

Article

Performance Enhancement of PLA Hybrid Biocomposites Using Flax Fiber and Agricultural Waste Biofillers: A Comparative Study with Jute-Based Systems Supported by Fuzzy CRITIC–COPRAS Analysis

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Abstract

The development of high-performance, sustainable biocomposites requires biodegradable matrices and optimized natural reinforcements. In this study, flax fiber-reinforced polylactic acid (PLA) hybrid biocomposites incorporating waste pistachio nut shells (WPNS), waste tea leaf fiber (WTLF), and waste quail eggshell (WQES) were developed and evaluated, with direct comparison to previously reported jute-based hybrid systems to assess the benefits of fiber substitution. The composites were fabricated via compression molding and characterized for their mechanical, thermal, acoustic, surface, and moisture-related properties. Replacing the jute with flax resulted in a consistent performance enhancement. Among the hybrids, the flax–WPNS composite exhibited the highest tensile and flexural performance, achieving tensile strength improvements of approximately 30–40% over neat PLA due to effective stress transfer and crack deflection. The flax–WTLF composite showed superior acoustic behavior, attaining a maximum sound absorption coefficient of approximately 0.65–0.70 at mid-to-high frequencies, attributed to its porous microstructure. In contrast, the flax–WQES composite demonstrated the highest thermal conductivity (0.54 W/(mK)) and apparent density (2.24 g/cm³), reflecting dense packing and the presence of CaCO₃-rich particles. Scanning electron microscopy revealed distinct microstructural mechanisms governing these property-specific responses, including differences in interfacial bonding, void distribution, and filler packing efficiency. An integrated fuzzy CRITIC–COPRAS multicriteria decision-making approach identified the flax–WPNS hybrid as the optimal overall formulation. The results clearly demonstrate that flax fibers outperform jute as a reinforcement in PLA-based hybrid biocomposites, and that targeted combinations of flax and waste-derived fillers enable multifunctional performance optimization for sustainable engineering applications.

Keywords: flax fiber; polylactic acid (PLA); waste-derived fillers; mechanical performance; thermal and acoustic properties; CRITIC–COPRAS



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1. Introduction

Biodegradable polymers have become increasingly important as the demand for environmentally friendly materials in the manufacturing and consumer sectors has increased. Among these materials, polylactic acid (PLA) has emerged as a leading material owing to its renewability and biocompatibility [1–3]. It offers good tensile stiffness, although its relatively low toughness, limited impact resistance, and slow crystallization rate restrict its functionality in structural and semi-structural engineering applications. These deficits have led researchers to progress in the area of reinforcing PLA with natural fibers and sustainable fillers to enhance its mechanical, acoustic, and thermal performance while maintaining biodegradability. Natural fibers are well-known for their abundance, low cost, biodegradability, renewability, and favorable strength-to-weight ratios [4–6]. Their incorporation into PLA aligns with global sustainability goals and supports the development of eco-friendly composite materials.

Among the available natural fibers, flax has been ranked as one of the most efficient reinforcements for PLA [7–11] owing to its high cellulose content, low microfibril angle, and intrinsic stiffness, which translate into excellent load-bearing capabilities [12,13]. Numerous studies have highlighted the exceptional reinforcing potential of flax in various matrices. For instance, recent research [14] reported that continuous flax/PLA laminates achieved tensile strengths up to 179 MPa and elastic moduli of 19.5 GPa. In contrast, 3D-printed flax/PLA composites [15] achieved a tensile strength of 253.7 MPa and a modulus of 23.3 GPa at approximately 30% fiber volume, which are significantly higher than those of neat PLA. Similarly, increasing the flax fiber content from 7.9 to 17.6 wt. % increased the impact energy absorption to approximately 25–30 J [16]. In addition, chemical and surface treatments further enhanced the flexural strength by 17% and interlaminar shear strength by 29% [17]. These improvements underscore the suitability of flax-reinforced PLA as a high-performance biodegradable composite.

In addition to natural fibers, agricultural waste fillers in the form of nut waste, leaf waste, and poultry waste, such as shells and feathers, have emerged as the most suitable and sustainable reinforcements for enhancing composite performance. Among these fillers, particularly in the nut waste category, waste pistachio nut shell filler (WPNS) [18–21] is promising because of its lignocellulosic composition and high hardness. In previous studies, WPNS-based composites have shown remarkable improvements across various polymer matrices. For example, alkali-treated WPNS increased the tensile and flexural moduli and improved the thermal stability of PLA by approximately 15 °C at a 20 wt. % loading [22]. In polyester matrices, a 5 wt. % WPNS addition enhanced tensile strength by 96.6%, impact strength by 66.7%, and microhardness by 53% [23], while in epoxy hybrid systems, WPNS increased the fatigue life to 38,725 cycles [24]. These results confirm that WPNS is a robust and sustainable filler that significantly enhances matrix stiffness, hardness, and thermal performance [25,26]. Particle-size-dependent reinforcement has also been reported in poly(methyl methacrylate) (PMMA), where WPNS particles smaller than 150 µm yielded tensile strengths of up to 69 MPa [20]. These findings further confirm that WPNS is a robust and sustainable filler capable of significantly enhancing matrix stiffness, hardness, and thermal performance across different polymer systems [27,28].

