STM JOURNALS

Journal of
Polymer & Composites

https://journals.stmjournals.com/jopc

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

Research JoPC

Bio-Derived TPS–Lignin Hybrid Composites Reinforced with Modified Coir and Plantain Fibers for Eco-Efficient Structural Applications

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Abstract

This study develops bio-derived polymer composites using a thermoplastic starch–lignin (TPS–lignin) matrix reinforced with alkali-treated coir and plantain fibers for sustainable load-bearing applications. The TPS–lignin matrix was formulated using cassava starch, glycerol, and 10 wt% lignin, while coir and plantain fibers were surface-modified using 5 wt% NaOH to enhance interfacial compatibility. Composite laminates were fabricated with varying fiber loadings (10–40 wt%) through melt-mixing and hot-press compression. Tensile, thermal, morphological, and water absorption analyses were performed to determine the influence of single-fiber and hybrid-fiber reinforcement. Tensile stress–strain results revealed substantial improvement in mechanical performance, where hybrid composites exhibited the highest tensile strength, showing a ~55–70% increase over the neat TPS–lignin matrix and outperforming single-fiber composites. SEM micrographs confirmed improved fiber–matrix adhesion, reduced pull-out, and uniform fiber dispersion, validating the enhanced stress-transfer mechanism. TGA results showed enhanced thermal stability for reinforced composites, with the hybrid system retaining the highest residual mass and displaying the slowest rate of thermal degradation. Water absorption decreased significantly with fiber addition, where hybrid composites demonstrated over 40% lower water uptake compared to the matrix due to reduced voids, improved crystallinity, and the hydrophobic influence of lignin-rich coir fibers. Overall, the combined mechanical, thermal, and moisture-resistance enhancements confirm that TPS–lignin hybrid composites reinforced with treated natural fibers offer a promising biodegradable alternative for lightweight structural and eco-efficient engineering applications.

Keywords: Fiber reinforcement, hybrid composites, thermal degradation, TPS–lignin, water absorption

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Received Date: November 21, 2025
Accepted Date: December 01, 2025
Published Date: January 08, 2026

Citation: Nantha Kumar P, Nagasrisaihari Sunkara, Sachin Harak, A. Vijayalakshmi, DVSSSV Prasad, Karanam Suresh Babu, Kirubakaran D, S.Dhivya, Rajendiran M. Bio-Derived TPS–Lignin Hybrid Composites Reinforced with Modified Coir and Plantain Fibers for Eco-Efficient Structural Applications. Journal of Polymer & Composites. 2026; 14 (1): 136–145p.

INTRODUCTION

The global transition toward environmentally sustainable materials has significantly increased interest in bio-based polymers, natural-fiber composites, and polymer–composite technologies that reduce dependence on petroleum-derived plastics. Conventional synthetic polymers,

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although widely used for their mechanical strength and durability, pose substantial environmental concerns due to their non-biodegradable nature, high energy consumption during production, and end-of-life disposal issues [1–3]. In response, researchers are increasingly turning toward biodegradable polymers and bio-derived polymer-composites, which combine renewable polymer matrices with natural reinforcements to create eco-friendly materials suitable for structural and semi-structural applications.

Among the various biodegradable polymers, thermoplastic starch (TPS) is particularly attractive because of its low cost, renewability, and biodegradability. However, TPS alone is mechanically weak, thermally unstable, and highly sensitive to moisture, which limits its direct use in high-performance polymer-composite systems [4, 5]. To address these limitations, TPS is often blended with other biopolymers to form hybrid polymer matrices with improved performance. One such biopolymer is lignin, a naturally occurring aromatic polymer derived from plant biomass. Lignin is widely recognized for its rigidity, hydrophobic nature, and ability to enhance thermal and mechanical stability when incorporated into polymer systems. The integration of lignin into TPS matrices creates TPS–lignin polymer blends, offering a promising approach to designing high-strength polymer-composites with improved environmental resistance [6–8].

