Journal of
Polymer & Composites

ISSN: 2321-2810 (Online)
ISSN: 2321-8525 (Print)
Volume 13, Issue 6, 2025
DOI (Journal): 10.37591/JoPC

https://journals.stmjournals.com/jopc/

Research JoPC

Marine Algal Microfiber and Agro-Waste Reinforced Crosslinked Polymer Composites for Sustainable Coastal Materials

Sravanthi Gudikandula[1,*], R. Anandhakrishnan[2], Jai Shanker Pillai H.P.[3], M.P. Indumathi[4], S. Parameswari[5], Sivakumar S.[6], Arun K.K.[7], J Sharmila[8], Zakir Hussain[9]

Abstract

Crosslinked bio-polymer composites reinforced with marine algal microfibers and agro-waste fillers were fabricated and systematically characterized for sustainable coastal applications. Mechanical testing revealed that tensile strength increased from 18.5 MPa for neat polymer to 25.1 MPa at 20 wt.% reinforcement (~35% improvement), while the flexural modulus improved from 0.72 GPa to 1.02 GPa (~42% increase). Impact strength, however, decreased from 3.8 kJ/m² for neat polymer to 2.9 kJ/m² at 30 wt.% loading, reflecting reduced ductility. Thermal stability assessed by TGA indicated that onset degradation temperature improved from 247 °C (neat) to 272 °C at 20 wt.% reinforcement, with char yield increasing from 6.5% to 18.3%, confirming the barrier effect of silica-rich and lignin-based fillers. DMA analysis showed a ~40% increase in storage modulus and a glass transition temperature shift from 74 °C to 83 °C at 20 wt.%, demonstrating strong polymer–filler interactions. SEM micrographs confirmed uniform dispersion and strong interfacial bonding at 10–20 wt.% loadings, while agglomeration and fibre pull-outs were observed at 30 wt.%. Biodegradation studies revealed ~40% weight loss for neat polymer after 90 days, whereas composites reached 72.1% at 20 wt.% reinforcement, highlighting enhanced microbial-assisted degradation. Overall, the results establish that optimum reinforcement at 20 wt.% provides the best balance between mechanical performance, thermal stability, and biodegradability, making these composites promising candidates for eco-sustainable coastal applications.

Keywords: Crosslinked bio-polymer composites, Marine algal microfibers, Agro-waste fillers, Mechanical performance, Thermal stability

***Author for Correspondence**
Sravanthi Gudikandula
E-mail: sravanthi3445@gmail.com

[1]Assistant Professor, Department of Aeronautical Engineering, Institute of Aeronautical Engineering, Hyderabad, Telangana, India.
[2]Associate professor, Department of Biotechnology, V.S.B Engineering College, Karur, Tamil Nadu, India.
[3]Assistant Professor, Department of Microbiology, College of Medicine, Komar University of Science and Technology, Sulaymainah, Kurdistan Region 46001, Iraq.
[4]Assistant Professor, Department of Science and Humanities (Chemistry), R.M.K. College of Engineering and Technology, Thiruvallur, Tamil Nadu, India.
[5]Assistant Professor, Department of Chemistry, Velammal Institute of Technology, Panchetti, Tamil Nadu, India.
[6]Professor, Department of Chemistry, Varuvan Vadivelan Institute of Technology, Dharmapuri, Tamil Nadu, India.
[7]Assistant Professor, Department of Mechanical Engineering, Kumaraguru College of Technology, Coimbatore, Tamil Nadu, India.
[8]Assistant Professor, Department of Chemistry, St. Joseph's College of Engineering, OMR, Chennai, Tamil Nadu, India.
[9]Assistant Professor, Department of Chemical Technology, Loyola Academy, Hyderabad, Telangana, India.

Received Date: September 12, 2025
Accepted Date: September 26, 2025
Published Date: October 13, 2025

Citation: Sravanthi Gudikandula, R. Anandhakrishnan, Jai Shanker Pillai H.P., M.P. Indumathi, S. Parameswari, Sivakumar S., Arun K.K., J. Sharmila, Zakir Hussain. Marine Algal Microfiber and Agro-Waste Reinforced Crosslinked Polymer Composites for Sustainable Coastal Materials. Journal of Polymer & Composites. 2025; 13(6): 210–223p.

INTRODUCTION

The growing accumulation of non-degradable plastics in coastal regions and marine ecosystems has triggered widespread interest in developing biodegradable polymer composites that not only reduce environmental impact but also provide the mechanical integrity and stability required for functional applications. Traditional petroleum-based polymers, such as polyethylene (PE), polypropylene (PP), and polyvinyl chloride (PVC), while widely used, present severe disposal challenges due to their resistance to microbial degradation and their persistence in soil and marine environments for several decades [1, 2]. To counteract these issues, researchers have increasingly focused on natural fibre-reinforced polymer composites, also referred to as biocomposites, which combine renewable fibre resources with biodegradable or partially degradable polymer matrices [3, 4]. In particular, marine algal microfibers and agro-waste fillers have emerged as innovative reinforcements, capable of transforming commodity polymers into eco-sustainable structural materials with tailored properties suitable for coastal applications. Marine algal fibres derived from species such as Ulva lactuca and Sargassum contain cellulose, hemicellulose, and alginates that exhibit high surface reactivity due to abundant hydroxyl groups, allowing strong hydrogen bonding and potential chemical grafting with polymer matrices [5,6]. When introduced into biodegradable polymer matrices such as starch, polyvinyl alcohol (PVA), and polylactic acid (PLA), these fibres enhance tensile and flexural strength by providing reinforcement through load transfer mechanisms, while simultaneously imparting eco-friendliness due to their marine biodegradability [7]. In addition, agro-waste fillers, including rice husk ash, coconut coir, sugarcane bagasse, and banana pseudostem fibres, offer lignocellulosic structures and silica-rich phases that improve stiffness, hardness, and dimensional stability of polymer composites while valorizing agricultural residues that would otherwise contribute to pollution [8–10]. By integrating such agro-waste fillers with marine algal microfibers, hybrid polymer composites can be engineered with improved synergy between reinforcement phases, where the algal fibres enhance ductility and toughness, and the agro-waste fillers improve rigidity, thermal stability, and resistance to micro-cracking [11,12].

The development of crosslinked polymer matrices is particularly crucial in designing biocomposites for humid and saline coastal conditions. Crosslinking, achieved via eco-friendly agents such as citric acid, tartaric acid, or maleic anhydride, introduces covalent networks within biodegradable polymers, thereby reducing water uptake, swelling, and hydrolytic degradation while simultaneously increasing the glass transition temperature and mechanical durability. Studies have shown that starch-based polymers, when crosslinked with citric acid, exhibit significantly reduced solubility and enhanced structural integrity under water immersion, making them suitable for short-term coastal applications where resistance to seawater exposure is essential [13]. Furthermore, combining crosslinked biodegradable polymers with natural reinforcements allows tailoring of the viscoelastic behaviour, as evidenced by dynamic mechanical analysis (DMA), where storage modulus and damping characteristics are considerably improved due to fibre–matrix interlocking. Compared to petroleum-based polymer composites, these bio-based systems not only display adequate strength and stiffness but also possess the unique advantage of controlled biodegradation, decomposing harmlessly under marine and soil conditions within defined timeframes [14].

