al., 2022; Mahesh et al., 2021).

Conclusions: Natural fibre composites present a viable pathway toward sustainable, high-performance materials across diverse industrial sectors. Overcoming challenges—standardisation of testing, moisture mitigation, and scale-up of fibre processing—requires concerted multidisciplinary research and industrial partnerships. The article concludes with a detailed research agenda focused on interfacial science, long-term durability studies, and application-driven design frameworks.

Keywords: natural fibre composites, lignocellulosic reinforcement, interfacial engineering, dynamic mechanical performance, cementitious composites, impact resistance

Introduction

The resurgence of natural fibres as structural reinforcements is a response to growing environmental imperatives and a recognition of the unique material properties offered by lignocellulosic feedstocks. Natural fibres—comprising cellulose, hemicellulose, lignin, and minor extractives—present a complex hierarchical architecture from the nano- to the macro-scale that governs mechanical response (Kadolph, 2016; Britannica, 2023). Historically, synthetic fibres and mineral fillers dominated composite reinforcement due to predictable properties and mature supply chains. However, concerns over lifecycle impacts, recyclability, and cost volatility have rekindled interest in renewable alternatives (Kozlowski et al., 2004; Kumar et al., 2019).

The literature reveals a dichotomy: a rich array of small-scale studies demonstrating promising mechanical outcomes (Peças et al., 2018; Sanjay et al., 2016) versus persistent obstacles inhibiting industrial deployment (Kumar et al., 2019). Primary obstacles include intrinsic variability in fibre properties linked to plant genotype, growth conditions, and extraction methods (Kadolph, 2016), moisture-induced property shifts due to hydrophilic cellulose and hemicellulose (Kozlowski et al., 2004), and challenges in achieving robust interfacial bonding with hydrophobic polymer matrices or alkaline cementitious environments (Pai & Jagtap, 2015).

This article addresses these concerns through an integrated, in-depth analysis that draws on the provided corpus of studies while elaborating theoretical mechanisms and practical strategies. The objective is to produce a publication-ready treatise that not only collates empirical observations but also probes the underlying mechanics—micromechanics, surface chemistry, and dynamic viscoelasticity—to inform future experimental design and industrial application.

Background and Problem Statement

Lignocellulosic fibres possess inherently anisotropic tensile properties. Single-fibre strength, linear density, and cross-sectional variability are quantifiable via standardised methods such as ASTM D1577 and ASTM D3822; however, translating single-fibre metrics to composite-scale performance is non-trivial due to complex fibre–matrix interactions (ASTM D1577, 1979; ASTM D3822, n.d.; Kadolph, 2016). The literature identifies three interlinked problem domains that complicate technology transfer: (1) material variability and characterization, (2) interfacial adhesion and moisture management, and (3) performance under dynamic loads and impact—critical for safety and transportation applications (Sanjay et al., 2016; Mahesh et al., 2021; Mawkhlieng & Majumdar, 2019).

Material variability arises from natural heterogeneity in cellulose microfibril angle, lumen presence, and microfibrillar composition. These microstructural features dictate stiffness and failure modes under tensile and flexural loading (Kadolph, 2016; Pai & Jagtap, 2015). Interfacial adhesion governs stress transfer and energy dissipation during loading; poor adhesion leads to fibre pull-out and premature composite failure (Peças et al., 2018; Kumar et al., 2019). Moisture ingress, facilitated by hydroxyl groups in cellulose, alters fibre dimensions and plasticises the matrix near the interface, degrading long-term performance (Kozlowski et al., 2004).

Dynamic and impact performance is particularly challenging. Natural fibres exhibit viscoelastic behavior influenced by the polymeric nature of cellulose and lignin, resulting in frequency- and temperature-dependent damping. For applications such as soft body armour or impact-resistant structural components, understanding how fibre fraction, orientation, and matrix compatibility influence ballistic and high-strain-rate response is essential (Shlykov et al., 2022; Mahesh et al., 2021; Anggoro & Kristiana, 2015).

Given these interdependent challenges, the literature reflects a pressing need for a coherent theoretical and practical framework that can guide experimentalists and industry practitioners in optimising natural fibre composites for target applications. This article fills that gap by synthesising mechanistic insights with applied case studies and proposing a systematic research agenda that aligns materials science fundamentals with industrial constraints.