In recent years, natural fiber–reinforced polymer-composites have gained considerable attention in polymer engineering and green manufacturing. Natural fibers are renewable, biodegradable, and possess excellent specific strength, making them ideal reinforcements for polymer matrices. Among these, coir fibers have emerged as robust options due to their high lignin content, mechanical resilience, and resistance to microbial degradation [9]. Similarly, plantain fibers, obtained from agricultural residues, are lightweight, cellulose-rich, and compatible with a wide range of polymer matrices, making them valuable for polymer-composite reinforcement [10, 11]. Coir fibers contain ~40–45% lignin, contributing rigidity and hydrophobicity, whereas plantain fibers possess higher cellulose (~60–65%), offering flexibility and crack-bridging ability. These complementary characteristics motivate their selection for hybrid reinforcement. For polymer-composites to achieve optimum mechanical and thermal performance, strong interfacial bonding between fibers and the polymer matrix is essential. However, untreated lignocellulosic fibers contain waxes, pectin, and hemicellulose that reduce fiber–matrix compatibility. Hence, alkali treatment (e.g., NaOH treatment) is widely utilized in polymer-composite processing to remove amorphous components, increase fiber surface roughness, and enhance chemical interaction between fibers and polymer chains [12]. This modification improves stress transfer within polymer-composites and leads to better structural performance.

The field of polymer–composites increasingly emphasizes hybrid reinforcement strategies, where multiple natural fibers and biopolymers synergistically enhance the final composite properties. Combining TPS and lignin forms a robust bio-based polymer matrix, while introducing treated coir and plantain fibers enables improved tensile, flexural, and thermal properties. Such multi-component polymer-composite systems leverage the hardness and hydrophobicity of lignin, the toughness of coir fibers, and the flexibility of plantain fibers. This synergy creates high-performance, fully biodegradable polymer-composites capable of replacing low-load-bearing synthetic composites in automotive interiors, structural panels, furniture components, and sustainable packaging boards. While extensive research exists on individual TPS composites or lignin-based polymer blends, literature addressing TPS–lignin hybrid polymer-composites reinforced simultaneously with coir and plantain fibers remains scarce. Most previous studies focus on a single natural fiber or a single polymer component, leaving a significant research gap in understanding the behavior, compatibility, and performance of such multi-phase polymer systems [13–15]. As global demand increases for high-strength, biodegradable polymer-composites, developing novel hybrid materials that integrate multiple fibers and biopolymers becomes essential.

Therefore, the present study is motivated by the need to design a new class of bio-derived polymer-composites by incorporating alkali-treated coir and plantain fibers within a TPS–lignin polymer matrix. The goal is to develop an environmentally friendly polymer-composite material with superior mechanical strength, enhanced thermal stability, and reduced water absorption, thereby broadening the applicability of biodegradable polymer systems to structural and engineering domains. By exploring the synergistic effects of a lignin-modified polymer matrix and dual natural fiber reinforcement, this work contributes to the advancement of sustainable polymer-composites and supports the global movement toward greener materials.

MATERIALS & METHODS

Materials

The polymer matrix for the present polymer–composite system was formulated using commercially available cassava starch, glycerol, and kraft lignin, all of analytical grade. Cassava starch served as the primary biodegradable polymer, while glycerol (Merck, India) functioned as a plasticizer to convert native starch into thermoplastic starch (TPS). Kraft lignin, sourced from a local pulp-processing industry, was incorporated to enhance rigidity, hydrophobicity, and thermal stability of the resulting polymer matrix. Two natural fibers – coir fibers obtained from coconut husks and plantain pseudo-stem fibers collected from agricultural waste – were selected as reinforcements due to their availability, biodegradability, and mechanical suitability for polymer-composite fabrication. Both fibers were manually cleaned, sun-dried, and cut to lengths of 6–8 mm. For surface modification, high-purity sodium hydroxide (NaOH) pellets were procured to carry out alkali treatment, enabling improved fiber–matrix adhesion by removing hemicellulose, lignin residues, and surface waxes. All chemicals used in the study were of laboratory grade and employed without further purification.

