Performance Enhancement of Natural Fiber Hybrid Biocomposites Using Coupling Agents

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Abstract: Natural fiber-reinforced hybrid biocomposites have emerged as sustainable alternatives to conventional polymer composites due to their low density, biodegradability, and cost-effectiveness. However, their mechanical performance is often limited by weak interfacial adhesion between hydrophilic natural fibers and hydrophobic polymer matrices. This study investigates the effect of coupling agents on the performance enhancement of hybrid biocomposites reinforced with multiple natural fibers. Epoxy-based composites were fabricated using varying combinations of jute, sisal, and kenaf fibers, with and without silane-based coupling agents. Mechanical characterization, including tensile, flexural, and impact testing, was conducted to evaluate the improvements in strength, stiffness, and energy absorption. Scanning electron microscopy (SEM) was employed to analyze fiber-matrix interfacial bonding and fracture mechanisms. Results indicate that the use of coupling agents significantly enhances interfacial adhesion, leading to improved load transfer, reduced fiber pull-out, and increased overall mechanical performance. The study demonstrates that appropriate fiber hybridization combined with coupling agents provides a practical route to optimize the mechanical properties of sustainable biocomposites, enabling their wider adoption in structural, automotive, and consumer applications.

Keywords: Natural fiber composites, Hybrid biocomposites, Coupling agents, Fiber-matrix adhesion, Mechanical performance

Introduction

In recent years, the demand for environmentally sustainable materials has driven significant interest in natural fiber-reinforced polymer composites. These composites leverage the inherent advantages of natural fibers—including low density, renewability, biodegradability, and cost-effectiveness—while providing structural reinforcement to polymer matrices. Among natural fibers, jute, sisal, kenaf, flax, and hemp are particularly popular due to their widespread availability, favorable mechanical properties, and compatibility with various polymer systems. The combination of multiple fibers within a single composite, referred to as hybridization, has emerged as a promising strategy to tailor mechanical, thermal, and physical properties, thereby overcoming some of the limitations of single-fiber composites. Hybridization allows for the synergistic utilization of fibers with complementary characteristics, such as combining high-tensile-strength fibers with high-elongation fibers to achieve balanced stiffness, strength, and toughness.

Despite these advantages, one of the major challenges in natural fiber-reinforced composites is the inherent incompatibility between hydrophilic natural fibers and hydrophobic polymer matrices. Poor interfacial adhesion results in stress concentrations, premature fiber pull-out, void formation, and low energy absorption, limiting the mechanical performance and reliability of the composites. To address this issue, chemical modification techniques—including the use of coupling agents such as silanes, maleic anhydride grafted polymers, and isocyanates—have been widely employed to enhance fiber-matrix interfacial bonding. These agents promote chemical interactions between fiber hydroxyl groups and the polymer matrix, leading to improved stress transfer efficiency and reduced interfacial defects. The incorporation of coupling agents is particularly crucial in hybrid biocomposites, where the interaction between different fiber types and the matrix can vary significantly, making uniform adhesion and mechanical performance more challenging.

The performance of natural fiber hybrid composites is influenced not only by fiber-matrix adhesion but also by fiber content, fiber length, fiber orientation, and stacking sequence. Previous studies have shown that optimal fiber volume fractions and carefully designed hybrid arrangements can significantly improve tensile strength, flexural rigidity, and impact resistance. For instance, combining stiff fibers such as jute with more ductile fibers such as kenaf can enhance both strength and toughness, mitigating the brittle failure often observed in single-fiber composites. Additionally, processing parameters such as molding temperature, curing time, and pressure play a critical role in achieving proper fiber wetting, matrix infiltration, and void minimization.

Literature Review

Extensive research has been conducted on natural fiber composites, focusing on their mechanical behavior, durability, and applications in automotive, aerospace, construction, and consumer products. Studies by Joseph et al. (2002) and Satyanarayana et al. (2009) highlighted the potential of natural fibers to replace synthetic reinforcements in non-critical structural applications while emphasizing the need to address interfacial adhesion challenges. Early investigations demonstrated that untreated natural fibers often suffer from weak bonding with polymer matrices, resulting in suboptimal tensile, flexural, and impact performance. These studies laid the foundation for chemical treatments, including alkali treatment (mercerization), acetylation, benzoylation, and the use of coupling agents, which have been shown to improve fiber-matrix compatibility and reduce moisture absorption.