In the leaf waste category, waste tea leaf fillers (WTLF) are the residues remaining after the extraction of industrial processing (withering, crushing, fermenting, drying) and are rich in cellulose, lignin, tannins, and phenolic constituents. The incorporation of spent tea leaf (STL) powder into natural-fiber-reinforced composites has been shown to significantly enhance mechanical performance. The addition of 10 g of STL to jute/cotton composites increased tensile strength by 33.46% and compressive strength by 38.86%, accompanied by improved crystallinity and thermal resistance [29]. In glass/kenaf epoxy hybrid systems, a

formulation containing 5 wt. % WTLF and 25 wt. % kenaf achieved the highest mechanical strength, while a higher WTLF content (25 wt. %) provided superior sound absorption due to enhanced porosity [30]. Similarly, incorporating 5 phr of ground tea leaves (GTL) into melt-spun PLA fibers improved tensile behavior and crystallinity, as evidenced by intensified XRD peaks at 16° and 21.8° , indicating filler-induced structural ordering [31]. These findings collectively demonstrate that WTLF can strongly influence both mechanical and functional properties of biocomposites.

In addition to agricultural waste, poultry waste, especially waste quail eggshell fillers (WQES), contains 94–97% CaCO_3 along with an organic membrane that improves interfacial bonding [32,33]. The incorporation of 5–10 wt. % WQES into epoxy matrices increased the tensile strength to 49.5–52.4 MPa, outperforming conventional limestone fillers [34]. Polyaniline/quail eggshell (PANI/QES) composites exhibited an ammonia detection limit of 5.24 ppm, demonstrating enhanced surface reactivity. Similarly, the incorporation of 9 wt. % eggshell powder into jute/epoxy composites increased tensile strength by 73.83%, flexural strength by 50.17%, and Shore D hardness by 21.43% [35]. Despite these mechanical advantages, the use of eggshell fillers in flax/PLA systems remains largely unexplored.

In our previous study [36], we demonstrated for the first time that PLA composites reinforced with jute fibers and filled with waste pistachio nut shells (WPNS), waste tea leaves (WTLF), and waste quail eggshells (WQES) exhibited significant improvements in tensile strength, acoustic damping, and flame resistance. Specifically, the WPNS-filled jute/PLA composites achieved the highest tensile strength (57.38 MPa), WTLF-filled composites achieved the highest sound absorption coefficient (0.643), and WQES-filled composites exhibited superior flame resistance and minimal water uptake (0.588%) owing to their CaCO_3 -rich composition. These findings establish the viability of multi-filler hybrid PLA systems and open avenues for enhancing their performance by replacing the primary reinforcement fibers.

Building on this foundation, the present study replaced jute with flax fiber, a stronger and stiffer bast fiber, to develop a new family of high-performance flax/PLA composites filled with the same three waste fillers (WPNS, WTLF, and WQES). The intention was to assess whether the inherently superior mechanical behavior of flax can synergize with WPNS, WTLF, and WQES to produce composites with enhanced strength, stiffness, and acoustic properties compared to their jute-based counterparts. As agricultural fillers differ in morphology and composition, with WPNS being lignocellulosic, WTLF being porous and phenolic-rich, and WQES being mineral-rich, flax-reinforced hybrids are expected to demonstrate reinforcement mechanisms different from those of jute/PLA systems.

However, the presence of multiple reinforcements and fillers complicates direct comparisons based solely on mechanical testing results. An optimal composite cannot be selected based on a single property because performance criteria such as strength, stiffness, density, water absorption, thermal stability, and acoustical behavior often conflict. To address this challenge, multi-criteria decision-making (MCDM) techniques have been increasingly applied in material research [37]. Methods such as the Analytic Hierarchy Process (AHP) [38], Viekriterijumsko KOMPromisno Rangiranje (VIKOR) [39], Technique for Order of Preference by Similarity to Ideal Solution (TOPSIS) [40], Weighted Aggregated Sum Product Assessment (WASPAS) [41], Multi-Attributive Border Approximation area Comparison (MABAC) [42], Step-wise Weight Assessment Ratio Analysis (SWARA) [43], and Complex Proportional Assessment (COPRAS) [42] have been used to rank composite formulations. Fuzzy-enhanced models further improve decision quality by accounting for uncertainty in experimental data and expert evaluations. Recent research [44] employed a Criteria Importance Through Inter-Criteria Correlation and Evaluation based on Distance from Average Solution (CRITIC-EDAS) hybrid model to optimize biofiller-vinyl ester com-

posites, identifying a formulation with 49.71 MPa tensile and 37.88 MPa flexural strengths as the optimal. Taguchi optimization has been shown to be effective in reducing wear by up to 89.91% in PLA hybrid composites [45]. Despite this progress, no study has yet applied the integrated fuzzy CRITIC–COPRAS approach to optimize flax/PLA composites filled with WPNS, WTLF, or WQES formulations used in the present [46].