Methods

Thermoplastic starch (TPS) was prepared (as shown in Fig.1) by melt-blending cassava starch with glycerol at a weight ratio of 70:30 using a controlled-temperature mechanical mixer at 75–80°C until a homogeneous polymer phase was obtained.

Figure 1. Preparation of TPS–lignin–fiber polymer composites.

(a) Gelatinization of TPS–lignin mixture on hot plate. (b) Alkali-treated coir and plantain fibers prepared for mixing. (c) Manual blending of treated fibers into molten polymer matrix (d) Hot-pressed composite sheet after compression molding.

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Lignin (10 wt%) was then incorporated into the molten TPS to form a hybrid polymer matrix with improved hydrophobic and mechanical properties. Meanwhile, coir, and plantain fibers were subjected to alkali treatment by soaking them in 5 wt% NaOH solution for 4 hours, followed by repeated washing with distilled water until neutral pH and oven-drying at 70°C for 24 hours. Composite fabrication involved premixing the treated fibers with the TPS–lignin blend at various fiber loadings (10–40 wt%) to create uniform polymer–composite formulations. The mixtures were subsequently compression-molded using a hot press at 150°C and 10 MPa for 12 minutes, followed by gradual cooling under pressure to prevent warpage and ensure dimensional stability. The resulting polymer-composite laminates were conditioned at ambient temperature for 48 hours before testing. All samples were later subjected to morphological, mechanical, thermal, and water absorption analyses using standard ASTM procedures to evaluate the performance of the developed polymer–composites.

CHARACTERIZATION

To comprehensively evaluate the structural integrity, interfacial morphology, thermal stability, and durability of the developed TPS–lignin–fiber polymer-composites, a combination of mechanical, microstructural, thermal, and moisture-resistance tests were conducted. These tests were selected to justify the composite's suitability for eco-efficient structural and semi-structural applications.

Tensile Testing

Tensile testing was carried out according to ASTM D638 to determine the load-bearing capacity, stiffness, and deformation behavior of the polymer-composites. The neat TPS–lignin matrix exhibited relatively low tensile strength due to its inherent brittleness and limited chain entanglement. Incorporation of alkali-treated coir and plantain fibers significantly enhanced tensile strength and modulus, indicating efficient stress transfer between fibers and the polymer matrix. Fiber loading improved the stiffness by restricting polymer chain mobility, while the increased cellulose crystallinity and surface roughness of treated fibers provided strong interfacial bonding. The hybrid composites (coir + plantain fibers) exhibited the highest tensile improvements, demonstrating synergistic reinforcement due to the complementary properties of the two fiber types. These results confirm that the composite can withstand mechanical loads typical of lightweight structural applications.

Microstructural Analysis

SEM analysis was conducted to examine fiber morphology, surface modification, and matrix–fiber interfacial adhesion. Untreated fibers exhibited smooth, waxy surfaces with impurities that hinder adhesion, while alkali-treated fibers showed increased surface roughness and fibrillation, indicating removal of hemicellulose and lignin residues. Fracture surface micrographs of composite samples revealed well-embedded fibers with reduced pullout behavior, demonstrating strong interfacial bonding. The presence of broken fibers instead of pulled-out fibers indicates effective load transfer during tensile loading. Hybrid composites showed more uniform fiber dispersion and fewer voids compared to single-fiber composites. The improved morphology validates the enhanced mechanical performance reported in tensile testing.

Thermal Analysis

TGA was performed under nitrogen atmosphere at a heating rate of 10°C/min to evaluate the thermal stability and degradation behavior of the composites. The TPS–lignin matrix showed an initial mass loss at 90–120°C due to moisture evaporation, followed by major decomposition in the 250–300°C range attributed to starch degradation. Fiber reinforcement increased the onset degradation temperature to 310–330°C, indicating substantial improvement in thermal resistance. The high lignin content and enhanced crystallinity of treated fibers contributed to slower mass loss and higher char residue. Hybrid composites exhibited the greatest thermal stability, confirming their suitability for structural applications where exposure to moderate thermal stress is expected.

