



Investigation of Mechanical Performance and Environmental Degradation of PLA-based Hybrid Composites Containing Natural and Carbonized Fillers

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ABSTRACT

This research analyzed the manufacturing and performance of polylactic acid (PLA) hybrid composites reinforced by rice husk (RH) and carbonized rice husk (RH carbon). Polylactic acid was mixed with thermoplastics such as ABS, PP, and PS for mechanical enhancements, while the hybrid fillers improved interfacial adhesion, thermal stability, and biodegradability. Using these raw materials, 10 composite formulations were polymerized by twin-screw extruder and compression molding, which were then subjected to thorough mechanical, thermal, microstructural, moisture absorption, and biodegradation behavior studies. Tensile, flexural, impact, and compressive properties were all at their highest for the optimized RH carbon composites, particularly Sample 8. According to TGA, the thermal stability improved in the presence of RH carbon; DSC passed evidence for increased crystallinity and thermal transitions. The SEM revealed strong filler-matrix adhesion with a lesser number of voids. Water absorption and soil burial tests ensured environmental instability of the composites. Increased RH carbon content diminished moisture absorption and biodegradation. The results emphasize the RH-RH carbon hybrid fillers to yield environmentally friendly composites for structural and biodegradable applications.

Keywords: Biopolymers; Blends; Polylactic acid-based composites; Carbonized rice husk; Environmental degradation.

1. INTRODUCTION

Biodegradable composites have caught great attention and focus due to the increased demand for materials that ensure sustainability and environment-friendly alternatives to petroleum-based polymers. Of the group of biodegradable polymers, polylactic acid (PLA) gets considerable attention for its renewable origin, biocompatibility, degradability, and composability. The major drawback to its wider industrial application lies in its brittleness, low-impact strength, and moisture sensitivity. The other thermoplastics (like acrylonitrile butadiene styrene (ABS), polypropylene (PP), and polystyrene) are studied to form blends with PLA to impart modifications in mechanical properties and processability. Natural fillers improve the mechanical property, allow cheap production, and encourage biodegradability of polymers. Rice husk (RH), being one of the most common agricultural wastes with a relatively high amount of cellulose, hemicellulose, and lignin, falls under the category of potential bio-fillers. Yet, its hydrophilicity and hence poor adhesion with hydrophobic polymer matrices may hamper mechanical stability. Thus, carbonized rice husk (RH carbon) has been utilized as an additional filler. The porous structure, thermal stability, and moisture resistance of RH carbon complement those of raw RH, promising a synergistic balance between biodegradability and durability. The

addition of two types of fillers namely, steel and hybrid to PLA materials can greatly improve the mechanical properties of the composites. For example, testing showed that wheat-cellulose fiber and vermiculite act synergistically to increase the elastic modulus by about 137% and the flexural modulus by more than 127% in PLA matrices, giving a good perspective regarding the strength enhancement of bio-composites through such hybrid reinforcements (Ariturk *et al.* 2024). At the same time, agricultural waste, like, horse gram powder, gave a 11.19% increase in tensile strength at optimum filler loading, establishing the importance of cheap natural fillers toward mechanical enhancement (Nataraj and Babu, 2024). Hybridization of PLA composites with continuous carbon/glass fibres exhibited 13% enhancement in tensile strength as compared to composites reinforced by single carbon fibers, thus affirming the hybrid reinforcement strategy in enhancing mechanical properties (Chen *et al.* 2024). From another viewpoint, real-world environmental degradation mechanisms hamper their dispersion under *in situ* conditions. Climatic aging was shown by (Vašíček *et al.* 2023) to compromise the mechanical properties progressively in PLA composites reinforced with natural fillers, thereby testing for long-term durable behavior. The use of waste fillers makes reinforcement simultaneously attractive from the mechanical and sustainability points of view, thus reducing dependency