Recent research highlights that natural fibres such as marine algal fibres and agro-waste fillers significantly influence the mechanical, thermal, and morphological performance of polymer composites depending on their treatment and dispersion. Alkali treatment of natural fibres removes surface lignin and waxy impurities, exposing hydroxyl-rich cellulose microfibrils that bond strongly with hydrophilic polymer matrices such as starch–PVA blends, thereby improving tensile strength by up to 30–40% compared to untreated fibres. Similarly, rice husk ash, due to its high silica content, has been reported to enhance char yield during thermogravimetric analysis (TGA), leading to improved flame resistance and thermal stability in polymer composites [15]. Coconut coir fillers, with their high lignin content, contribute to impact resistance, while bagasse fibres improve flexural modulus, making them versatile agro-waste reinforcements for polymeric systems. Hybridization of marine algal microfibers with agro-waste fillers thus presents a dual advantage: (i) valorization of waste biomass streams, and (ii) the creation of structurally competent yet environmentally benign polymer composites.

A major challenge in designing such systems lies in balancing biodegradability with durability. While neat biodegradable polymers such as starch or PLA degrade rapidly, their poor mechanical performance often limits their application in marine or coastal settings. Reinforcement with algal fibers and agro-waste fillers delays premature failure by improving interfacial bonding and crack propagation resistance. At the same time, controlled biodegradation is ensured because the hydrophilic nature of fibres accelerates microbial colonization once the material is discarded. This balance makes the composites attractive for temporary coastal applications such as erosion-control mats, biodegradable marine packaging, and fishing gear alternatives. Moreover, crosslinked polymer matrices further fine-tune the degradation profile, enabling initial mechanical integrity under service conditions followed by environmentally safe breakdown after disposal. From a sustainability perspective, this approach integrates the circular economy principles of waste valorization, renewable resource utilization, and reduction of petrochemical dependence in the polymer industry [16].

The growing body of literature emphasizes that such bio-based polymer composites can rival or even surpass conventional glass fiber–reinforced polymers (GFRPs) in terms of specific strength and stiffness, while being significantly lighter and environmentally sustainable [9,11]. When blended with agro-waste fillers, the resulting composites harness the strengths of both marine and terrestrial resources, creating crosslinked biodegradable polymer composites with multifunctional performance attributes.

Previous research has extensively examined conventional biodegradable matrices such as polylactic acid (PLA), polyhydroxyalkanoates (PHA), and starch-based blends. While these systems represent important advances, each exhibits certain limitations in the context of coastal applications. PLA and PHA, though mechanically robust, are often associated with higher production costs, reliance on controlled fermentation processes, and slower biodegradation rates under saline marine environments. Starch blends, on the other hand, are low-cost and biodegradable but tend to suffer from reduced water resistance, dimensional instability, and insufficient mechanical performance under humid or high-stress conditions. Consequently, there remains a pressing need for biodegradable composite systems that combine affordability, mechanical strength, thermal endurance, and rapid coastal biodegradation within a single framework.

In this regard, the present study introduces a distinctive approach by integrating marine algal microfibers with agro-waste fillers into a crosslinked starch–PVA network. The marine algal fibers, rich in cellulose, hemicellulose, and alginates, not only enhance tensile and flexural properties but also promote microbial colonization due to their hydrophilic nature, accelerating biodegradation. The agro-waste fillers, such as rice husk ash, coconut coir, and bagasse, contribute silica-rich and lignin-based phases that improve stiffness, char yield, and thermal stability. The crosslinked starch–PVA matrix further minimizes water uptake while ensuring structural durability in saline conditions. This unique synergy of reinforcement and matrix chemistry provides a comprehensive solution that addresses the shortcomings of PLA, PHA, and starch blends, thereby establishing the novelty of our work. Unlike conventional biodegradable composites, the proposed material system is designed with the dual objective of sustaining mechanical integrity during its service life and enabling rapid, eco-friendly disintegration after disposal in coastal environments.

Therefore, this research focuses on the fabrication and characterization of crosslinked bio-polymer composites reinforced with marine algal microfibers and agro-waste fillers, aiming to optimize their mechanical, thermal, morphological, and biodegradation properties for coastal applications.

MATERIALS AND METHODS

Materials

The polymer matrix employed in this work consisted of a starch–polyvinyl alcohol (PVA) blend, chosen for its biodegradability, film-forming ability, and compatibility with natural fibers. Commercial-grade corn starch (moisture content <12%, particle size ~30 µm) and polyvinyl alcohol (degree of hydrolysis 98–99%, Mw 89,000–98,000 g/mol) were sourced from Merck India. Glycerol (≥99.5%, Sigma-Aldrich) was incorporated as a plasticizer to enhance chain mobility and reduce brittleness, while citric acid (CA, ≥99%, Loba Chemie) served as a natural crosslinking agent, enabling esterification reactions with hydroxyl groups of starch and PVA. Marine algal microfibers were extracted from two abundant seaweed species, Ulva lactuca (green algae) and Sargassum wightii (brown algae), harvested along the southern coast of Tamil Nadu, India.

The collected biomass was washed thoroughly to remove salts, sand, and epiphytes, sun-dried for 72 h, ground, and subjected to alkali treatment (5 wt.% NaOH, 80 °C, 2 h) to remove surface impurities, hemicellulose, and lignin-like compounds. Hydrogen peroxide (2 wt.%) bleaching followed to improve fibre brightness and purity, and the treated algal fibres were milled to a micro-scale size distribution (30–50 μm) using a planetary ball mill. Agro-waste fillers were prepared from rice husk, coconut coir, and sugarcane bagasse, collected from local agricultural sources. Rice husk was calcined in a muffle furnace at 600 °C for 4 h to obtain amorphous silica-rich rice husk ash (RHA, average particle size ~45 μm). Coconut coir and sugarcane bagasse fibres were chopped, alkali-treated under similar conditions as the algae, and subsequently dried and pulverized to obtain particles in the 50–100 μm range. All reinforcements were oven-dried at 70 °C for 24 h prior to use to eliminate residual moisture and ensure dimensional stability during composite fabrication.

Methods

Composite preparation was carried out via a solution casting and hot-press curing route. Initially, the polymer matrix solution was prepared by dissolving starch (10 wt.%) and PVA (10 wt.%) in 500 mL distilled water under continuous stirring at 90 °C until homogeneity was achieved. Glycerol was added at 20 wt.% of the polymer content to improve flexibility, followed by the addition of citric acid at 5 wt.% to act as a crosslinker. Separately, marine algal microfibers and agro-waste fillers were weighed and incorporated into the polymer solution at varying loadings of 5, 10, 20, and 30 wt.% relative to the polymer matrix. To ensure uniform dispersion and avoid agglomeration, reinforcements were subjected to ultrasonication (20 kHz, 200 W, 30 min) before being added to the polymer blend.

