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Research paper

Valorisation of Coffee Husk Fiber into High Performance PLA Biocomposites for Sustainable Food Packaging Applications

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Abstract

Agro-waste valorisation offers a sustainable route for developing environmentally responsible materials, particularly for packaging applications. In this study, coffee husk fiber (CHF), a lignocellulosic by-product of coffee processing, was chemically treated and utilised as a reinforcing agent in polylactic acid (PLA) to develop high-performance biocomposites. The CHF underwent alkaline treatment followed by benzoylation to enhance fiber–matrix compatibility by reducing its hydrophilicity. Composites were fabricated using compression moulding with varying CHF loadings (2.5%, 5%, and 7.5% by weight). The influence of fiber treatment on the mechanical, morphological, and water absorption properties of the composites was systematically investigated. Results showed a notable improvement in impact strength and flexural stiffness, particularly at 5% of CHF content treated. Water absorption was significantly reduced in treated composites, indicating enhanced durability. SEM revealed improved fiber dispersion and bonding, while FTIR confirmed successful chemical modifications. The findings demonstrate the feasibility of upcycling coffee processing waste into biodegradable packaging materials, supporting circular economy and waste reduction initiatives.

Keywords*: Coffee husk fiber, Polylactic acid, Agro-waste valorization, Biodegradable composites, Sustainable packaging, Fiber surface modification*

1. Introduction

Increasing global environmental concerns have led to a growing interest in the development of green composites that are biodegradable and sustainable. Among the most widely consumed beverages globally, coffee ranks second only to petroleum in terms of global trade value, with cultivation spread across more than 50 countries. However, coffee processing generates substantial amounts of agricultural waste, with coffee husks comprising approximately 40–50% of the processed bean mass [1]. While currently utilised in biofuel production, fertilisers, and as dietary fiber, the high caffeine content in coffee husks renders them unsuitable for livestock feed [1].

Due to their wide availability and low cost, coffee husk fiber (CHF) is a promising filler material for polymer composites. Natural fiber-reinforced composites offer several advantages such as biodegradability, high specific strength, low density, and reduced environmental impact [2]. When combined with biodegradable polymers such as polylactic acid (PLA), these composites become more sustainable. PLA, produced via fermentation of starch-rich crops, is a leading biopolymer with excellent tensile properties and compostability [3]. However, its inherent brittleness and low impact resistance limit its practical applications [4].

A sustainable biocomposite is defined as a material with minimal or no environmental impact throughout its lifecycle. However, the incorporation of untreated natural fibers into polymers may lead to hydrophilic behaviour, water absorption, poor fiber–matrix adhesion, and mechanical deterioration [5]. To overcome these challenges, chemical modification of fibers is necessary to improve their compatibility with hydrophobic polymer matrices.

Among various chemical treatments, benzoylation is recognised as an effective method to enhance the hydrophobicity of natural fibers and improve fiber–matrix adhesion. This process involves the reaction of benzoyl chloride with hydroxyl groups in cellulose under alkaline conditions, forming ester linkages that improve interfacial bonding [5]. Benzoylation has been shown to reduce water absorption and enhance thermal stability and mechanical properties [6].

Several studies have investigated the incorporation of agricultural waste into polymer matrices to produce eco-friendly composites. For example, rice husks, wheat straw, and coffee husks have been successfully utilised as reinforcing agents [7,8]. Compared to other agro-waste fibers, CHF offers a unique combination of properties that make it particularly suitable for biocomposite applications. CHF possesses a relatively high lignin content (~23.7%), which contributes to improved thermal insulation and dimensional stability, making it

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advantageous for applications like food packaging that require structural integrity under varying conditions. Additionally, CHF contains a substantial proportion of cellulose (~24.5%) and hemicellulose (~29.7%), which contribute to its mechanical reinforcement potential. Unlike many other agricultural by-products, CHF is abundantly available as a by-product of coffee processing, especially in coffee-producing regions like South India, yet remains underutilised. Its lightweight, fibrous structure, coupled with its compatibility with surface treatments such as benzoylation, allows for enhanced interfacial adhesion with hydrophobic polymer matrices like PLA. These distinctive features set CHF apart from conventional agro-fibers and justify its selection in this study as a novel and sustainable reinforcement agent for biocomposite development [1].