on virgin raw materials with an associated positive effect on the environment. Conversely, the observed balance between mechanical enhancement and environmental stability emphasizes the need for a further investigation into the degradation mechanisms of such hybrid composites under different environmental conditions. The main degradation mechanisms facing PLA composites result from hydrolytic- and biodegradation-type mechanisms. Water diffuses into the polymer matrix to initiate hydrolysis of the ester link under both aerobic and anaerobic circumstances. Also, plant-based fibers can also accelerate hydrolysis by aiding water absorption and microbial colonization into the polymer matrix. In general, such fillers severely increase the degradation rate (Momeni *et al.* 2023). Synthetically speaking, PLA composites with renewable and biodegradable raw materials give good promise. Their biodegradation rate of about 90% within 12 weeks in composting environments marks them as potential candidates for more environmentally friendly disposal methods (Juan and Zhang, 2013). Developing green composites, therefore, has also led to improvements in waterproof and corrosion resistance properties by using bamboo and wheat straw as main materials, thus allowing their use in eco-friendly product design (Huang *et al.* 2022). In particular, compatibilized PLA/ABS blends may experience a toughening of their properties, including thermal resistance with SEBS-g-MA (styrene-ethylene-butylene-styrene grafted with maleic anhydride). A different study found that SEBS-g-MA increases the impact strength of the blends, alongside HDT and thermal stability. At a ratio composition of 60/30/10 wt% PLA/ABS/SEBS-g-MA, the blends have been proven to show better mechanical properties than neat PLA, thus supporting the application of compatibilizers for improving phase compatibility and interfacial adhesion (Costa *et al.* 2023). On the other hand, PLA/PLAE blends (where PLA is toughened with PLA-based thermoplastic elastomer, PLAE) have shown immense mechanical properties, with tensile strength measuring 70 MPa and elongation at break exceeding 140%. The ductility is aided by well-dispersed nanoscale elastomeric domains, which serve as energy-absorbing sites during deformation and thereby give rise to enhanced toughness without much compromise on stiffness (Cai *et al.* 2016). In the case of ABS composites, additional components such as polybutylene terephthalate (PBT) and multi-walled carbon nanotubes (MWCNTs) have been introduced for enhancing mechanical properties. Loading of 0.3 wt% MWCNTs in ABS/PBT hybrids brought about superior enhancement to tensile and flexural strength, indicating a synergy between nanoscale fillers and thermoplastic reinforcement for facilitating load transfer and stiffening (Farajian *et al.* 2022). Similarly, a significant enhancement of mechanical properties was found in polypropylene composites through the design of all-PP systems. Also, better tensile properties, especially in unidirectional laminate configurations were obtained on the β -nucleated random PP copolymer matrix

reinforced with α -PP homopolymer fibers. The main working mechanism lies in interfacial stress transfer, which distributes the load effectively and suppresses failure initiation, which is accounted for by the matrix-fiber interface (Abraham *et al.* 2009). Recently, hybrid composites with PLA, ABS, or PP are considered widely in structural, automotive, and packaging industries. Of these, PLA composites with reinforcement fibers of natural, man-made cellulose, and glass fibers exhibit better impact strength and crash resistance when compared with their counterparts based on PP composites. Very good improvements, in performance, were mostly caused by the cellulosic fibers that reinforce the composites coupled with the selective modification of the polymer-filler interphase, enabling an efficient stress transfer and hence interfacial bonding enhancement (Jaszkiewicz *et al.* 2013). These developments further suggest the feasibility of developing PLA as a biodegradable material alternative to petrochemical-based materials, especially in highly impact-resistant applications. Mechanical properties of PLA/ABS composites were further enhanced by the use of Short Carbon Fibres (SCFs), particularly in components fabricated using 3D-printing processes. Incorporation of SCF increased the hardness and impact strength while the tribological properties measured in terms of specific wear rate were found to both increase and decrease in varying measures. The wear mechanisms observed in these composites were ploughing, micro-cutting, and delamination, all of which were affected by the dispersion and alignment of SCFs within the matrix (Suresha *et al.* 2023). These results indicate the intricacy involved in reinforcing fiber-based hybrid systems wherein the mechanical and wear properties may not improve congruently.

Mechanically, rice husk and biochar addition have been investigated widely in PLA composites for performance enhancement and environmental sustainability (Zhao *et al.* 2024). The incorporation of biochar can help enhance the tensile strength. Low-loading incorporation of biochar is considered the best possible reinforcement method, since composites with 5 wt.% biochar loading have achieved tensile strengths up to 36 MPa, indicating good load transfer, and improved filler-matrix interaction (Anerao *et al.* 2023). The impact resistance modifications are, however, dependent on the incorporation amount of biochar. At 1 wt.%, the impact strength improves due to fine dispersion and energy absorption, while at elevated loadings, there is agglomeration that results in decreased toughness (Anerao *et al.* 2023). Similarly, flexural strength is greatly influenced through biochar concentration with values reaching a maximum at 3 wt.% and, thereafter, decreasing gradually due to filler agglomeration and inferior dispersion. Hot compression molding was an effective fabrication method for rice husk reinforced PLA composites, as it enhanced tensile strength and hardness through improved filler compaction and uniform

distribution within the composite matrix (Kumar *et al.* 2024). Additionally, dynamic mechanical analysis in a similar system (rice husk biochar/high-density polyethylene composites), would enhance viscoelastic properties, and hence PLA composites can also benefit the same under similar conditions (Zhang *et al.* 2017). Structurally, the SEM evidences suggested that the consistency in the distribution of rice husk and biochar in the PLA matrix is important in ensuring uniform mechanical behavior. Better dispersion means better stress transfer, and agglomerated regions become weak points (Kumar *et al.* 2024; Zhang *et al.* 2017). Even if these were promising renunciations supporting the use of biofillers in reinforcing PLA composites, issues of filler aggregation, interface adhesion, and processing-induced degradation prevent industrial implementation of PLA composites. Hence, more research is needed in filler loading, surface treatment, and processing for achieving a reasonable compromise between maximum mechanical enhancement and least impact on processability and environmental stability.