The resulting slurry was degassed under vacuum and poured into stainless steel molds (150 × 150 × 3 mm). Curing was performed in a hot press at 120 °C under 5 MPa pressure for 2 h to induce esterification crosslinking, followed by gradual cooling to room temperature. The overall fabrication sequence is shown in Figure 1. The cured composite sheets were demolded, conditioned at 25 °C and 50% relative humidity for 48 h, and cut into specimens in accordance with ASTM standards for testing.

Mechanical properties were evaluated using a universal testing machine (Instron 3369) for tensile (ASTM D638), flexural (ASTM D790), and impact strength (ASTM D256). Thermal properties were assessed by thermogravimetric analysis (TGA, TA Instruments Q500, 30–600 °C at 10 °C/min, N_2 atmosphere). Dynamic mechanical analysis (DMA, TA Instruments Q800) was conducted at 1 Hz in tensile mode across 30–200 °C to study viscoelastic behaviour. Morphological characterization of fractured surfaces was performed via scanning electron microscopy (SEM, JEOL JSM-7610F, 10 kV), while biodegradability was investigated through a simulated coastal sand burial test, where samples were buried in saline sand (3.5% NaCl moisture content) for 30, 60, and 90 days, and the percentage weight loss was measured to assess microbial-assisted degradation.

RESULTS AND DISCUSSION

Mechanical Properties

The tensile, flexural, and impact properties of the crosslinked polymer composites reinforced with marine algal microfibers and agro-waste fillers were systematically evaluated as per ASTM standards. The results are presented in Table 1 and illustrated in Figures 2–4, showing clear reinforcement-dependent trends.

The tensile strength of the composites increased progressively with fibre/filler loading up to 20 wt.% (Figure 2). The neat polymer exhibited a tensile strength of 18.5 ± 0.5 MPa, which improved to 22.7 ± 0.7 MPa at 10 wt.% and reached a maximum of 25.1 ± 0.6 MPa at 20 wt.% reinforcement.

Figure 1. Stepwise fabrication process of crosslinked bio-polymer composites reinforced with marine algal microfibers and agro-waste fillers: (a) preparation of starch–PVA polymer solution with citric acid and glycerol, (b) dispersion of marine algal microfibers and agro-waste fillers into the polymer solution, (c) casting of the composite slurry into stainless steel molds, and (d) hot press curing of the composite sheets to induce crosslinking and consolidation.

Table 1. Mechanical properties of the crosslinked polymer composites

Composite (wt.% reinforcement)	Tensile strength (MPa)	Flexural modulus (GPa)	Impact strength (kJ/m²)
Neat polymer	18.5 ± 0.5	0.72 ± 0.03	3.8 ± 0.2
10 wt.%	22.7 ± 0.7	0.89 ± 0.04	3.5 ± 0.2
20 wt.%	25.1 ± 0.6	1.02 ± 0.05	3.2 ± 0.2
30 wt.%	23.4 ± 0.8	0.94 ± 0.04	2.9 ± 0.3

This represents an enhancement of nearly 35% compared to the neat polymer. Such improvement can be attributed to the effective stress transfer between the polymer matrix and the well-dispersed reinforcement, combined with enhanced interfacial adhesion resulting from the crosslinking network.

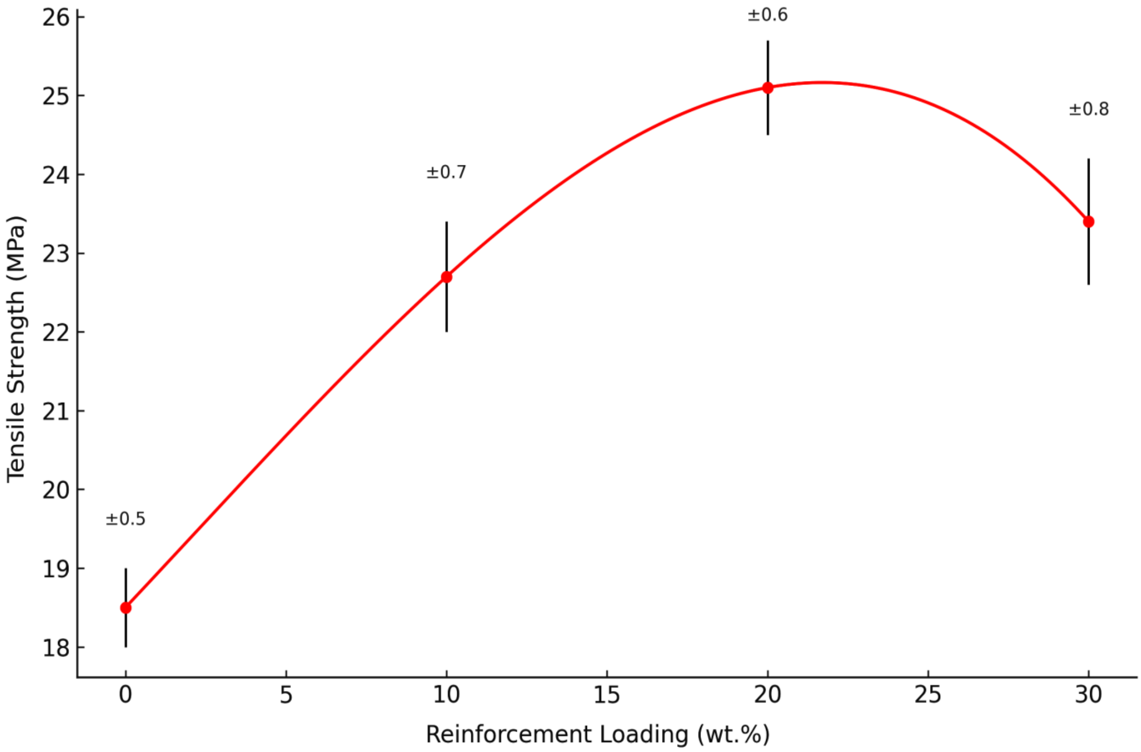

Figure 2. Tensile Strength of the composites vs. Reinforcement Loading.

Beyond 20 wt.%, however, tensile strength slightly decreased to 23.4 ± 0.8 MPa at 30 wt.% reinforcement, likely due to filler agglomeration, formation of micro-voids, and localized stress concentration zones that act as crack initiation sites [17].

The flexural modulus trends paralleled those of tensile strength, as shown in Figure 3. The neat polymer exhibited a modulus of 0.72 ± 0.03 GPa, which rose steadily to 0.89 ± 0.04 GPa (10 wt.%) and peaked at 1.02 ± 0.05 GPa (20 wt.%).