Literature Gap

While numerous studies document property improvements using treated fibres or hybrid reinforcements, there exists limited synthesis that unites surface chemistry, micromechanics, and dynamic performance into a coherent design process. Studies often report promising mechanical enhancements but stop short of connecting those outcomes to scalable manufacturing considerations and long-term durability metrics (Peças et al., 2018; Abbas et al., 2022). Moreover, standardisation of test methods and reporting remains incomplete—making cross-study comparisons difficult and slowing technology maturation (ASTM D1577, 1979; ASTM D3822, n.d.). There is a distinct gap in knowledge about the translation of laboratory-scale treatments (e.g., chemical modifications, nanocellulose inclusion) into industrially viable processes that preserve or enhance dynamic energy absorption and environmental resilience

(Kusmono et al., 2022; Shlykov et al., 2022). This work seeks to synthesise and extend current understanding by offering an integrated theoretical framework and identifying prioritized research directions to bridge laboratory promise and industrial reality.

Methodology

This article employs a critical integrative literature synthesis, complemented by mechanistic extrapolation grounded in fundamental materials science. The methodology emphasises textual analysis rather than new experimental data to ensure adherence to the given references while enabling deep theoretical elaboration.

Literature Selection and Scope: The corpus comprises classical textile and fibre handbooks, encyclopedic overviews of natural fibres, peer-reviewed studies on dynamic performance and surface morphology, reviews of industrial applications, material-specific studies (e.g., kenaf, sisal, ramie), ASTM standards for fibre characterization, and targeted application reports such as soft body armour case studies (Kadolph, 2016; Britannica, 2023; Shlykov et al., 2022; Sanjay et al., 2016; Mawkhlieng & Majumdar, 2019; ASTM D1577, 1979).

Analytical Framework: The synthesis follows a multi-scale approach:

1. **Constituent Level (Nano–Micro):** Analysis of cellulose nanostructure, microfibril angle, lignin distribution, and porosity. Citations connect structural features to mechanical metrics using established fibre science principles (Kadolph, 2016; Kozlowski et al., 2004).
2. **Fibre Level (Micro–Macro):** Discussion of single-fibre metrics (linear density, tensile strength) and their measurement (ASTM D1577; ASTM D3822), including implications for composite design (Pai & Jagtap, 2015).
3. **Interface Level:** Examination of surface treatments, chemical coupling strategies, and interfacial failure mechanisms. Emphasis on alkaline, silane, acetylation, and enzymatic approaches reported across the literature and their theoretical bases (Peças et al., 2018; Bahja et al., 2021).
4. **Composite Scale:** Mechanistic models of stress transfer, crack deflection, and energy absorption, informed by viscoelastic composite theory and empirical studies of dynamic response (Shlykov et al., 2022; Mahesh et al., 2021).
5. **Application Mapping:** Translation of material performance to specific industrial use-cases: cementitious reinforcements, protective armour systems, and structural components impacted by dynamic loads (Abbas et al., 2022; Anggoro & Kristiana, 2015; Mawkhlieng & Majumdar, 2019).

Quality Assurance and Citation Strategy: All major claims are accompanied by citations to the provided literature. Where inferential connections are made—such as extrapolating microstructural influences on macroscopic damping behavior—the argument is explicitly framed as theoretical interpretation built upon cited empirical observations (Shlykov et al., 2022; Kozlowski et al., 2004).

Narrative and Elaboration: The methodology intentionally favours detailed, discursive explanation over summarisation to meet the elaboration requirement. Each conceptual point is expanded upon with theoretical rationale, counterarguments, and implications for experimental design and industrial scalability.

Results

The synthesis produced several major findings across scales, which are described in detail below. These findings consolidate empirical observations from the reference corpus and elucidate mechanistic pathways that underpin observed behaviour in natural-fibre-reinforced composites.

1. **Constituent and Microstructural Determinants of Mechanical Behavior**

   Natural fibres derive mechanical properties from a hierarchical organisation: crystalline cellulose microfibrils embedded in an amorphous hemicellulose–lignin matrix create a composite-like cell wall architecture. The microfibril angle—defined as the angle between cellulose microfibrils and the fibre axis—is a decisive parameter controlling stiffness and tensile behaviour; lower microfibril angles yield greater axial stiffness and tensile strength (Kadolph, 2016). Lumen size, which contributes to porosity and density, affects linear density measurements (ASTM D1577) and composite packing efficiency (ASTM D1577, 1979). The presence of extractives and hemicellulose increases hydrophilicity, influencing moisture uptake and swelling under humid conditions (Kozlowski et al., 2004). These constituent-level traits explain observed variability in single-fibre strength and the sensitivity of composite properties to processing conditions (Pai & Jagtap, 2015).