2. MATERIALS AND METHODS

In the research work, a multi-component hybrid composite was fabricated by utilizing biodegradable and thermoplastic polymers with natural and carbonized agro-waste fillers. The main polymer matrix, polylactic acid (PLA, Grade 3052D), was obtained from India Industries Pvt. Ltd., Chennai, India. As a pellet, PLA was pre-dried at 60°C for 6 hours in a hot-air furnace to remove moisture that would otherwise lead to hydrolytic degradation on processing. Acrylonitrile Butadiene Styrene (ABS), as a granule, was obtained from Aadila Lakshmi Plastic Traders Pvt. Ltd., Hyderabad, India. Polypropylene (PP, Grade PETOPLN EH 251) was obtained from PETKIM Petrokimya Holding A.S., Turkey, and Polystyrene (PS, SABIC®125PS) was obtained from SABIC, Kingdom of Saudi Arabia. Both PP and PS were in powder form.

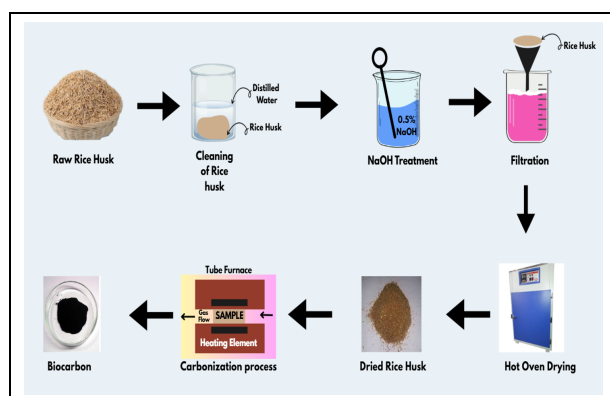


Fig. 1: Sequence of method followed for the preparation of biocarbon

This study utilises rice husk (RH) and its carbonized equivalent (RH Carbon) as hybrid fillers due

to their similar composition and potential to boost composite performance. The usage of both forms of the identical raw material offers compatibility and enhanced interfacial adhesion to the PLA matrix, owing to their comparable functional groups and structural properties. The compatibility offers greater mechanical interlocking, thereby enhancing the composite's overall toughness and lifespan. Through the synergistic interaction of the two fillers, the addition of untreated rice husk which is made up of lignocellulosic components and biocarbon that lowers water absorption and improves thermal stability significantly enriches the composite. The rice husk was acquired locally from a rice mill and comminuted into a fine powder for utilisation. The rice husk received an alkali treatment with sodium hydroxide (NaOH), which also increased the interfacial adhesion between the filler and the polymer matrix. By making the fibres more wettable and rougher on the surface, the treatment reduces surface elements including lignin, hemicellulose, waxes, and oils that might prevent adherence. Before soaking in a 0.5% NaOH solution for four hours, the rice husk was first cleaned in distilled water as part of the treatment process. The technique entailed drying the husk at 40°C for 6 hours to remove moisture and firm its structure. Biocarbon (RH Carbon) was generated by carbonizing dry rice husk powder in a furnace as illustrated in Fig. 1. Table 1 illustrates the physical and mechanical characteristics of various materials used in this study.

Table 1. Physical and mechanical characteristics of PLA, PP, PS and ABS

Property	PLA	ABS	PP	PS
Density (PLA)	1.25 g/cm ³ (21.5 °C),	1.05 g/cm ³ (21.5 °C),		1.05 g/cm ³
Melt flow index (MFI)	(210 °C/2.16 kg)	21.3 g/10 min (220 °C/10 kg)	24 g/10 min (230 °C/2.16 Kg)	7 g/10 min (200°C/5 kg)

2.1 Blend Preparation

All the polymers were pre-dried at 60°C for 24 hours in an air-circulated oven to remove any trace of moisture. The blend formulations were prepared through predetermined weight percent of PLA, ABS, PP, PS, RH, and RH Carbon, as shown in Table 2. Blending was carried out through a NEO PLAST HIGH TORQUE ZV20 counter-rotating twin screw extruder (TSW). Barrel temperatures were controlled through a controlled gradient: 160°C at the feed zone and increased to 190°C at the die head. A screw speed of 60 rpm was set to ensure proper mixing. The composite blends were then molded into standard test specimens after the extrusion process through NEOPLAST semi-compression molding equipment (Gujarat, India) with water cooling, and with a pressure of 196 bar. The molded samples were cured under room conditions for a minimum of 46 hours before testing.