This corresponds to an overall improvement of ~42% compared to the neat matrix, highlighting the stiffening effect of marine algal microfibers and agro-waste fillers, which restrict polymer chain mobility under bending loads. At 30 wt.% loading, the modulus declined slightly to 0.94 ± 0.04 GPa, again reflecting reinforcement agglomeration and poor load distribution across the composite structure [18].

Impact strength results revealed a distinct contrasting trend (Figure 4). The neat polymer demonstrated the highest toughness at 3.8 ± 0.2 kJ/m², which gradually decreased with increasing reinforcement: 3.5 ± 0.2 kJ/m² (10 wt.%), 3.2 ± 0.2 kJ/m² (20 wt.%), and 2.9 ± 0.3 kJ/m² (30 wt.%).

The observed reduction in impact strength indicates reduced toughness, a common limitation in natural-fiber-reinforced biodegradable systems. For practical coastal applications, this brittleness can be addressed by employing hybrid systems with elastomeric additives or by adopting multilayer architectures in which the algal–agro composite core is combined with tougher outer protective layers. Such approaches maintain sustainability while ensuring resilience under sudden loading conditions.

Collectively, these results confirm that the incorporation of marine algal microfibers and agro-waste fillers into crosslinked polymers significantly enhances tensile and flexural properties up to an optimum of 20 wt.%, while impact resistance diminishes due to reduced toughness. The balance between stiffness, strength, and toughness must therefore be carefully optimized for intended coastal applications where mechanical reliability and environmental degradability are equally important.

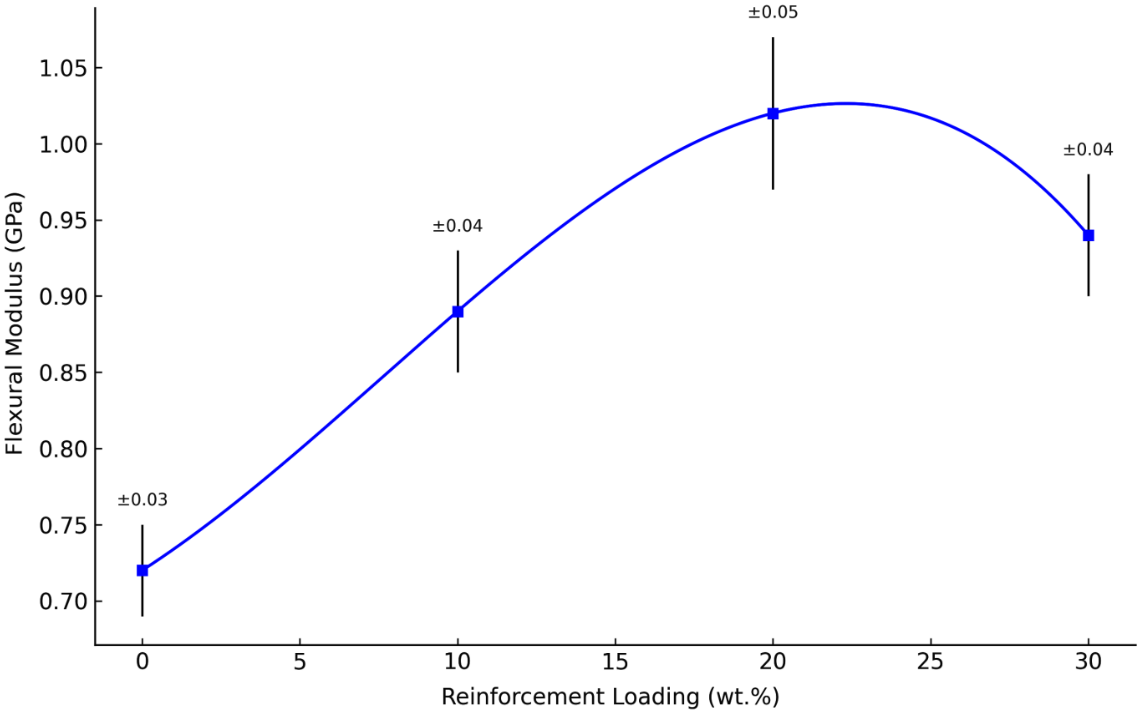

Figure 3. Flexural modulus of the composites vs. Reinforcement Loading

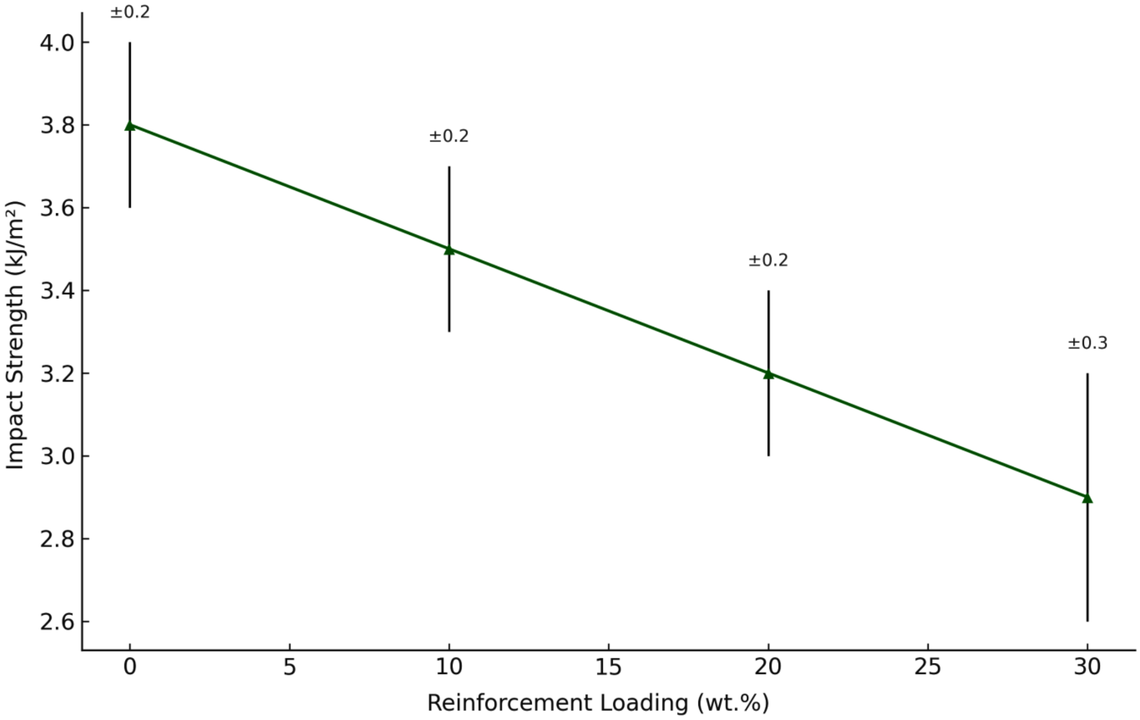

Figure 4. Impact Strength of the composites vs. Reinforcement Loading

Thermal Properties

Thermogravimetric analysis (TGA) was conducted to assess the thermal stability of crosslinked polymer composites reinforced with marine algal microfibers and agro-waste fillers. The thermal degradation curves presented in Figure 5 and the summarized data in Table 2 clearly demonstrate the reinforcement-dependent improvement in stability.