The objective of this study was to develop and evaluate flax fiber–reinforced PLA hybrid biocomposites incorporating waste pistachio nut shells (WPNS), waste tea leaf fiber (WTLF), and waste quail eggshell (WQES) as sustainable biofillers. This study aimed to investigate the synergistic effects of flax fibers and different agricultural waste fillers on the mechanical, thermal, acoustic, and surface properties of PLA composites and to compare their performance with previously reported jute-based systems. Furthermore, an integrated fuzzy CRITIC–COPRAS multi-criteria decision-making approach was employed to identify the optimal composite formulation based on multiple performance attributes.

2. Materials and Methods

2.1. Materials

Extraction of Biofillers: Waste Pistachio Nut Shell (WPNS), Waste Tea Leaf (WTLF) and Waste Quail Eggshell (WQES) Particles

In the present study, two different agro-wastes, waste tea leaf residues (WTLF) and waste pistachio nutshells (WPNS), and one poultry waste biofiller, waste quail eggshells (WQES), were used as secondary reinforcements. Pistachio shells were collected from food-processing outlets in the Jammu and Kashmir regions, whereas tea leaf residues were obtained from the Ooty tea factory, Coimbatore, India. Quail eggshells were sourced from commercial poultry farms in Namakkal, India, and then cleaned for further processing. All these fillers inherently exhibit variations in their particle morphology, chemical constituents, and moisture levels owing to environmental factors, maturation stage, and prehandling practices.

To minimize this heterogeneity, all fillers were subjected to a standardized purification protocol, such as chemical pretreatment. The chemical modifications induced by alkali treatment, including the reduction of hemicellulose and lignin-related functional groups and the exposure of cellulose-rich hydroxyl sites, were previously confirmed by FTIR analysis and reported in our earlier study [36]. The same treatment protocol was used to ensure consistency and comparability. Initially, the materials were washed with a 5% NaOH solution to remove surface-bound oils, organic residues, and other contaminants. After thorough rinsing with deionized water, the treated biofillers were oven-dried at 100 °C for 12 h in a hot-air oven (Model: NSW-143, Narendra Scientific Works, New Delhi, India) to ensure complete moisture removal. A comprehensive schematic illustrating the preparation routes for WPNS, WTLF, and WQES and the development of the composites is shown in Figure 1.

2.1.1. Particle Size of the Biofillers

Particle size analysis of biofillers is essential because it directly influences the dispersion quality, interfacial bonding, and stress transfer within polymer matrices. Variations in particle size can significantly affect the mechanical, thermal, and functional performance of biocomposites. The WPNS and WTLF were pulverized using a planetary ball mill (Model: NST-2A, Insmart Systems, Hyderabad, India) to obtain controlled particle size fractions between 50 µm and 75 µm. This window size was intentionally chosen to promote homogeneous dispersion and improve the filler–matrix adhesion within the PLA. Literature reports [47–49] indicate that particles finer than 50 µm are prone to agglomeration, whereas those exceeding 75 µm may offer insufficient surface area for effective stress transfer; therefore, an intermediate distribution is preferred for optimum reinforcement performance.

After milling, the fillers were passed through a sieve stack to ensure uniformity and stored in a desiccator until composite fabrication was performed. In contrast to WPNS and WTLF, which were processed in the micrometer size range, WQES was subjected to intensive size reduction to obtain a substantially finer particulate system. Rather than being treated solely as a conventional micrometer-scale filler, the effective fineness and dispersion potential of WQES were quantitatively evaluated using the Brunauer–Emmett–Teller (BET) surface area (Model: BELSORP-MAX, MicrotracBEL Corp., Osaka, Japan) and Barrett–Joyner–Halenda (BJH) pore size analyses, as per ASTM D3663 [50]. The WQES exhibited a specific surface area of $1.51 \text{ m}^2/\text{g}$ and a mean pore diameter of 34.27 nm (Figure 2), confirming its mesoporous micro–nanoscale nature and high surface availability. This finer scale is attributed to the high calcium carbonate content (90–95%) of eggshells, which benefits from the reduced particle dimensions by enhancing the dispersion, interfacial contact area, and filler–matrix interactions in the PLA systems. Previous studies have similarly reported improved reinforcement efficiency and functional performance for finely divided eggshell-derived CaCO_3 fillers [51–54].