Table 2. Blend composition

Sample No.	PLA (wt.%)	ABS (wt.%)	PP (wt.%)	PS (wt.%)	RH (wt.%)	RH Carbon (wt.%)
1	28	22	10	10	15	15
2	25	25	12	10	15	13
3	30	20	12	8	10	20
4	28	25	10	10	10	17
5	25	22	15	8	15	15
6	30	25	10	8	7	20
7	28	20	15	10	12	15
8	30	22	10	10	5	23
9	28	25	12	8	10	17
10	25	20	15	10	10	20

2.2 Mechanical Characterization

2.2.1 Tensile Test

Tensile testing was performed according to ASTM D638 on U-shaped dumbbell specimens of size $160 \times 13 \times 3 \text{ mm}^3$. The tests were conducted on an Instron 3369 universal testing machine at a crosshead rate of 5 mm/min and room temperature of 25°C. The data point is the mean of five individual samples.

2.2.2 Compression Testing

Compression testing was in accordance with ASTM D695-10 with specimens of $25 \times 25 \times 10 \text{ mm}^3$. A 50 kN load cell universal testing machine was run at a crosshead speed of 1 mm/min. Three samples of each composition were run.

2.2.3 Flexural Test

Flexural strength was determined by ASTM D790 on $80 \times 13 \text{ mm}^2$ test specimens in a 3-point bending fixture with a 50 mm support span and at a 1.4 mm/min loading rate.

2.2.4 Impact Test

The impact resistance was evaluated in accordance with ASTM D256 utilizing notched test specimens ($63.5 \times 12.7 \times 3 \text{ mm}^3$). A V-notch of 2.54 mm was created, and fracture energy absorbed was determined utilizing an impactometer.

2.3 Thermal Analysis

2.3.1 Thermogravimetric Analysis (TGA)

Thermal stability was measured using a Pyris-7 TGA. 5–10 mg samples were scanned from 20°C to 900°C at 10°C/min under a nitrogen purge (20 mL/min). Degradation profile was recorded to determine onset and peak degradation temperatures.

2.3.2 Differential Scanning Calorimetry (DSC)

Differential Scanning Calorimetry was performed to determine the thermal transitions, including glass transition temperature, crystallization temperature, and melting temperature of the hybrid composites. The measurements were carried out using a DSC 8000 system (PerkinElmer, USA) under a nitrogen environment to prevent oxidation.

For each test, about 6–8 mg of the composite was sealed in an aluminum pan. The samples were heated from 30°C to 250°C at a rate of 10°C/min, then cooled to 30°C, followed by a second heating cycle to eliminate thermal history. The thermal transition temperatures and enthalpies were recorded from the second heating cycle, and the data were used to assess the influence of filler composition on the crystallinity and thermal behavior of the composites.

2.4 Microstructure Analysis

2.4.1 Scanning Electron Microscopy (SEM)

The SEM analysis was carried out with a Hitachi S-3400 N in order to examine the fracture surfaces. Charging was avoided by gold-coating the samples and they were scanned at an accelerating voltage of 20 kV to look for matrix-filler interaction and morphology. X-Ray Diffraction (XRD) was carried out on formed samples using a Rigaku MiniFlex 600 diffractometer equipped with Cu K α radiation ($\lambda = 1.54060 \text{ \AA}$). Scanning was in the range of 10°–90° (2 θ) with a step size of 0.111°/s for high-resolution measurement of crystallinity.

The water absorption (WA) behavior of these hybrid composites of PLA/ABS/PP/PS was studied by submerging oven-dried specimens in plain water, distilled water, and 5% NaCl solution for 24 hours at room temperature. The percentages of water absorbed were recorded periodically by weighing the specimens and using the standard formula:

$$WA(\%) = \frac{w_t - w_0}{w_0} \times 100 \quad \dots (1),$$

where, w_t and w_0 initial and final weights, respectively. The test gave an indication of how the composites react with respect to hydrophilicity and diffusion behaviors in different aqueous environments.

2.5 Soil Burial Degradation Test

In analyzing the biodegradability of samples, the molded sample was subjected to soil burial tests for 60 days. The samples were buried at a depth of 10 cm in natural garden soil at ambient temperature conditions (temperature: $25 \pm 2^\circ\text{C}$ relative humidity: 60-70%). To promote microbial growth and imitate normal

environmental conditions, the soil was kept moist. After 60 days of burial, retrieval of samples was done from the soil, washing the samples gently with water to bring off soil particles, and air-drying for subsequent testing. Tensile strength for each sample was measured prior to burial (0-day) and after soil exposure for 60 days. Three specimens were tested and the average was reported for each sample under each condition.