2. **Single-Fibre Characterisation and Its Translation to Composite Performance**

   Standardised tests for linear density (ASTM D1577) and tensile strength (ASTM D3822) provide indispensable baseline metrics. However, the literature emphasises that single-fibre strengths often overestimate composite reinforcement potential due to scale effects, fibre–fibre interactions, and stress concentration in matrices (Kadolph, 2016; Pai & Jagtap, 2015). For example, high single-fibre tensile strength measured under idealised gripping and low strain-rate conditions may not translate to composite-level performance under complex loading modes, particularly impact or high-strain-rate events where matrix cracking and fibre pull-out dominate failure (Mahesh et al., 2021).

3. **Interfacial Bonding: Surface Morphology, Treatments, and Adhesion Mechanisms**

   Interfacial bonding remains the pivotal lever for improving property transfer. Surface morphology—characterised by fibrillation, surface roughness, and the presence of waxy cuticles—determines mechanical anchorage potential and chemical reactivity (Pai & Jagtap, 2015). Chemical treatments such as alkali (mercerisation), silanization, acetylation, and enzymatic processing modify surface energy, expose reactive hydroxyl groups, and reduce hemicellulose content to improve compatibility with hydrophobic polymer matrices or to mitigate adverse reactions in alkaline cementitious systems (Peças et al., 2018; Bahja et al., 2021). The literature documents improved tensile strength, reduced water uptake, and better dispersion following such treatments, but emphasises trade-offs: aggressive treatments can damage cellulose crystallinity and reduce intrinsic fibre strength (Peças et al., 2018; Kusmono et al., 2022).

4. **Dynamic and Viscoelastic Composite Performance**

   Natural fibre composites exhibit viscoelastic response and frequency-dependent damping due to the polymeric nature of cellulose and the viscous contributions of hemicellulose and lignin. Shlykov et al. (2022) report that varying fibre proportions strongly affects dynamic performance; increasing fibre fraction can both stiffen the composite and reduce damping per unit volume, but microstructural heterogeneity introduces complex internal frictional mechanisms that can enhance energy dissipation under certain configurations (Shlykov et al., 2022). The inclusion of nanocrystalline cellulose (NCC) and well-dispersed nanoscale reinforcements can augment interfacial bonding and improve impact resistance by bridging microcracks and providing additional energy dissipation pathways (Kusmono et al., 2022; Mahesh et al., 2021).

5. **Cementitious Composites Reinforced with Natural Fibres**

   The incorporation of natural fibres into cementitious matrices offers improved tensile toughness, crack arresting, and post-crack flexural performance, particularly at low fibre fractions where the risk of fibre-matrix chemical degradation is limited (Abbas et al., 2022; Bahja et al., 2021). However, alkaline degradation of cellulose and hemicellulose remains a significant concern in portland cement environments; strategies include fibre pre-treatment, surface coating, and matrix modification with supplementary cementitious materials to reduce alkalinity and extend durability (Kozlowski et al., 2004; Abbas et al., 2022). Morphological studies (Bahja et al., 2021) show that treated sisal fibres exhibit better dispersion and improve crack-bridging mechanisms, translating to higher energy absorption before failure.

6. **Applications in Protective Systems and Impact-Resistant Structures**

   Natural fibres have been experimentally demonstrated in soft body armour and ballistic applications when hybridised with high-performance fibres or when used as secondary layers to absorb and dissipate energy via delamination, fibre pull-out, and frictional mechanisms (Mawkhlieng & Majumdar, 2019; Anggoro & Kristiana, 2015). Anggoro and Kristiana (2015) combine ramie fibres with epoxy matrices for body armour prototypes, suggesting viability when fibre architecture and matrix ductility are optimised. Nevertheless, the literature cautions that natural fibres alone rarely replace advanced synthetic fibres in high-threat ballistic scenarios without significant composite design optimisation and hybridisation (Mawkhlieng & Majumdar, 2019).

7. **Processing and Scale-Up Considerations** Translation to industry-scale production is impeded by supply chain variability, limited standardisation in processing protocols, and the need for environmentally benign, cost-effective fibre treatments (Kumar et al., 2019; Peças et al., 2018). Mechanically based fibre extraction and chemical retting processes present trade-offs between fibre quality and environmental footprint. The literature underscores the importance of integrating agronomic practices, standardised extraction, and quality control to reduce property scatter and enable predictable composite performance (Kadolph, 2016; Kumar et al., 2019).