Furthermore, recent work has shown that torrefied and chemically modified CHF significantly enhances the performance of PLA-based composites. Improvements in thermal stability and mechanical strength of PLA composites reinforced with torrefied CHF have been reported, highlighting the importance of combined thermal and chemical treatments [9].

This study focuses on the fabrication and characterisation of CHF-reinforced PLA composites with varying fiber loadings (2.5%, 5%, and 7.5%), processed using compression moulding. The work evaluates the effects of chemical pretreatment on CHF and assesses the mechanical properties, morphology, and water absorption characteristics of the composites. The findings contribute to the field of sustainable materials by demonstrating the viability of CHF/PLA composites in potential packaging applications.

2. Materials and Methods

2.1 Materials

PLA 4043D, a biodegradable thermoplastic derived from renewable resources, was obtained in pellet form from Natur Tech India Ltd., Chennai, India. Due to its biocompatibility, biodegradability, and mechanical performance, PLA is widely used in eco-friendly composite applications [3]. Coffee husks were locally sourced from farms in Erode, Tamil Nadu, and served as the natural fiber reinforcement because of their high cellulose, hemicellulose, and lignin content [1]. Chemicals used for fiber treatment, including sodium hydroxide (NaOH), benzoyl chloride, ethanol, and methanol, were procured from Star Chemicals and Harenba Instruments, Erode, and used without further purification.

2.2 Pretreatment of CHF

To improve interfacial adhesion between CHF and PLA, chemical pretreatment was performed. Initially, the CHF was sun-dried to reduce the moisture content to about 15%. The fibers were then treated with 10% NaOH solution at room temperature for one hour to remove surface impurities and to activate hydroxyl groups on the fiber surface. Alkaline treatment is known to disrupt hydrogen bonding in the fiber structure, leading to improved fiber roughness and enhanced bonding potential with polymer matrices [5].

The alkali-treated fibers were then washed repeatedly with distilled water until a neutral pH was achieved, followed by drying for 24 hours. Subsequently, the fibers underwent benzoylation, a chemical surface modification that enhances fiber hydrophobicity and compatibility with hydrophobic polymers. This involved immersing the fibers in a mixture of benzoyl chloride and NaOH (ratio 9:1) for 30 minutes at room temperature. Benzoylation replaces hydroxyl groups on the fiber surface with benzoyl groups, thus reducing hydrophilicity and improving mechanical properties of the resulting composites [5,6]. The entire process is illustrated in Scheme 1.

After the reaction, fibers were thoroughly rinsed with ethanol to remove residual benzoyl chloride and dried. The dried CHF was then ground and sieved to a uniform particle size of approximately 0.2 mm, which improves dispersion and mechanical homogeneity within the PLA matrix [9].

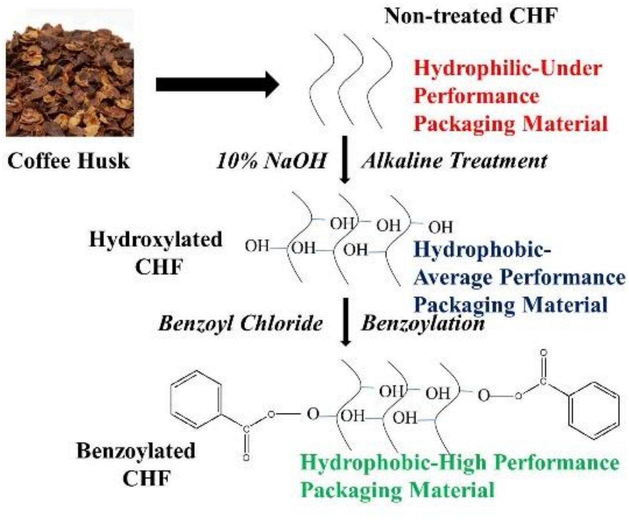

Fig. 1: Illustration for the preparation of different functionalized CHF.