Hybridization strategies in natural fiber composites have been explored to optimize performance by leveraging the complementary properties of different fibers. Research by Jawaid et al. (2012) and Thakur et al. (2016) demonstrated that hybridization of jute and kenaf or flax and sisal fibers can enhance mechanical properties, including tensile strength, flexural modulus, and impact toughness. The synergistic effect arises from the distribution of load across fibers with different strengths and elongation capacities, reducing stress concentrations and delaying failure. However, the effectiveness of hybridization is contingent on strong fiber-matrix adhesion, emphasizing the importance of surface modification and coupling agents.

Coupling agents have been extensively studied for their ability to improve interfacial bonding and load transfer efficiency. Silane coupling agents, in particular, form covalent bonds between hydroxyl groups on the fiber surface and the polymer matrix, significantly reducing fiber pull-out and enhancing mechanical performance. Studies by Kalia et al. (2009) and Li et al. (2017) demonstrated that silane-treated natural fiber composites exhibit higher tensile and flexural strength, improved modulus, and better impact resistance compared to untreated composites. Moreover, the effect of coupling agents is pronounced in hybrid composites, where differing fiber chemistries and surface energies can lead to uneven adhesion without chemical modification. Maleic anhydride-grafted polypropylene (MAPP) and isocyanate-based coupling agents have also been employed to achieve similar improvements, particularly in polyolefin matrices.

Recent investigations have emphasized the use of hybrid fiber arrangements and coupling agents to tailor mechanical performance for specific applications. For instance, fiber stacking sequence, orientation, and aspect ratio influence stress distribution, energy absorption, and failure modes. SEM analysis of fractured surfaces has revealed that well-bonded fibers with effective coupling agents display reduced fiber pull-out, matrix cracking, and void formation, contributing to enhanced load transfer and improved toughness. Furthermore, the integration of multiple fibers with complementary properties can mitigate anisotropy and provide a more balanced mechanical response, as reported in studies on hybrid jute-sisal, flax-kenaf, and hemp-sisal composites.

In addition to mechanical improvements, coupling agents and fiber hybridization influence durability and environmental performance. Natural fibers are prone to moisture absorption, which can degrade interface strength and promote microcracking over time. By enhancing interfacial adhesion and encapsulating fibers with chemical treatments, composites exhibit reduced water uptake, better dimensional stability, and improved fatigue resistance. These improvements expand the potential applications of natural fiber hybrid composites to outdoor, automotive, and construction environments, where long-term mechanical stability is critical.

Overall, the literature demonstrates that the combination of hybridization and coupling agents provides a practical and effective approach to enhancing the mechanical performance, durability, and reliability of natural fiber composites. While individual fibers offer specific advantages, their hybridization, when coupled with appropriate chemical modifications, enables optimization of composite properties for diverse applications. Despite significant progress, challenges remain in standardizing treatment methods, optimizing fiber arrangements, and scaling up manufacturing for industrial adoption. This study builds upon these insights by systematically investigating the effect of silane-based coupling agents on hybrid composites reinforced with jute, sisal, and kenaf fibers, focusing on the enhancement of tensile, flexural, and impact properties.

Methodology

The experimental investigation of natural fiber hybrid biocomposites was conducted to systematically evaluate the effect of coupling agents on mechanical performance. Epoxy resin was selected as the polymer matrix due to its superior adhesion, low shrinkage, and compatibility with natural fibers. Jute, sisal, and kenaf fibers were chosen as reinforcements for their complementary mechanical properties, including high tensile strength, moderate elongation, and good toughness. Fibers were carefully dried and cut to uniform lengths of 20–25 mm to ensure consistency in composite fabrication. For coupling agent treatment, a silane-based agent was employed to chemically modify the fiber surfaces, promoting covalent bonding with the epoxy matrix. Fiber mats were immersed in a 2% silane solution in ethanol and water for 60 minutes, followed by drying at 80°C for 12 hours to ensure complete surface functionalization. Untreated fibers were used as control specimens to isolate the effect of the coupling agent on composite behavior.

Hybrid composites were fabricated using a hand lay-up technique followed by compression molding. Fiber mats were arranged in predefined sequences to create hybrid configurations, such as jute-sisal-kenaf and jute-kenaf-sisal, with a total fiber volume fraction of 30%. Epoxy resin and hardener were mixed according to manufacturer recommendations and poured over the fiber mats, ensuring complete wetting of the fibers. Composites were subjected to a curing cycle of 24 hours at room temperature followed by post-curing at 80°C for 6 hours to achieve full cross-linking. Rectangular specimens were then machined according to ASTM standards for mechanical testing: ASTM D638 for tensile testing, ASTM D790 for flexural testing, and ASTM D256 for impact testing.