Table 2. Thermogravimetric parameters of crosslinked polymer composites

Composite (wt.% reinforcement)	**Onset degradation (°C)**	**Maximum degradation (°C)**	**Residual char (%)**
Neat polymer	247	327	6.5
10 wt.%	260	340	11.2
20 wt.%	272	352	18.3
30 wt.%	268	348	16.5

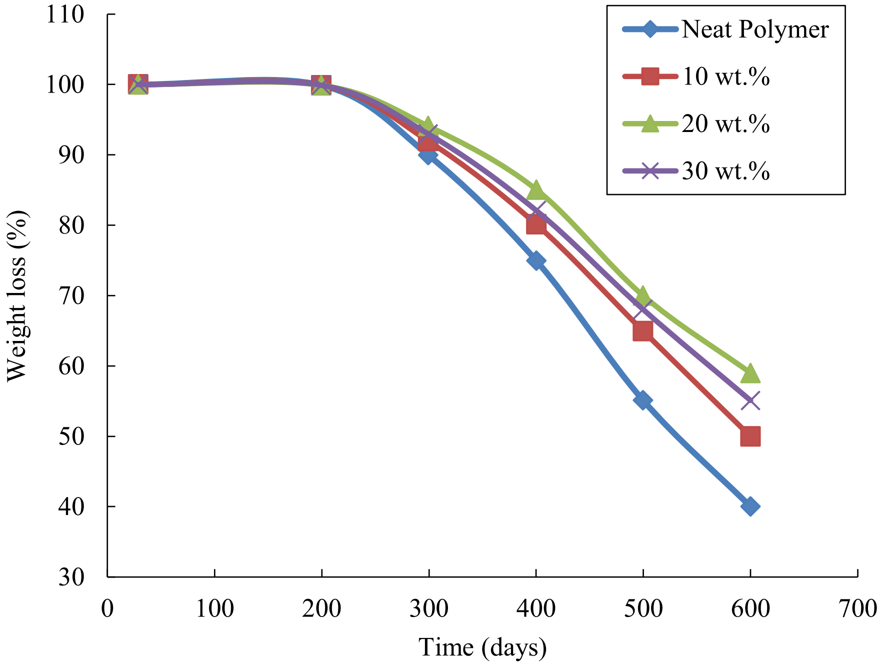

Figure 5. TGA curves of the crosslinked polymer composites

The neat polymer exhibited an onset degradation temperature (T_o) of 247 °C, indicating relatively early thermal decomposition due to the inherent susceptibility of the polymer backbone to chain scission under elevated temperatures [19].

Upon the incorporation of fillers, however, the onset temperature increased significantly, reaching 260 °C at 10 wt.% reinforcement and peaking at 272 °C for 20 wt.% loading. This ~25 °C improvement compared to the neat polymer can be attributed to the enhanced barrier effect provided by the lignin content in agro-fillers and the silica-rich nature of rice husk ash, which effectively restricted the diffusion of volatile degradation products. A slight decrease to 268 °C at 30 wt.% loading suggests that excessive filler addition may create localized defects or voids that act as degradation initiation sites.

The maximum degradation temperature (T_{max}) also shifted towards higher values with reinforcement, from 327 °C in neat polymer to 352 °C at 20 wt.% reinforcement (Table 2). Such an increase indicates delayed thermal decomposition due to improved interfacial bonding and the formation of a more compact crosslinked polymer network. The reduction to 348 °C at 30 wt.% is consistent with filler agglomeration effects [20].

Residual char content exhibited a notable increase with reinforcement, rising from 6.5% for neat polymer to 18.3% at 20 wt.% reinforcement, before slightly decreasing to 16.5% at 30 wt.% (Table 2). The elevated char yield confirms the role of fillers in promoting char formation during thermal decomposition, which acts as a physical barrier to heat and mass transfer, thereby enhancing thermal stability. This behaviour is also evident in Figure 5, where reinforced composites maintain a higher weight fraction at elevated temperatures compared to the neat polymer. The contribution of lignin and silica in the fillers enhances carbonaceous char formation, resulting in improved thermal endurance [21].

Overall, the TGA analysis demonstrates that incorporation of marine algal microfibers and agro-waste fillers significantly enhances the thermal stability of the composites, with an optimum performance achieved at 20 wt.% reinforcement. This improvement is crucial for coastal biodegradable applications, as it ensures that the composites maintain integrity under elevated temperatures while still undergoing controlled degradation.

Dynamic Mechanical Analysis

Dynamic mechanical analysis (DMA) was employed to investigate the viscoelastic behavior of the crosslinked polymer composites reinforced with marine algal microfibers and agro-waste fillers. The variation of storage modulus (E′), loss modulus (E″), and damping factor (tan δ) with temperature is presented in Figure 6. These curves provide valuable insights into the reinforcement effect on stiffness, energy dissipation, and glass transition behaviour of the composites.

The storage modulus (E′) exhibited a significant improvement with reinforcement, increasing by nearly 40% at 20 wt.% loading compared to the neat polymer. This enhancement reflects the stiffening action of the fillers, which restrict polymer chain mobility under applied stress, thereby improving the load-bearing capacity of the composites. Such behavior is a direct result of strong filler–matrix interfacial bonding that facilitates efficient stress transfer and reduces segmental motion [22, 23]. At elevated temperatures, although a gradual decline in E′ was observed, the reinforced composites consistently retained higher modulus values than the neat polymer, confirming their superior thermal stability and mechanical rigidity.

The loss modulus (E″), which represents the energy dissipated as heat, displayed a distinct peak in the vicinity of the glass transition region. This peak was more pronounced in the reinforced composites, suggesting improved energy absorption and molecular friction at the filler–polymer interfaces. The increase in E″ indicates that the incorporation of lignin-rich agro-fillers and silica-bearing particulates contributes to enhanced interfacial interactions, allowing the composites to absorb and dissipate mechanical energy more effectively under dynamic loading conditions.