2.3 Fabrication of CHF/PLA Composites

Composite sheets were prepared using the compression moulding method, known for producing void-free, well-consolidated composites with uniform fiber distribution [10]. Fiber loadings were varied at 2.5%, 5%, and 7.5% by weight. Each CHF/PLA blend was mixed thoroughly and moulded into a stainless steel die (20 cm × 20 cm × 3 mm) under a hydraulic press. The moulding was performed at 150°C and 5 MPa pressure for 3–4 hours to ensure complete consolidation and degassing [7].

Control samples using untreated CHF were also fabricated under identical conditions to compare the effect of surface treatment.

2.4 Characterisation of Composites

2.4.1 Scanning Electron Microscopy (SEM)

The fractured surfaces of the composites were examined using SEM to assess fiber–matrix adhesion and surface morphology. Before imaging, samples were coated with gold to ensure conductivity. SEM provided insights into dispersion uniformity and void presence in the composite microstructure [9].

2.4.2 Fourier Transform Infrared Spectroscopy (FTIR)

FTIR spectra of the composites were recorded using a Bruker spectrometer over the 4000–500 cm^{-1} range to identify functional groups and verify chemical bonding. Key peaks such as hydroxyl (~3400 cm^{-1}), carbonyl (~1750 cm^{-1}), and ester groups indicated the success of the benzoylation process and fiber–matrix interactions [16].

2.4.3 Impact Strength

Impact strength was measured using the Charpy method according to ASTM D256. Specimens (50 mm × 13 mm × 3 mm) were notched and tested using a pendulum impact tester. This test evaluates the toughness of the composite by determining the energy absorbed upon fracture [10].

2.4.4 Flexural Strength

Flexural strength was assessed by a three-point bending test in accordance with ASTM D790. Rectangular samples (60 mm × 13 mm × 3 mm) were tested using a universal testing machine with a span length of 50 mm and a crosshead speed of 2 mm/min. This method reflects the material's ability to resist deformation under bending loads [11].

2.4.5 Water Absorption

Water absorption behaviour was evaluated using ASTM D570. Dried specimens (65 mm × 65 mm × 5 mm) were weighed, submerged in distilled water, and reweighed at hourly intervals for 24 hours. Water absorption percentage was calculated to evaluate moisture uptake and the effectiveness of the benzoylation treatment in reducing fiber hydrophilicity [5,16].

3. Results and Discussion

3.1 SEM Morphological Analysis

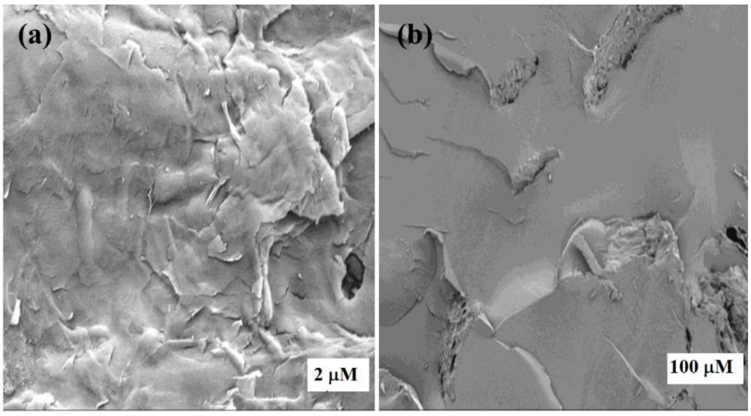

Fig. 2: SEM morphologies of CHF/PLA composites by using both (a) Treated CHF and (b) Non-treated CHF.

SEM micrographs revealed the morphological features of the fractured surfaces of the composites. Treated CHF/PLA composites displayed fewer voids and better fiber–matrix interlocking (Fig.1a), confirming the effectiveness of chemical treatment in enhancing adhesion. In contrast, untreated composites showed fiber pull-out and interfacial gaps (Fig. 1b), indicating weak bonding and poor load transfer.

Such interfacial discrepancies are known to deteriorate mechanical performance, particularly under flexural and impact loading. SEM observations corroborated the results from mechanical tests [6,9].