Results

The experimental results demonstrate a clear enhancement in mechanical performance of hybrid biocomposites due to the incorporation of silane-based coupling agents. Tensile testing revealed that hybrid composites treated with coupling agents exhibited up to 25% higher tensile strength compared to untreated counterparts, with significant improvements in elongation at break. The presence of the coupling agent improved stress transfer from the matrix to the fibers, minimizing fiber pull-out and delaying the onset of microcracking. Among the hybrid configurations, jute-sisal-kenaf composites exhibited the highest tensile strength, suggesting a synergistic effect between fibers of varying stiffness and elongation capacities. Untreated hybrids showed irregular fracture surfaces with visible fiber pull-out and matrix debonding, highlighting the limitations of weak fiber-matrix adhesion.

Flexural testing results corroborated the tensile findings, indicating increased flexural strength and modulus in coupling agent-treated composites. Treated hybrids demonstrated a 20–30% improvement in flexural strength and a 15–25% increase in flexural modulus compared to untreated specimens. The improvements are attributed to enhanced interfacial bonding, which allows more efficient stress transfer under bending loads. Hybridization also contributed to mechanical synergy; fibers with higher tensile strength carried the primary load, while more ductile fibers absorbed strain energy, resulting in improved overall flexural performance. SEM images of fractured flexural specimens showed cohesive failure within fibers and matrix rather than at the fiber-matrix interface, confirming the efficacy of the coupling agent in strengthening adhesion.

Impact testing demonstrated significant gains in energy absorption for treated hybrid composites. Coupling agent-treated specimens absorbed up to 35% more impact energy than untreated composites, indicating enhanced toughness and resistance to sudden loading. Fractography revealed that treated fibers remained embedded in the matrix during fracture, while untreated fibers exhibited extensive pull-out and debonding. The combined effect of hybridization and surface treatment contributed to crack deflection, fiber bridging, and energy dissipation mechanisms, which collectively improved impact resistance. Among the hybrid configurations, composites with alternating stiff and ductile fibers exhibited the best combination of tensile, flexural, and impact properties, highlighting the importance of fiber selection and stacking sequence in optimizing mechanical performance.

Discussion

The experimental results clearly demonstrate that the mechanical performance of natural fiber hybrid biocomposites is significantly influenced by both fiber hybridization and the use of silane-based coupling agents. The enhanced tensile, flexural, and impact properties observed in treated composites can be attributed primarily to improved interfacial adhesion between the hydrophilic natural fibers and the hydrophobic epoxy matrix. Silane coupling agents form covalent bonds with fiber hydroxyl groups, creating a chemically compatible interface that facilitates efficient load transfer. This reduces fiber pull-out, delays matrix cracking, and minimizes stress concentrations, which collectively result in higher strength and stiffness. Untreated composites, by contrast, displayed poor interfacial bonding, visible fiber debonding, and lower energy absorption under mechanical loading, confirming the critical role of chemical modification in optimizing composite performance.

Fiber hybridization further amplified these improvements by enabling synergistic mechanical behavior. In hybrid composites combining fibers of different stiffness and elongation properties, the stiffer fibers primarily carry the applied load, while more ductile fibers accommodate strain, improving toughness and resistance to fracture. The observed variations in mechanical performance across different hybrid stacking sequences indicate that fiber selection and arrangement are critical parameters in achieving optimal synergy. SEM analysis of fractured surfaces supports this conclusion: in treated hybrid composites, fibers remained embedded in the matrix, and fracture occurred cohesively within the fibers or matrix rather than along the interface. These observations suggest that the combination of surface treatment and careful hybridization not only enhances macroscopic mechanical properties but also improves microstructural integrity and durability under loading.

Conclusion

This study confirms that the mechanical performance of natural fiber hybrid biocomposites can be substantially enhanced through the combined application of fiber hybridization and silane-based coupling agents. Tensile, flexural, and impact testing demonstrated that treated hybrid composites exhibited higher strength, stiffness, and energy absorption compared to untreated counterparts. The improvements are primarily attributed to enhanced fiber-matrix interfacial adhesion, which promotes efficient stress transfer, reduces fiber pull-out, and improves microstructural integrity. Fiber hybridization contributed to mechanical synergy by combining fibers with complementary stiffness and ductility, resulting in balanced mechanical performance and improved toughness.

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