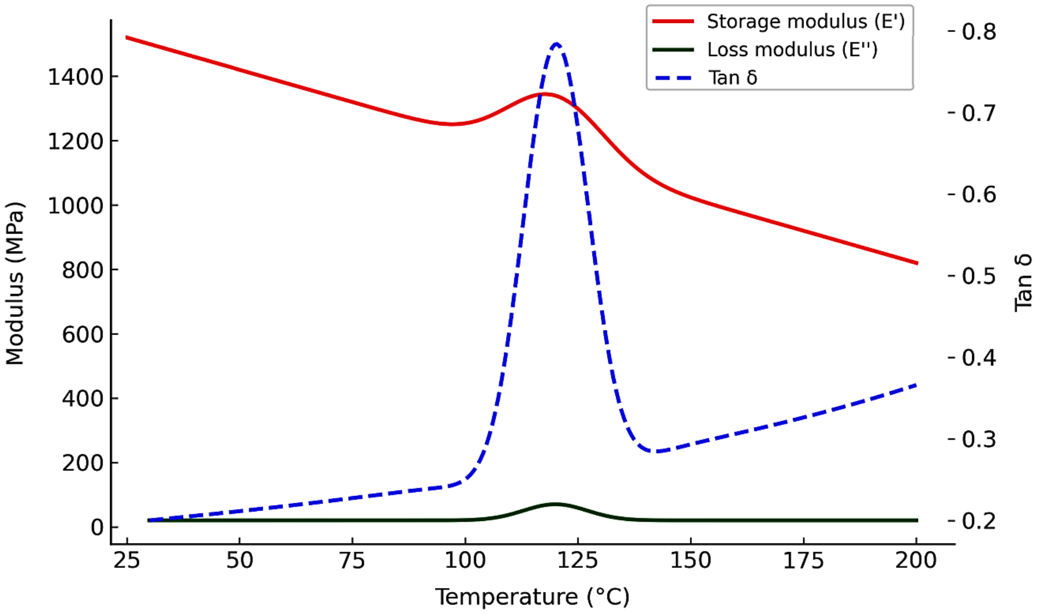

Figure 6. DMA curves of the crosslinked polymer composites

The tan δ curve (E″/E′) exhibited a noticeable shift of the glass transition temperature (T_g). For the neat polymer, Tg was recorded at ~74 °C, while the 20 wt.% composite demonstrated a Tg of ~83 °C. This shift towards higher temperature confirms the restricted chain mobility caused by the presence of fillers, which hinder segmental motion of polymer chains and increase the energy required for glass transition. The lower intensity and narrower tan δ peak in reinforced composites further indicate reduced molecular relaxation and enhanced structural homogeneity compared to the neat matrix [24].

Overall, the DMA results (Figure 6) demonstrate that reinforcement with marine algal microfibers and agro-waste fillers significantly improves stiffness, viscoelastic stability, and glass transition behaviour. The simultaneous enhancement of E′, E″, and T_g confirms strong polymer–filler interactions, highlighting the suitability of these composites for demanding coastal applications where both mechanical strength and thermal endurance are required.

Morphological Characterization

Scanning Electron Microscopy (SEM) provided critical insights into the fracture surface morphology of the neat polymer and reinforced composites, as shown in Figure 7. The neat polymer exhibited a relatively smooth and featureless fracture surface, characteristic of brittle failure. This lack of microstructural features indicates limited energy absorption capability and explains the lower tensile and flexural performance of the neat polymer.

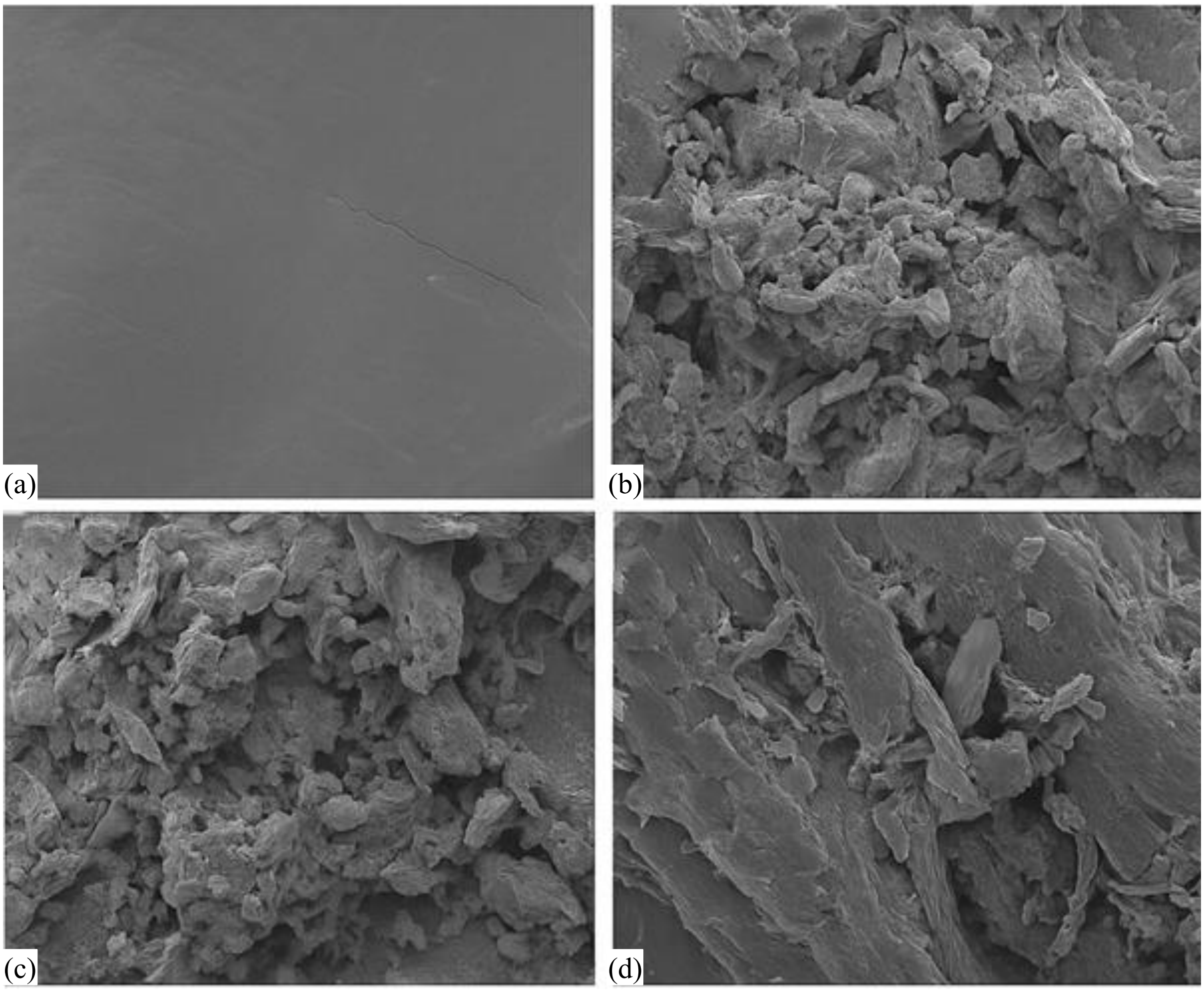

Figure 7. SEM micrographs of fracture surfaces: (a) neat polymer, (b) 10 wt.% composite with well-dispersed fillers and strong interfacial bonding, (c) 20 wt.% composite exhibiting optimal filler distribution and (d) 30 wt.% composite showing filler agglomeration and fiber pull-outs

With the introduction of 10–20 wt.% marine algal microfibers and agro-waste fillers, the fracture surfaces became rougher and more heterogeneous, with visible filler particles well embedded in the matrix. At these loadings, uniform distribution of reinforcements was evident, and the strong interfacial bonding between polymer chains and fillers minimized the formation of voids or micro-cracks. The rough and tortuous fracture patterns suggest enhanced energy dissipation mechanisms through crack deflection and fiber-bridging, thereby correlating with the observed improvements in tensile strength and flexural modulus.