3.2 FTIR Structural Analysis

FTIR spectra of the 5% treated CHF/PLA composite (Fig.2) exhibited prominent absorption bands around 3400 cm^{-1} (–OH stretching), 1748 cm^{-1} (C=O stretching from ester bonds), and 1086 cm^{-1} (C–O–C stretching), confirming successful esterification during benzoylation [16]. These peaks correspond to the chemical interactions between the PLA matrix and the treated fiber surfaces, affirming effective chemical modification.

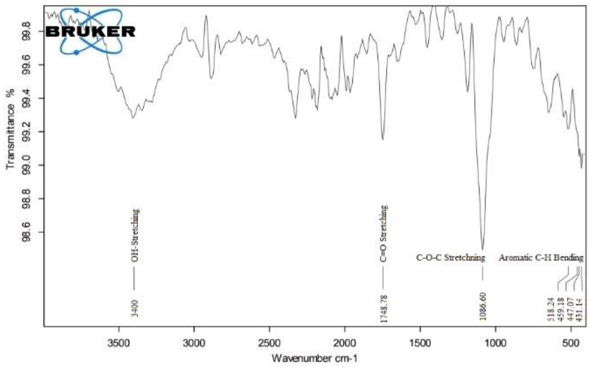

Fig. 3: FTIR spectra of the obtained CHF/PLA composite.

Additionally, bands between 2000–2500 cm^{-1} indicated unreacted aromatic compounds, a known limitation when benzoylation is not fully followed by ethanol/methanol washing [9]. Nonetheless, the reduction in hydroxyl-related peaks and the emergence of aromatic groups demonstrate that the CHF surface was effectively modified, improving compatibility with the PLA matrix [5].

3.3 Impact Strength

Impact strength is a critical mechanical property that reflects a material's ability to withstand sudden loads and resist fracture. Neat PLA is known for its brittleness and low impact resistance, which restricts its applications in structural components [3]. As shown in Fig. 3, the incorporation of chemically treated CHF significantly enhances the impact resistance of PLA composites. At 2.5% treated CHF loading, the impact strength increased to 768.9 J/m, while at 5% loading, it peaked at 942.0 J/m. However, at 7.5% loading, a slight decline to 665.8 J/m was observed, likely due to fiber agglomeration and matrix discontinuity [1,9].

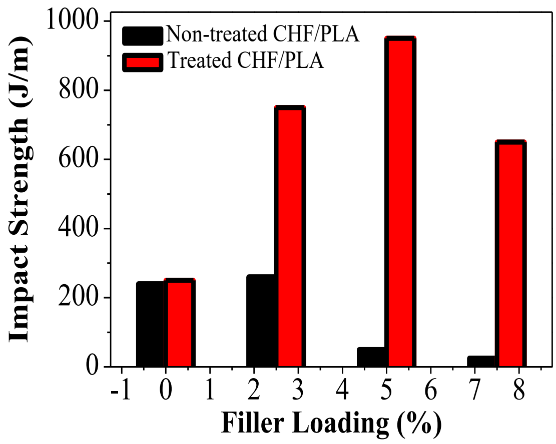

Fig. 4: Impact Strength of CHF/PLA composites at different loading levels of both treated and non-treated CHF.

This behaviour aligns with previously reported results, where chemically treated fibers improved the energy absorption capacity due to enhanced fiber–matrix interfacial bonding [5]. Benzoylation reduces fiber hydrophilicity and facilitates better dispersion, improving stress transfer from the matrix to the fiber under impact conditions [6,9].

In contrast, composites reinforced with untreated CHF exhibited significantly lower impact strength. This was attributed to poor interfacial bonding, leading to micro-crack formation and propagation under loading. Similar observations have been reported for other agro-fiber composites, demonstrating the necessity of chemical surface treatment [7].

3.4 Flexural Properties

Flexural strength and stiffness are essential parameters for evaluating a composite's resistance to deformation under bending loads—crucial for structural integrity in packaging applications. Fig. 4 illustrates the flexural strength of CHF/PLA composites at varying fiber loadings for both treated and untreated fibers. For treated CHF composites, the highest flexural strength (84.0 MPa) was observed at 2.5% fiber loading. This value declined with increasing filler content, dropping to 68.4 MPa at 5% and further to 57.9 MPa at 7.5%. This trend can be attributed to fiber agglomeration and increased interfacial voids at higher loadings, which act as stress concentration points and hinder effective stress transfer from the matrix to the fibers.