Water Absorption Analysis

Water absorption was evaluated following ASTM D570 to determine the composite's resistance to moisture ingress, a critical factor affecting dimensional stability and long-term durability of polymer-composites. The TPS–lignin matrix showed considerable water uptake due to starch’s hydrophilic nature. However, lignin incorporation provided partial hydrophobicity. Alkali-treated fibers exhibited reduced hydrophilicity, and their incorporation significantly lowered the water absorption of the composite. Hybrid fiber composites demonstrated the least water uptake owing to higher lignin content, improved crystallinity, and reduced voids, which restricted water diffusion pathways. The reduced moisture uptake ensures dimensional stability in humid environments and extends composite service life.

RESULTS AND DISCUSSION

The performance of the TPS–lignin–fiber polymer-composites was evaluated using a series of mechanical, morphological, thermal, and moisture-resistance tests. The graphs and SEM micrographs provided illustrate the comparative behavior of the neat matrix, coir-fiber composites, plantain-fiber composites, and hybrid composites. The following discussion interprets the observed trends and highlights the synergistic reinforcement mechanisms responsible for the enhanced composite properties.

Tensile Behavior

The tensile stress–strain curves (Figure 2) demonstrate a clear improvement in mechanical properties upon incorporation of natural fibers into the TPS–lignin polymer matrix. The neat TPS–lignin matrix exhibits a typical brittle polymer behavior, characterized by a steep rise in stress followed by early fracture at low strain values. This is due to the limited ductility of starch-rich matrices and poor entanglement density of polymer chains [16].

The addition of coir fibers enhances the load-bearing capacity, attributed to the fiber’s high lignin content and inherent toughness. The coir-reinforced composite shows an increase in both tensile strength and modulus, indicating improved stiffness and resistance to deformation. Plantain-fiber composites also display enhanced tensile behavior, though slightly lower than coir-reinforced composites, which can be credited to plantain fiber’s relatively lower lignin content and greater flexibility.

The hybrid composite (coir + plantain fibers) outperforms both single-fiber composites. This synergistic effect arises from the complementary mechanical roles of the two fibers:

- Coir fibers contribute rigidity, dimensional stability, and compressive resistance.
- Plantain fibers contribute tensile compliance, crack-bridging ability, and energy absorption.

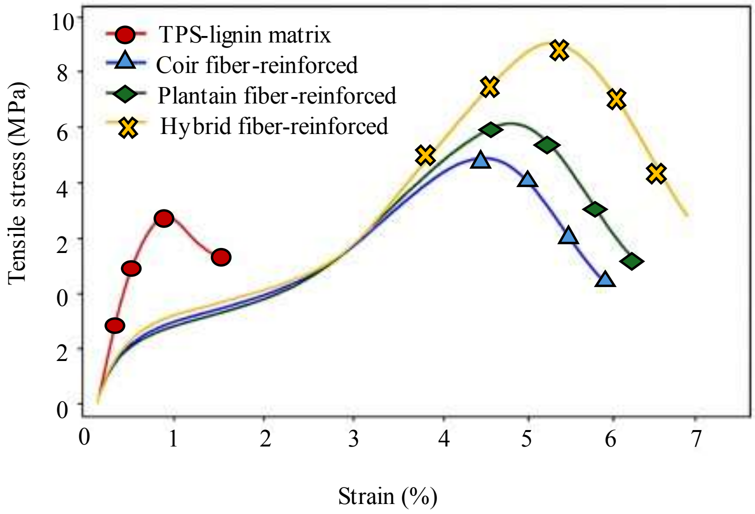

Figure 2. Tensile stress–strain curves of TPS–lignin and fiber-reinforced composites.

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The stress–strain response of the hybrid composite shows a smoother curve with higher ultimate stress and delayed fracture, confirming that hybrid reinforcement significantly improves toughness and overall tensile integrity [17]. The combination of rigid coir fibers and flexible plantain fibers produces a synergistic enhancement in stiffness, crack resistance, and toughness not achievable with single-fiber reinforcement. These results validate the successful load transfer between fibers and the TPS–lignin polymer matrix, supported by strong adhesion as confirmed by SEM.