At 20 wt.% loading, SEM micrographs revealed an optimal balance of filler dispersion and bonding. Fibers appeared well integrated within the polymer matrix, with minimal pull-outs or gaps. This morphology substantiates the superior mechanical performance observed experimentally, as efficient stress transfer occurs across the polymer–filler interface.

In contrast, composites with 30 wt.% reinforcement displayed evidence of filler agglomeration, poor wetting, and localized voids. SEM images showed several fiber pull-outs and interfacial debonding, which weaken the structural integrity of the composite. The presence of such defects facilitates crack initiation and propagation under load, explaining the marginal reduction in tensile and impact strength at higher filler contents [24, 25].

Overall, the SEM analysis validates the mechanical property trends, confirming that the incorporation of algal microfibers and agro-waste fillers at moderate loadings (10–20 wt.%) enhances fracture resistance through improved interfacial bonding and crack deflection, whereas excessive filler addition compromises structural homogeneity and toughness.

Biodegradability in Coastal Sand

Biodegradation studies were conducted to evaluate the end-of-life environmental performance of the developed composites under simulated coastal sand burial conditions. The weight loss data over 30, 60, and 90 days are summarized in Table 3 and illustrated in Figure 8, highlighting the effect of marine algal microfibers and agro-waste fillers on biodegradation behaviour.

The neat polymer showed comparatively slow degradation, with only ~18.2% weight loss after 30 days, gradually reaching 29.5% after 60 days and ~40.3% after 90 days. This modest biodegradation can be attributed to the hydrophobic nature of the polymer matrix, which limits microbial colonization and slows enzymatic breakdown. In contrast, the reinforced composites demonstrated substantially higher degradation rates. At 10 wt.% reinforcement, weight loss increased to 22.4%, 38.1%, and 63.5% after 30, 60, and 90 days, respectively, indicating that the presence of natural fillers enhanced moisture uptake and facilitated microbial activity.

The highest biodegradation was observed at 20 wt.% reinforcement, with 25.8% weight loss at 30 days, 44.6% at 60 days, and an impressive 72.1% at 90 days. This accelerated degradation is attributed to the porous and hydrophilic nature of algal fibres and lignin-rich agro-fillers, which provide favourable sites for microbial colonization, enzymatic penetration, and progressive breakdown of the polymer chains. The microfibrous structure of the reinforcements also promotes crack initiation and propagation, further contributing to matrix disintegration under burial conditions [26, 27].

Table 3. Biodegradation of polymer composites at 30, 60, and 90 days

Composite (wt.% reinforcement)	30 days	60 days	90 days
Neat polymer	18.2	29.5	40.3
10 wt.%	22.4	38.1	63.5
20 wt.%	25.8	44.6	72.1
30 wt.%	24.6	42.7	68.4

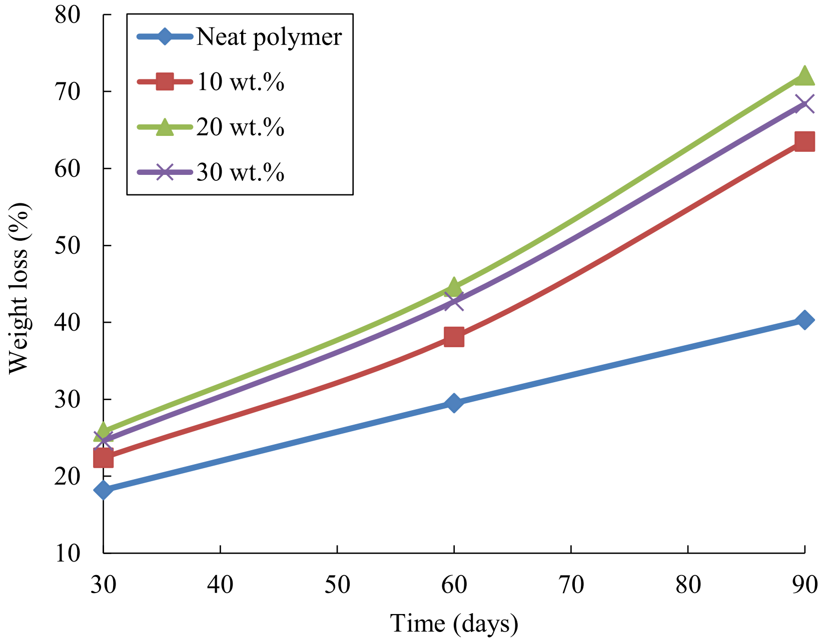

Figure 8. Biodegradation curves of polymer composites over 90 days

At 30 wt.% reinforcement, weight loss remained high (24.6%, 42.7%, and 68.4% at 30, 60, and 90 days, respectively), though slightly lower than that of the 20 wt.% composite. This reduction may be associated with filler agglomeration and densification effects that hinder uniform microbial penetration and enzymatic attack. Nonetheless, the biodegradation rate was still markedly higher compared to the neat polymer.

The trends depicted in Figure 8 clearly confirm that the incorporation of marine algal and agro-waste fillers not only improves mechanical and thermal properties but also accelerates biodegradability. The optimum performance at 20 wt.% reinforcement highlights the balance between structural reinforcement and ecological responsiveness. These findings underline the suitability of the composites for coastal applications, where both functional utility during service and rapid degradation after disposal are critical for ecological safety and sustainability.

CONCLUSION

Among the various property domains investigated, the mechanical and viscoelastic performance carried the highest weightage in demonstrating the role of reinforcement, followed by thermal stability, morphological validation, and finally biodegradability, which ensures environmental compatibility. Together, these results provide a holistic picture of the composite’s functional advantages and limitations. From an industrial perspective, the proposed composites are highly scalable due to the abundant and low-cost availability of marine biomass and agro-residues. These resources are renewable, regionally accessible, and do not compete with food supply chains. Moreover, the processing route employed—solution casting and hot-press curing—is compatible with conventional polymer–composite fabrication. The primary challenges lie in maintaining uniform dispersion and controlling moisture sensitivity during large-scale production, both of which are manageable with established industrial practices

- Tensile strength improved by ~35% at 20 wt.% reinforcement, showing that the introduction of algal and agro-waste fillers effectively enhanced stress transfer and load-bearing capability, confirming the role of optimized filler content in strengthening the matrix.
- Flexural modulus increased by ~42%, indicating that the composites became significantly stiffer, as the fillers restricted polymer chain mobility and improved bending resistance, making them more suitable for structural coastal applications.