Conversely, composites containing untreated CHF exhibited significantly lower flexural strength across all loadings, primarily due to poor interfacial bonding and ineffective stress distribution under load. The presence of surface impurities and hydroxyl groups on untreated fibers contributes to weak adhesion with the hydrophobic PLA matrix, causing fiber pull-out and interfacial debonding under stress. These observations are consistent with previous studies on untreated agro-waste fillers in thermoplastic matrices, where insufficient bonding compromised the mechanical integrity of the composites [4, 6, 10].

In addition to strength, the flexural stiffness—a measure of material stiffness—increased with higher treated CHF content. This is due to the rigid nature of the lignocellulosic fibers, which restrict polymer chain mobility and enhance resistance to bending deformation. Despite the reduction in strength at higher filler content, the improved stiffness reflects the fibers' role as reinforcement agents contributing to the overall stiffness of the composite. Notably, the 5% treated CHF composite presented an optimal balance between stiffness and strength, supporting its selection as the best-performing formulation.

Benzoylation played a critical role in improving the flexural performance at lower loadings. The chemical modification enhanced fiber–matrix adhesion, promoting more efficient stress transfer and reducing the presence of interfacial voids. These improvements are visible in SEM micrographs, where treated fibers showed better dispersion and matrix embedding compared to untreated ones.

Overall, the flexural behaviour of the treated CHF/PLA composites confirms that proper surface treatment and filler content optimisation are essential for achieving desirable mechanical properties. Similar trends have been reported in studies involving mercerised, torrefied, or silane-treated fibers, highlighting the universal importance of interfacial engineering in biocomposite systems [6, 9, 11-15].

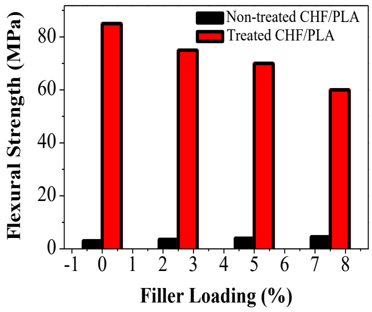

Fig. 4: Flexural Strength of CHF/PLA composites at different loading levels of both treated and non-treated CHF.

3.5 Water Absorption

Water absorption behaviour is critical for biocomposites used in packaging and outdoor applications. Fig. 5 illustrates the moisture uptake of both treated and untreated CHF/PLA composites. Treated fibers showed significantly reduced water absorption due to the benzoylation process, which lowers the number of hydroxyl groups available for hydrogen bonding with water molecules [5,16].

For instance, at 2.5% filler loading, the water absorption reduced from 2.63% (untreated) to 2.26% (treated), and at 7.5%, from 5.49% to 3.15%. This behaviour confirms the hydrophobisation effect of benzoyl chloride treatment [6]. Similar findings have shown that chemical surface modification effectively reduces the hydrophilicity of agro-waste fibers [1].

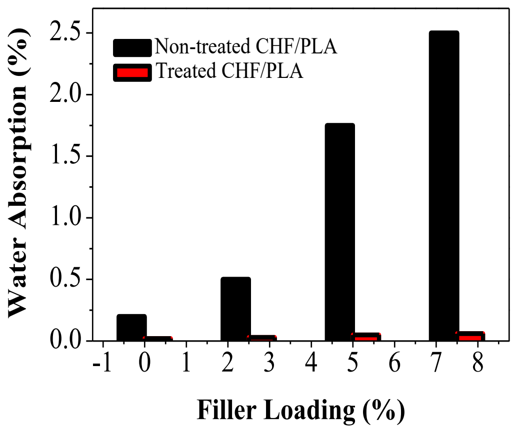

Fig. 5: Water Absorption of CHF/PLA composites at different loading levels of both treated and non-treated CHF.

The increase in water uptake with filler loading is attributed to increased interfacial voids and a higher surface area of the fibers exposed to water. It has also been highlighted that natural fiber content beyond an optimal level leads to moisture-driven degradation, compromising dimensional and mechanical stability [8].

3.6 Comparative study

Table 1: Performance Comparison of CHF/PLA Composite with Other Packaging Materials.