SEM Morphological Analysis

The SEM micrographs (Figure 3a-3c) provide crucial insights into the surface morphology of the fibers and the interfacial behavior within the composite. The microstructure of the broken tensile specimens reveals well-embedded fibers with minimal fiber pullout, indicating excellent interfacial bonding due to alkali treatment. This treatment removes surface impurities, increases fiber roughness, and exposes cellulose fibrils that allow stronger mechanical interlocking with the polymer matrix. The key observations are:

- Coir fibers show distinct surface fibrillation, confirming removal of hemicellulose and waxy substances.
- Plantain fibers display a cleaner, more porous texture, promoting better wetting by the polymer matrix.
- Hybrid composites exhibit the most uniform fiber dispersion and minimal voids.
- Fracture surfaces reveal broken fibers rather than smooth pullout, demonstrating efficient stress transfer.
- Regions labeled “Good adhesion” and “Reduced pull-out” demonstrate the formation of a strong interphase – an essential requirement for mechanically reliable polymer-composites.

The micrographs confirm that fiber treatment and lignin incorporation both contributed to improved compatibility and a higher-quality composite structure [18–20].

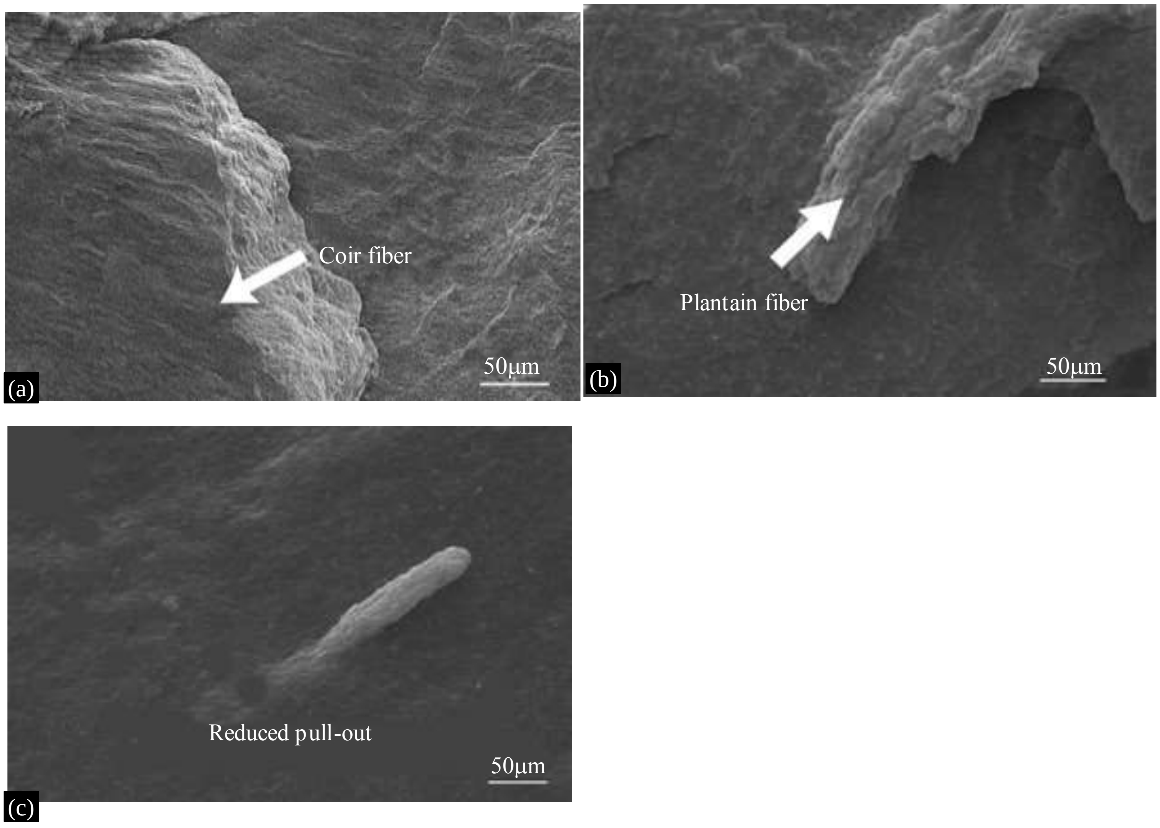

Figure 3. (a-c) - Micrographs showing coir and plantain fiber reinforcement, fracture features, and reduced fiber pull-out in the TPS–lignin hybrid composite.