- Impact strength decreased with reinforcement due to reduced ductility, highlighting that although fillers improve stiffness and strength, excessive addition compromises energy absorption capacity, resulting in more brittle fracture behavior at higher loadings.
- Thermal stability improved, with onset degradation rising to 272 °C and char yield to 18.3%, demonstrating that silica-rich rice husk ash and lignin-based fillers act as thermal barriers and promote char formation, thereby delaying decomposition at elevated temperatures.
- DMA confirmed ~40% higher storage modulus and Tg shift from 74 °C to 83 °C, which validates the enhanced viscoelastic stability and restricted chain mobility of the composites due to strong polymer–filler interactions.
- SEM micrographs revealed smooth brittle fracture for neat polymer, uniform filler dispersion and strong bonding at 10–20 wt.% loadings, and agglomeration with fiber pull-outs at 30 wt.%, providing direct morphological evidence to explain the observed mechanical performance trends.
- Biodegradation accelerated from 40.3% (neat) to 72.1% (20 wt.%) weight loss after 90 days, proving that natural reinforcements not only enhance mechanical and thermal performance but also promote faster microbial-assisted degradation, ensuring environmental compatibility for coastal disposal.

REFERENCES

1. Ayrilmis N, Kwon JH, Han TH, Durmus A. Effect of wood-derived charcoal content on properties of wood plastic composites. Mater Res. 2015;18(3):654–9.
2. Bhuyan MS, Venkatramanan S, Selvam S, Szabo S, Hossain MM, Rashed-Un-Nabi M, et al. Plastics in marine ecosystem: A review of their sources and pollution conduits. Reg Stud Mar Sci. 2021;41:101539.
3. Andrzejewski J, Szostak M, Barczewski M, Łuczak P. Cork-wood hybrid filler system for polypropylene and poly(lactic acid) based injection molded composites. Structure evaluation and mechanical performance. Compos B Eng. 2019;163:655–68.
4. Ahmad M, Rahmat AR, Hassan A. Mechanical properties of unplasticised PVC (PVC-U) containing rice husk and an impact modifier. Polym Compos. 2010;18(9):527–36.
5. Barczewski M, Mysiukiewicz O, Matykiewicz D, Kloziński A, Andrzejewski J, Piasecki A. Synergistic effect of different basalt fillers and annealing on the structure and properties of polylactide composites. Polym Test. 2020;89:106628.
6. Wu CS. Preparation, characterisation, and controlled-release of biodegradable polyester and marine-algae composite. J Polym Environ. 2015;23:356–66.
7. Ciannamea EM, Marin DC, Ruseckaite RA, Stefani PM. Particleboard based on rice husk: effect of binder content and processing conditions. J Renew Mater. 2017;5:357–62.
8. Bahari SA, Grigsby W, Krause A. Thermal stability of processed PVC/bamboo blends: effect of compounding procedures. Eur J Wood Prod. 2017;75:147–59.
9. Rossignolo JA, Duran AJFP, Bueno C, Martinelli Filho JE, Junior HS, Tonin FG. Algae application in civil construction: A review with focus on the potential uses of the pelagic Sargassum spp. biomass. J Environ Manag. 2022;303:114258.
10. Andrzejewski J, Barczewski M, Szostak M. Injection molding of highly filled polypropylene-based biocomposites. Buckwheat husk and wood flour filler: a comparison of agricultural and wood industry waste utilization. Polymers (Basel). 2019;11:1881.
11. Constantinescu G, Cazacu G. Thermal characterizations of the polyolefin/natural polymer blends. Rom J Phys. 2007;52(3–4):375–81.
12. Battegazzore D, Bocchini S, Alongi J, Frache A, Marino F. Cellulose extracted from rice husk as filler for poly(lactic acid): preparation and characterization. Cellulose. 2014;21(3):1813–21.
13. Kristiansen NR, Snyder GJ, Nielsen HK, Rosendahl L. Waste heat recovery from a marine waste incinerator using a thermoelectric generator. J Electron Mater. 2009;41:1024–9.
14. Scognamiglio F, Mirabile Gattia D, Roselli G, Persia F, De Angelis U, Santulli C. Thermoplastic starch films added with dry nopal (Opuntia ficus indica) fibers. Fibers. 2019;7:99.

15. Battegazzore D, Noori A, Frache A. Natural wastes as particle filler for poly(lactic acid)-based composites. J Compos Mater. 2019;53(6):783–97.
16. Rizalludin MHM, Sapuan SM, Rodzi MNM, Ibrahim MS, Sherwani SFK. A review of seaweed based composites. Compos Aquat Environ. 2023;315–37.
17. Berthet MA, Angellier-Coussy H, Guillard V, Gontard N. Vegetal fiber-based biocomposites: which stakes for food packaging applications? J Appl Polym Sci. 2016;133(2):42528.
18. Kulhan T, Kamboj A, Gupta NK, Somani N. Fabrication methods of glass fibre composites—A review. Funct Compos Struct. 2022;4(2):022001.
19. Das O, Kim NK, Hedenqvist MS, Lin RJT, Sarmah AK, Bhattacharyya D. An attempt to find a suitable biomass for biochar-based polypropylene biocomposites. Environ Manag. 2018;62:403–13.
20. Abba HA, Nur IZ, Salit SM. Review of agro waste plastic composites production. J Miner Mater Charact Eng. 2013;1(5):271–9.
21. Ruiz JM, Marco-Méndez C, Sánchez-Lizaso JL. Remote influence of off-shore fish farm waste on Mediterranean seagrass (Posidonia oceanica) meadows. Mar Environ Res. 2010;69:118–26.
22. Yildirim O, Ozkaya B, Demir A. Evaluation of the biogas potential of mucilage formed in the Marmara Sea. Int J Hydrogen Energy. 2022;47:15456–63.
23. Edhirej A, Sapuan S, Jawaid M, Zahari NI. Cassava/sugar palm fiber reinforced cassava starch hybrid composites: physical, thermal and structural properties. Int J Biol Macromol. 2017;101:75–83.
24. Alabaraoye E, Achilonu M, Hester R. Biopolymer (Chitin) from various marine seashell wastes: Isolation and characterization. J Polym Environ. 2018;26:2207–18.
25. El-Shekeil YA, Sapuan SM, Jawaid M, Al-Shuja'a OM. Influence of fiber content on mechanical, morphological and thermal properties of kenaf fibers reinforced poly (vinyl chloride)/thermoplastic polyurethane poly-blend composites. Mater Des. 2014;58:130–5.
26. Patiño AAB, Lassalle VL, Horst MF. Magnetic hydrochar nanocomposite obtained from sunflower husk: A potential material for environmental remediation. J Mol Struct. 2021;1239:130509.
27. Bodirlău R, Teacă C-A, Spiridon I. Influence of natural fillers on the properties of starch-based biocomposite films. Compos B Eng. 2013;44:575–83.