Discussion

The preceding results highlight several intertwined themes that warrant deep interpretive analysis: the primacy of interfacial engineering, the role of microstructural control in dynamic performance, the nuanced balance between treatment efficacy and fibre integrity, and the pragmatic constraints of scaling natural fibre technologies. This discussion examines these themes, interrogates assumptions, identifies limitations, and articulates a strategic research and development roadmap.

1. **Interfacial Engineering as the Central Design Variable**

The evidence positions the fibre–matrix interface as the fulcrum upon which composite performance pivots. Effective stress transfer during static and dynamic loading requires adhesion mechanisms that balance chemical bonding with mechanical interlocking. Alkali treatments remove surface impurities and increase roughness, improving mechanical interlock, but can depolymerise hemicellulose and reduce tensile strength if not controlled (Peças et al., 2018). Silane coupling offers covalent bridging between fibre hydroxyls and polymer matrices, yet efficacy depends on synthesis parameters and application conditions (Peças et al., 2018). Acetylation reduces hydrophilicity and stabilises dimensional behaviour, improving durability in humid or cementitious systems, but it may alter fibre stiffness and modulate energy dissipation capacity (Bahja et al., 2021).

Design insight: interfacial strategies must be application-specific. For load-bearing structural composites where stiffness is critical, treatments that preserve cellulose crystallinity while improving adhesion should be prioritised. For energy-absorbing applications (impact, ballistic), treatments that preserve controlled interfacial debonding and fibre pull-out mechanisms may be advantageous, as these mechanisms dissipate kinetic energy through frictional work (Mahesh et al., 2021; Shlykov et al., 2022).

2. **Microstructure–Performance Linkages in Dynamic Regimes**

Dynamic response is sensitive to fibre architecture (orientation, length distribution, and volume fraction) as well as to intrinsic viscoelasticity of the constituents. The microfibril angle and cellulose crystallinity impart frequency-dependent stiffness and damping characteristics that manifest at composite scales (Kadolph, 2016; Shlykov et al., 2022). Intriguingly, a certain degree of heterogeneity—such as controlled fibrillation or deliberately roughened surfaces—may enhance energy dissipation by introducing additional micro-mechanisms for frictional loss (Shlykov et al., 2022). Counter-argument and nuance: while heterogeneity can increase damping, it can also introduce stress concentrators that reduce strength and accelerate fatigue failure. Thus, an optimal design window exists where energy dissipation is maximised without catastrophic loss of structural integrity. Quantifying this window requires systematic parametric studies linking microstructural metrics (microfibril angle distributions, lumen fraction) to composite-level viscoelastic functions (storage and loss modulus across frequency and temperature), an area underdeveloped in current literature (Shlykov et al., 2022).

3. **Treatment Trade-offs: Strength Versus Durability Versus Environmental Impact**

   Many effective chemical treatments utilise caustic or organic solvent chemistries that raise environmental and cost concerns. The literature includes promising explorations such as enzymatic retting and milder chemistries that offer selective removal of non-cellulosic components (Peças et al., 2018; Kusmono et al., 2022). However, enzymatic processes are often slower and require precise control. Implication for industrial adoption: life-cycle analyses and cost-benefit assessments are necessary to choose appropriate treatments at scale. Treatments that improve durability and reduce maintenance costs over a structure's life may justify higher upfront processing costs—particularly for infrastructure or safety-critical applications. Conversely, low-cost, minimal-treatment approaches may be appropriate for non-critical load-bearing applications where recyclability and end-of-life considerations predominate (Kumar et al., 2019).

4. **Cementitious Matrices: Chemical Compatibility and Long-Term Performance**

   The alkaline environment of Portland cement can hydrolyse cellulose and hemicellulose chains, weakening fibres over time (Kozlowski et al., 2004). Ash admixtures and blended cements reduce free calcium hydroxide concentration, offering a pathway to mitigate degradation. Fibre coatings (e.g., polymeric encapsulation) can physically separate fibres from alkaline pore solutions but may impede mechanical bonding unless engineered for controlled adhesion (Abbas et al., 2022). Studies of treated sisal in cementitious matrices indicate improved morphological integrity and mechanical performance when treatment and mix design are co-optimised (Bahja et al., 2021).

   Research need: long-term durability studies under wet–dry and freeze–thaw cycles, combined with microstructural evolution analyses, are crucial to establish service-life predictions. The literature provides snapshots of short-term improvements but lacks multi-year field performance datasets (Abbas et al., 2022).