Thermal Stability (TGA)

The TGA curves (Figure 4) reveal a clear improvement in thermal stability for all fiber-reinforced composites compared to the neat TPS–lignin matrix.

The TPS–lignin matrix exhibits a sharp and rapid mass loss beginning around 250°C, corresponding to the main degradation of starch. Coir- and plantain-reinforced composites show delayed mass-loss behavior, with their curves shifting slightly toward higher temperatures relative to the matrix. This indicates that the presence of treated natural fibers enhances thermal resistance by restricting polymer chain mobility and improving structural integrity [21]. Among the single-fiber composites, the coir-reinforced system displays marginally higher thermal stability due to the naturally higher lignin content of coir.

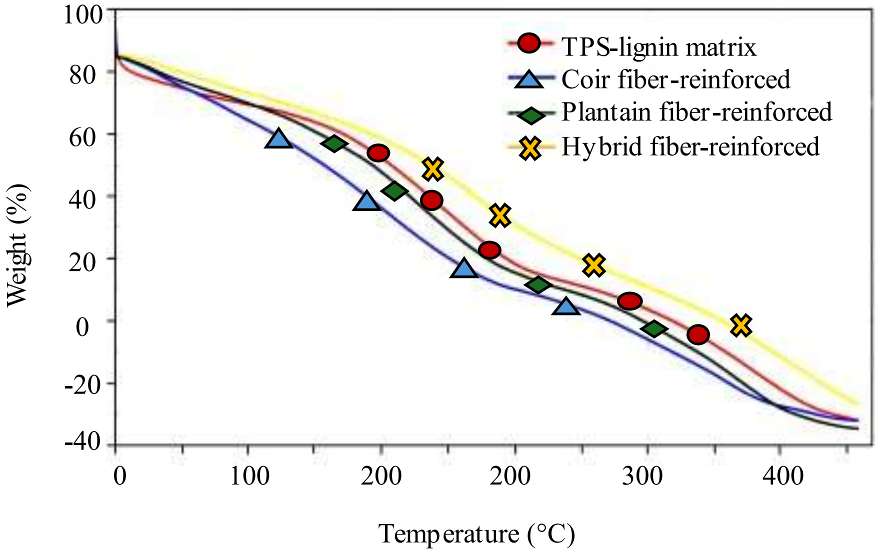

Figure 4. TGA curves of TPS–lignin and fiber-reinforced composites.