3. RESULTS AND DISCUSSIONS

3.1 Mechanical Characterization

Fig. 2a shows that tensile performance of hybrid PLA/ABS/PP/PS composites with different percentages of rice husk (RH) and carbonized rice husk (RH carbon) differed considerably in the ten different formulations. Experimental tensile strength values varied from about 67 MPa and 88 MPa and showed that adding bio-based fillers to the polymer matrix can significantly improve or degrade tensile performance depending on their amount and morphology. The highest tensile strength value was obtained for Sample 8, 88 MPa, due to its highly optimized composition with high RH carbon content (23%) and low RH content (5%). The excellent tensile performance can be attributed to the efficient stress transfer due to the increased interfacial adhesion between RH carbon and the polymer matrix. RH carbon with its rigid, porous microstructure and high surface area permits efficient mechanical interlocking in the PLA matrix and thus improves its tensile load-carrying capacity. Conversely, the lowest tensile strength value, about 67 MPa, was obtained for Sample 2 due to its relatively high untreated RH content of 15% and low RH carbon ratio. Untreated RH with lignocellulosic and hydrophilic nature may be contaminated with surface impurities and have poor interfacial bonding, resulting in fiber pull-out or void formation under tensile loading. Intermediate tensile performance was obtained for Samples 3, 5, and 9, with balanced content of RH and RH carbon. These samples yielded medium to high strength values (of approximately 74–80 MPa), suggesting that synergistic action of raw and carbonized fillers can be harnessed if in equilibrium.

The measured values of flexural strength are from 115 MPa to 165 MPa (Fig. 2b), clearly showing the strong impact of filler content and its dispersion on the resistance of the composite to deformation under bending loading. Of all the tested samples, Sample 8 showed the best flexural strength (~165 MPa) followed by Sample 6 with approximately 150 MPa. The two high values can be easily explained by their optimized hybrid filler content, i.e., high content of RH carbon (23% and 20%, respectively) and low fraction of raw RH. The fillers in the form of RH carbon increase the stiffness and modulus of the composite due to its graphitized structure, thermal stability, and high compatibility with the polymer matrix. In addition to that, the enhancement of RH carbon

dispersion in the matrix and the elimination of lignocellulosic contaminations allow for a continuous and strong stress path under flexural loading. In contrast, Sample 2 showed the lowest flexural strength, approximately 115 MPa, in accordance with its higher fraction of raw RH and low content of RH carbon. The addition of untreated RH, being more hydrophilic and less thermally stable, possibly leads to poor fiber matrix adhesion, microvoid formation, and local stress concentrations during bending. The limitation prevents the composite from evenly dispersing loads, thus reducing its resistance to flexural failure. Reasonable flexural strength values for Samples 3, 4, and 10 were in the 130–140 MPa range. These samples contained a balanced relative ratio of RH and RH carbon, which suggested that an optimal hybrid filler synergy can be attained without undue reliance on either. Synergy between raw RH imparting some degree of fiber stiffness and RH carbon imparting structural rigidity and heat resistance is significant in determining overall bending performance.

Impact strength values for all ten samples lay between the approximate range of 5.9 and 7.0 kJ/m², thus indicating fairly favorable toughness profiles for the composites prepared (Fig. 2c). These variations are directly dependent on the filler types and their proportions, particularly on the relative content of raw rice husk (RH) and carbonized rice husk (RH carbon), and also on the thermoplastic matrix compositions involved. Sample 10 had the best impact strength of about 7.0 kJ/m². Among the reasons that justify it are the neat matrix combination with higher PP and ABS (15% and 20%, respectively) with 20% RH carbon, which could have promoted energy absorption during the fracture. Both ABS and PP are ductile polymers and capable of resisting dynamic loads, whereas RH carbon, being porous and stiff, probably initiated crack deflection and provided microstructural reinforcement without overly compromising flexibility. On the other side, Sample 3 displayed the lowest impact strength (~5.9 kJ/m²), very likely due to a lower content of ductile polymer combined with an unsuitable filler layout. RH carbon helps maintain rigidity and mechanical integrity, but excessive filling or poor dispersion can cause localized stress concentrations that lead to premature fractures upon impact. Intermediate impact strength levels were found for Samples 5, 6, and 9, with values between 6.3 and 6.6 kJ/m². These formulations show a more synergistic interaction of matrix flexibility with filler-induced rigidity and thus point toward the existence of an optimal filler-to-polymer ratio for fracture resistance.

The Shore D (Fig. 2d) hardness ranged from 78 to 89, with Samples 6, 8, and 10 having the higher hardness values. These samples had a high RH carbon content and moderate to high PP content, thereby indicating surface rigidity and abrasion resistance to be

reciprocally dependent on filler hardness and its structural properties. Being thermally stable and graphitic in nature, the RH carbon endowed a hard surface finish onto the composites. However, Sample 1 with low RH carbon and increased RH showed the lowest hardness value of about 78; this further confirms the role of RH carbon in imparting higher surface mechanical resistance.