Material	Impact Strength (J/m)	Water Absorption (%)	Relative Cost	Biodegradability	Reference
CHF/PLA (5% Treated)	942.0	3.15	Low	Biodegradable (with PLA base)	This study
PLA/Starch Composite	~720.5	~5.10	Medium	Biodegradable	[4]
PET (Polyethylene Terephthalate)	~1200.0	~0.10	High	Non-biodegradable	[3]
Wheat Straw/PLA Composite	~860.0	~4.35	Medium	Biodegradable	[2]
Rice Husk/PP Composite	~890.0	~1.80	Medium	Partially biodegradable	[7]

Table 1 provides a comparative analysis of the performance characteristics of CHF/PLA composites against other commonly used packaging materials, including PLA/starch, wheat straw/PLA, rice husk/PP composites, and conventional PET. The 5% treated CHF/PLA composite developed in this study demonstrates an impressive impact strength of 942.0 J/m, which surpasses that of PLA/starch and

wheat straw/PLA composites. Moreover, its water absorption is significantly lower than other natural fiber-based composites, owing to the benzoylation treatment that enhances fiber hydrophobicity and interfacial adhesion.
While PET exhibits superior mechanical strength and minimal water absorption, its lack of biodegradability and high environmental footprint limit its appeal for sustainable applications. On the other hand, the CHF/PLA composite combines good mechanical performance, reduced moisture uptake, biodegradability, and low material cost, making it a viable candidate for eco-friendly food packaging. The inclusion of agro waste such as coffee husk fibers, not only contributes to waste valorisation but also enhances the functionality of the composite, supporting circular economy goals. This benchmarking validates the suitability of the proposed material in replacing or supplementing conventional packaging alternatives sustainably.

3.7 Future Directions

The current study establishes the feasibility of using chemically treated CHF as a reinforcing agent in PLA-based biocomposites for sustainable food packaging. However, several avenues remain open for future exploration. A key direction is to investigate the compostability and environmental degradation behaviour of these composites under both industrial and domestic conditions. Such studies will provide a clearer understanding of the materials' end-of-life behaviour and their contribution to waste minimisation.
Further research should also assess the UV resistance and thermal ageing of the composites to determine their durability in real-world storage or outdoor applications. While benzoylation has proven effective for fiber surface modification, its scalability and environmental footprint raise concerns. Hence, greener treatment alternatives, including enzymatic, silane, or plasma methods, should be explored to enhance the environmental compatibility and industrial applicability of the process.
Another promising direction involves enhancing the functional performance of CHF/PLA composites. The integration of barrier-enhancing or bioactive additives could improve moisture, oxygen, and microbial resistance—key properties for food-grade packaging materials. Additionally, to evaluate the commercial potential of these materials, it is imperative to conduct a techno-economic assessment alongside a life cycle analysis (LCA). These tools would help quantify their environmental benefits over conventional plastics and guide decisions in large-scale manufacturing.
Finally, with the rise of digital fabrication, future studies could adapt these composites for additive manufacturing technologies, such as fused deposition modelling (FDM). This would not only support the development of customisable, biodegradable packaging products but also extend the material's applications beyond packaging into broader sustainable design domains.

4. Conclusion

This research validates the potential of utilising coffee husk fiber, an abundant agro-industrial by-product, in the fabrication of PLA-based biocomposites for sustainable food packaging. Through alkaline and benzoylation treatments, the inherent hydrophilicity of CHF was reduced, resulting in improved fiber–matrix interaction, as confirmed by SEM and FTIR analyses.
Composites reinforced with 5% treated CHF demonstrated optimal performance, exhibiting a balance between mechanical strength, flexibility, and water resistance. The increase in impact strength and flexural stiffness, along with decreased water uptake, highlights the positive influence of fiber surface modification on composite performance. These enhancements are crucial for packaging applications that demand both strength and moisture tolerance.
By integrating agro waste into a biodegradable polymer matrix, this work contributes meaningfully to circular material cycles, offering an eco-friendly solution for managing agricultural residues and reducing reliance on conventional plastics. The study establishes a pathway for scalable, sustainable packaging innovations rooted in waste valorisation.

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