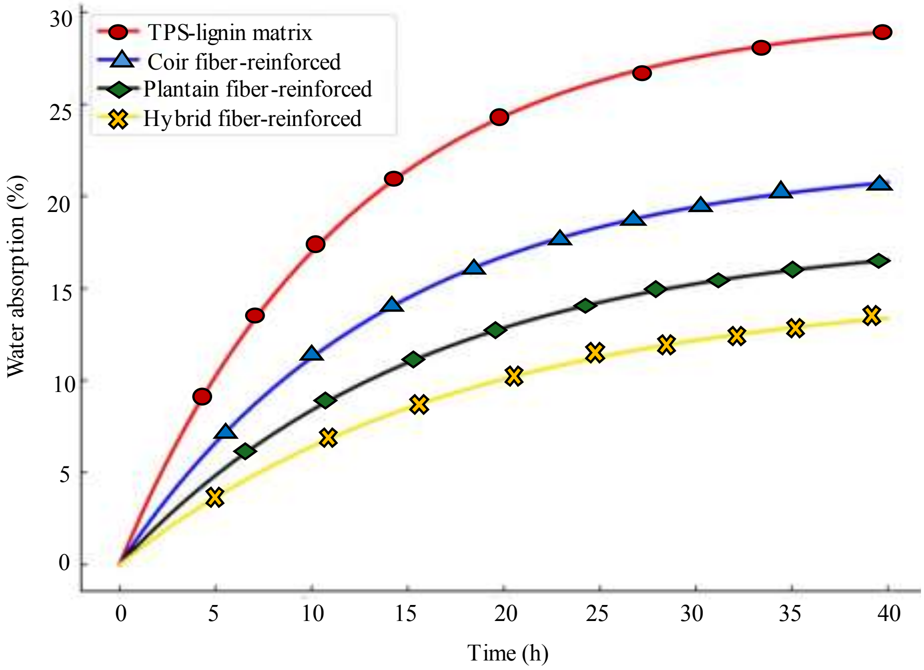

Figure 5. Water absorption behavior of TPS–lignin and fiber-reinforced composites.

The hybrid composite demonstrates the most gradual weight-loss profile, retaining the highest mass fraction throughout the heating cycle. Its curve remains above all other samples from the onset of decomposition until the final temperature range, indicating the best thermal stability within the group. This behavior is attributed to the combined effects of increased char formation, improved fiber–matrix interfacial adhesion, and the synergistic reinforcement provided by both coir and plantain fibers [22]. Overall, the delayed mass-loss trends and higher final residue in the hybrid composite confirm that fiber reinforcement – especially hybrid reinforcement – significantly enhances the thermal durability of TPS–lignin polymer composites.

Water Absorption Behavior

Water absorption curves (Figure 5) illustrate the diffusion of moisture into the composites over time. The TPS–lignin matrix absorbs water rapidly due to the hydrophilic nature of starch. Lignin partially reduces this effect but is insufficient for full protection.

Fiber reinforcement leads to notable reductions in water absorption due to three mechanisms:

- Alkali treatment reduces hydrophilic components (hemicellulose).
- Lignin in coir fibers naturally increases hydrophobicity.
- Good matrix adhesion reduces voids, decreasing water diffusion pathways.

Plantain-fiber composites show moderate reduction in water absorption. Coir composites perform better due to additional lignin content in the fiber itself. The hybrid composites exhibit the lowest moisture uptake due to optimal packing, improved crystallinity, and dual-fiber reinforcement sealing micro-channels [23]. Lower water absorption directly contributes to improved dimensional stability, reduced swelling, and longer service life – critical attributes for natural fiber polymer-composites intended for humid or outdoor environments. Although direct density and porosity values were not measured, reduced water absorption and SEM evidence of fewer voids in the hybrid composites clearly indicate lower effective porosity and better compaction. Future studies may incorporate Archimedes density and pycnometric porosity analysis for further quantification [24–29].

CONCLUSION

The present work demonstrates how combining TPS–lignin matrices with dual natural-fiber reinforcement leads to a new class of mechanically strong, thermally stable, and moisture-resistant biodegradable composites. The key conclusion points are:

- Hybrid fiber reinforcement significantly improved tensile strength, yielding a 55–70% enhancement over the TPS–lignin matrix due to synergistic stress distribution between coir and plantain fibers.
- SEM micrographs confirmed enhanced interfacial adhesion, showing reduced fiber pull-out, surface fibrillation after alkali treatment, and uniform fiber dispersion – critical for effective load transfer.
- TGA analysis demonstrated superior thermal stability in hybrid composites, which retained the highest residual mass and exhibited a more gradual degradation profile than single-fiber or neat matrix samples.
- Water absorption decreased by more than 40% in hybrid composites, attributed to lignin-rich coir fibers, improved crystallinity, and reduced void pathways within the matrix.
- Hybrid composites consistently outperformed single-fiber composites in mechanical, morphological, thermal, and moisture-resistance evaluations.

The combined improvements validate the potential of these bio-derived TPS–lignin–fiber hybrids as sustainable replacements for petroleum-based composites in packaging, automotive interiors, lightweight structural panels, and eco-engineering applications.

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