5. **Protective Applications and Hybrid Composites**

   Natural fibres show promise as components of multi-layer protective systems. Their low density and capacity for controlled failure mechanisms can complement high-strength synthetic fabrics by attenuating residual energy and reducing back-face deformation (Mawkhlieng & Majumdar, 2019; Anggoro & Kristiana, 2015). Hybridisation strategies—layering natural fibres with aramids, UHMWPE, or ceramic backings—can leverage the strengths of each material class

   Caveat: ballistic threats are defined by specific energy and mass parameters. Validation requires standardised testing against certified protocols. Existing work demonstrates concept feasibility but is not definitive for operational deployment without comprehensive ballistic certification (Mawkhlieng & Majumdar, 2019).

6. **Standardisation and Quality Control**
   The absence of universal standards for natural fibre composite testing complicates material qualification. ASTM standards for single-fibre metrics are valuable but insufficient; composite-level standards that address dynamic testing, ageing, and environmental conditioning must be developed to enable industry confidence (ASTM D1577, 1979; ASTM D3822, n.d.). Moreover, supply chain controls—from cultivar selection to retting and decortication—must be codified to reduce property scatter (Kadolph, 2016).

7. **Sustainability and Circularity Considerations**
   Beyond mechanical performance, natural fibre composites offer tangible environmental benefits: lower embodied energy, reduced carbon footprint, and potential for biodegradation or recyclability under certain conditions (Kozlowski et al., 2004; Kumar et al., 2019). Yet, the full lifecycle advantages depend on processing choices and end-of-life strategies. For instance, chemical treatments that impede biodegradation could undermine circularity unless balanced with reuse or safe disposal pathways (Peças et al., 2018).

Limitations

This synthesis is constrained by reliance on the provided literature, which, while broad, represents a curated subset of existing work. Empirical generalisations are tempered by variability in experimental protocols across studies. Where theoretical extrapolation is utilised—such as in linking microfibril angle distributions to composite damping spectra—these should be validated by targeted experimental investigations. Furthermore, while industrial considerations are discussed, economic analyses and large-scale supply chain modelling are outside the scope of this article and require dedicated studies.

Future Scope

To accelerate the transition of natural fibre composites from laboratory promise to industrial reality, the following research priorities are recommended:

1. **Standardised Composite Testing Protocols:** Develop composite-level standards for dynamic, ballistic, and durability testing that account for environmental conditioning. This will enable reliable cross-study comparison and certification.

2. **Multi-scale Characterisation and Modelling:** Integrate nano- and micro-scale measurements (e.g., SAXS, AFM, micro-CT) with meso-scale mechanical testing to build predictive models of composite behavior across frequencies and temperatures. Such models should incorporate fibre property distributions rather than nominal averages to account for natural variability.

3. **Green Processing Technologies:** Invest in enzymatic and low-impact chemical treatments for fibre surface modification that balance efficacy with environmental impact and cost-effectiveness.

4. **Hybridisation Strategies for High-Performance Applications:** Explore engineered hybrid laminates that place natural fibres in roles that exploit their energy dissipation potential while relying on synthetic fibres for primary load-bearing, enabling both performance and sustainability gains.

5. **Long-term Field Studies:** Implement multiyear monitoring of structural elements and protective systems incorporating natural fibres to gather data on ageing, moisture effects, and maintenance requirements.

6. **Supply Chain and Agronomic Integration:** Link agronomy, post-harvest processing, and materials engineering to create predictable, scalable fibre supply chains with documented quality metrics.

7. **Life-Cycle and End-of-Life Strategies:** Integrate life-cycle assessments with material development to ensure that processing choices preserve the environmental benefits of natural fibres and enable circularity where feasible.

Conclusion

Natural fibre composites present a compelling, multifaceted opportunity to couple performance with sustainability. The body of evidence synthesised here demonstrates that, when thoughtfully engineered, natural fibres can improve stiffness, toughness, and energy absorption in polymeric and cementitious matrices. Nevertheless, widespread industrial adoption hinges on resolving interfacial challenges, establishing standards, and developing environmentally responsible processing routes. The theoretical analyses presented in this article underscore the necessity of multi-scale, application-driven research that bridges material fundamentals with pragmatic industrial constraints. By prioritising interfacial science, robust characterisation, and scalable green processing, the field can unlock the full potential of lignocellulosic reinforcements for a broad suite of structural and protective applications.

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