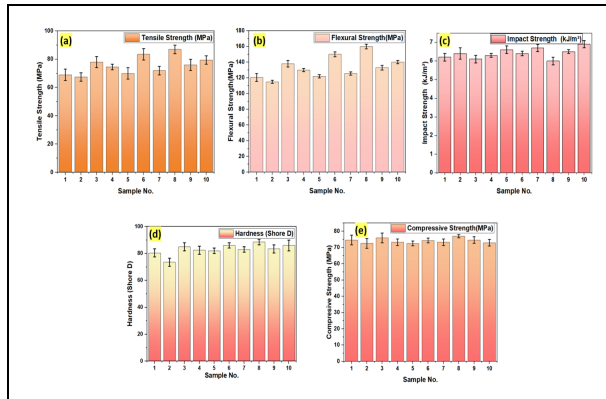


Fig. 2: Mechanical properties of the composites

Compressive strength results as presented in Fig. 2 (e) showed values between 67 and 76 MPa. Sample 8 exhibited the highest strength, around 76 MPa, also in good agreement with samples that gave the best response in other testing regimes. The large RH carbon content increased the density of the structure and thus decreased deformation of the matrix under compression. With higher RH content, composites showed more deformation, probably due to void formation in the matrix or weak filler-polymer interface under compressive strain. Samples 6 and 10 also gave the best performances, confirming the importance of an optimized balance of hybrid fillers.

The water absorption of the hybrid composites is represented in Fig. 3(a-j). Each of the 10 different formulations shows a distinct uptake profile after being immersed for 24 hours in plain water, distilled water, and in an NaCl solution. Through all tested samples, water absorption follows the trend: NaCl solution > plain water > distilled water because of the osmotic pressure exerted by NaCl that accelerates moisture ingress, while distilled water showed the least absorbance due to the absence of ions. With higher amounts of raw rice husk content, samples absorbed higher quantities of water, gaining peak values of as much as ~0.23%, owing to the hydrophilic nature of lignocellulosic fibers and higher porosity. On the other hand, absorption levels went down significantly in samples having more RH carbon content (such as samples 6, 8, 10), with final values less than 0.15%, because of the hydrophobic nature and thermal stability of biocarbon and the compact structure thereof, which restricts water penetration. Water absorption levels for all samples increased rapidly during the first 6-10

hours. Following this rapid phase, the water absorption rate started to slowly plateau as samples approached equilibrium. Such a behavior is indicative of Fickian-type diffusion kinetics, which are further influenced by the compatibility between matrix and filler and the dispersion of the filler. The study shows that, overall, RH carbon is key to improving the moisture resistance of biocomposites, while high RH content must be controlled with caution to avoid dimensional instability in humid or aqueous environments.

3.2 Biodegradation Test

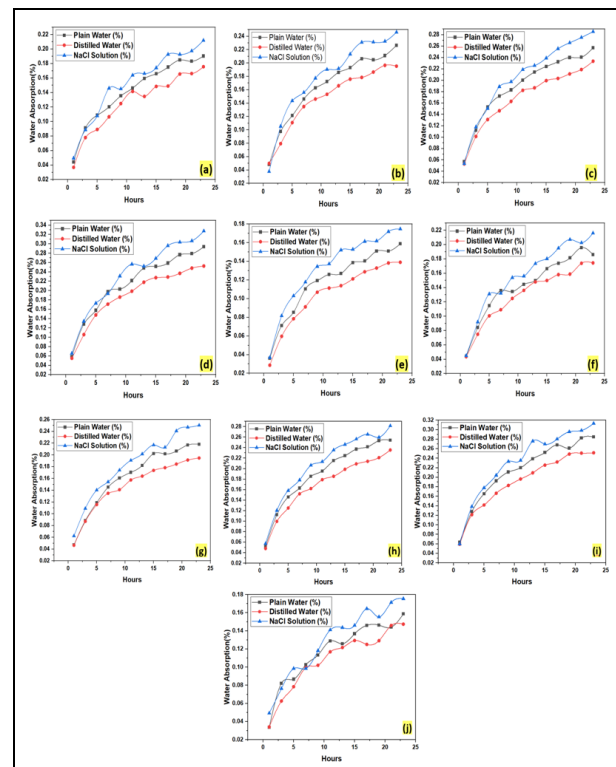


Fig. 3: Absorption rate of samples

Generally, the samples showed a steady weight loss with time as per the normal biodegradation trend: initial rapid degradation followed by a relatively slow steady-state disappearance after about 60 days (figure 4b). The trend mainly denotes the initial hydrolysis of plainly PLA amorphous regions and the microbial degradation of lignocellulosic rice husk in the early period, followed by the diffusion limitations and surface saturation afterwards. Among all the formulations, Sample 2 with the highest RH loading (15%) and a lower amount of RHC (13%) barely stood the most significant degradation of about 3.7% weight loss in 60 days. The highly-loaded RH being hydrophilic and biodegradable would retain moisture and keep the glowing microbial activity in favor of fast degradation.

Meanwhile, Sample 8 that had the lowest RH(5%) and the highest RHC (23%) level presented the

least degradation (~2.13%), proving the moisture-barrier character and thermal stability afforded by RHC. The carbonized filler restricted water absorption and the microorganism's entry, thereby promoting the composite's durability. Other samples such as 3, 4, 6, and 10, with moderate to well-balanced RHC content (17–20%), had intermediate degradation values between 2.5–3.0%, indicating that though only low amounts of RH were required to initiate biodegradation, the presence of RHC was an effective barrier to the progression of degradation.

The tensile-strength (Fig. 4 a) results show a consistent decrease in tensile strength for all the samples following soil exposure, with the reductions ranging somewhere between 12% and 16%. Samples with a higher RH content, namely Samples 2, 5, 7, and 9, witnessed higher tensile-strength reductions (up to around 16%), owing to the fact that lignocellulosic RH is more hydrophilic and biodegradable. The moisture uptake during burial probably weakened the fiber–matrix interface, induced microcracking, and expedited polymer chain scission. On the other hand, those samples containing a higher proportion of RH carbon, namely Samples 3, 6, and 8, showed comparatively lesser strength degradation (~12–13%), owing to the carbonized filler being protective, hydrophobic, and thermally stable against hydrolytic attack and thus preserving its integrity. For instance, having the highest 0-day UTS of 87 MPa, Sample 8 managed to hold on to a strength near 76.6 MPa even after 60 days, which reflected only a 12% loss. Conversely, Sample 2 was subjected to a drop from 67.5 MPa down to 56.4 MPa, being more susceptible at another RHC-RH ratio than likely the higher one, emphasizing the effects of filler composition on long-term durability and resistance to mechanical degradation under nature-simulating conditions.

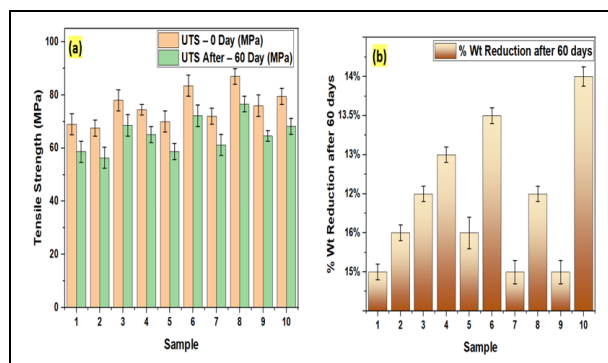


Fig. 4: Biodegradation results

3.3 Thermal Behaviour

The TGA profiles of the ten composite samples show a characteristic two-stage degradation pattern, characteristic of hybrid polymer composites containing both thermoplastics as well as bio-fillers illustrated in

Fig. 5. The first stage of weight loss, occurring at around 260–370°C, is caused by the degradation of low thermal stability components like hemicellulose and amorphous cellulose contained in rice husk fillers. The peak degradation—seen in the magnified inset of the TGA plot—occurs between 300–360°C, and is caused by the thermal degradation of the PLA/ABS/PP/PS matrix along with char formation. Among all the samples tested, Sample 8 has the highest thermal stability as supported by the delayed onset of decomposition and lower mass loss in the initial stage. This high thermal resistance is most probably caused by the high content of carbonized rice husk (biochar), which is thermally resistant and promotes char formation, thus protecting the matrix from early degradation. Sample 1 has the lowest thermal stability since it starts decomposing at the lowest temperature and has the lowest residue on decomposition, which reflects a lower biochar content and higher content of thermally labile materials. The residual mass at about 800°C is variable in samples, and the higher values are for composites with higher biochar proportion. This observation supports the hypothesis that biochar is not only a thermal insulant but also a thermally stable residue. Overall, the use of rice husk and carbonized rice husk as hybrid fillers has a remarkable influence on the thermal degradation process, and increased content of the carbonized filler increases the thermal resistance and stability of the composite.

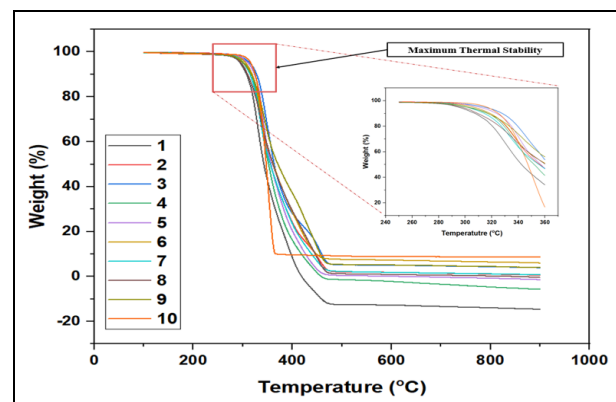


Fig. 5: TGA test results

3.4 DSC Analysis

Differential Scanning Calorimetry thermograms of Samples 1–10, as depicted in Fig. 6, show clear thermal transitions, such as glass transition (T_g), cold crystallization (T_{cc}), and melting temperature (T_m), important for determining the thermal behavior of PLA-based hybrid composites. In the series of samples, glass transition temperature (T_g) was found in the range 55–65°C, indicating the beginning of segmental mobility of the PLA matrix. Sample 6 and Sample 10 showed slightly elevated T_g values, indicating increased filler–matrix interactions that inhibit chain mobility due to rice husk biochar addition. Cold crystallization (T_{cc}) peaks were

seen in the range 100–140°C with varying intensity and position. Samples 3, 6, and 9 showed broader and intense T_{cc} peaks, indicating higher chain rearrangement on heating. This can be explained as an outcome of hybrid fillers having an effect to promote heterogeneous nucleation. The melting endotherm (T_m) was seen consistently between 150–165°C in all the samples. Sample 9 and 10, however, show sharper and higher T_m peaks, indicating higher crystalline order and thermal resistance due to optimal filler dispersion. Overall, the DSC data indicate that the addition of hybrid fillers (rice husk and its biochar) significantly impacts thermal transitions. Increased crystallinity and stable T_g values in some samples indicate higher thermal stability, higher chain packing, and potential applicability to thermally demanding applications.

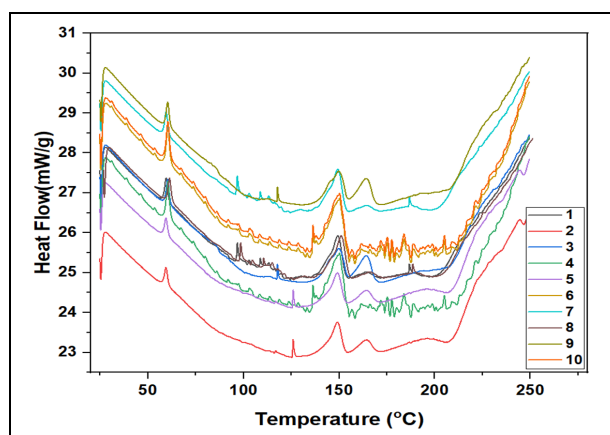


Fig. 6: DSC analysis

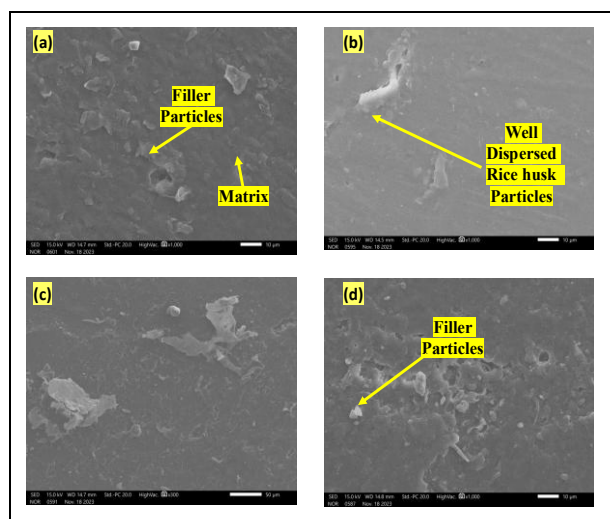


Fig. 7: Microstructural analysis

3.5 Microstructural Analysis

SEM micrographs (Fig. 7 a–d) reveal the morphology of the surface and interfacial characteristics of the PLA-based bio-filler-reinforced hybrid composites. In all the instances, the filler particles are

well bonded to the PLA matrix, as indicated by the lack of any gaps or pull-outs in the filler areas. Image (a) clearly reveals the uniform dispersion of filler particles, which reflects good compatibility and interfacial adhesion within the matrix. Likewise, image (b) indicates good bonding of the biomass fillers with the matrix with no appreciable detachment or debonding. The photos (c) and (d) also validate this observation and reveal that the filler particles are dispersed well in the PLA matrix with hardly any microvoids present. The microvoids are negligible and do not affect the overall structural strength of the composite material. The uniform bonding and even distribution of the fillers in all the samples indicate that the processing conditions were optimally suitable, which enhanced the interaction between the polymer and bio-filler phases. This is the cause of the improved mechanical and thermal stability of the composite system.

4. CONCLUSION

This study systematically evaluated the mechanical, thermal, microstructural, water absorption, and environmental degradation behaviour of PLA based hybrid composites reinforced with rice husk and carbonised rice husk. The hybrid incorporation significantly improved composite performance, with RH providing mechanical reinforcement and RH carbon enhancing thermal stability and reducing hydrophilicity. Thermal analyses confirmed higher degradation onset temperatures and increased crystallinity with greater rice husk carbon content, while SEM revealed uniform filler dispersion and strong filler matrix bonding. Increased RH carbon reduced water absorption and slowed environmental degradation, whereas higher RH content accelerated weight loss. After 60 days, tensile strength declined by 10 to 20 %, indicating controlled degradability with retained structural integrity. Overall, the use of agro waste and biochar supports sustainable material development, making these composites suitable for semi structural biodegradable applications in automotive, packaging, agricultural, and consumer products.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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