Figure 1. Isolation of waste biofillers and sequential steps in composite fabrication.

2.1.2. Flax Fiber Mat

The flax fibers used in this study were procured from Go Green Products (Coimbatore, Tamil Nadu, India) and supplied in mat form for composite fabrication. Initially, it was cleaned with distilled water to eliminate dust, surface impurities, and processing residues. The fibers were subsequently dried at 60 °C for 24 h and cut into uniform mats suitable for composite layup. Consistent with procedures widely reported in earlier research on natural fiber/PLA systems [14,17], alkaline surface modification was performed to enhance interfacial compatibility. The flax mats were immersed in 5 wt. % NaOH solution for 4 h,

thoroughly rinsed until neutral pH was achieved, and then oven-dried at 80 °C for 6 h. Raw flax fibers typically contain 65–75% cellulose, 12–18% hemicellulose, and 2–5% lignin, and exhibit relatively high moisture absorption owing to the hydrophilic functional groups present in hemicellulose and pectin [12]. Alkali treatment selectively removes portions of hemicellulose, lignin, waxes, and surface pectic substances, resulting in a 10–15% increase in cellulose exposure and the development of a microscopically roughened surface topology. This modification reduced the inherent hydrophilicity of flax, improved the wetting of the fiber by PLA, and promoted stronger interfacial adhesion by facilitating mechanical interlocking. Numerous studies [5,11,17] have demonstrated that NaOH-treated flax fibers produce higher tensile, flexural, and interlaminar shear strengths in PLA-based composites than untreated fibers, primarily because of the improved load transfer efficiency and minimized interfacial voids.

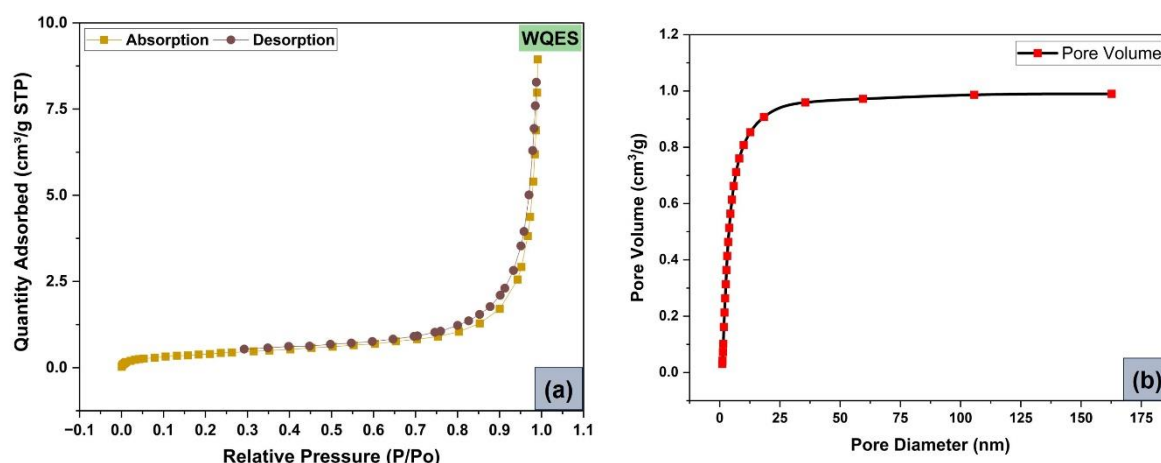


Figure 2. (a) Nitrogen adsorption–desorption isotherms and (b) BJH pore size distribution of waste quail eggshell (WQES).

2.1.3. Sample Composition Design and Justification

The formulation of the composite sets (Table 1) was designed to decouple the baseline reinforcing behavior of the waste biofillers from their synergistic interaction with flax fibers while remaining consistent with the processing constraints and prior optimization studies.

Table 1. Composite formulations used in the present study.

Sample ID	PLA (wt. %)	Flax (wt. %)	WPNS (wt. %)	WTLF (wt. %)	WQES (wt. %)
S1	100	—	—	—	—
S9	60	40	—	—	—
S3	60	—	40	—	—
S4	60	—	—	40	—
S5	60	—	—	—	40
S10	60	32.5	7.5	—	—
S11	60	32.5	—	7.5	—
S12	60	32.5	—	—	7.5

Composites S3–S5 (PLA + WPNS/WTLF/WQES) were prepared with a relatively high filler loading of 40 wt. % to deliberately amplify the intrinsic influence of each biofiller on the mechanical, thermal, and acoustic responses of the PLA matrix in the absence of fibrous reinforcement. At a low filler content (≤ 10 wt. %), the effects of particulate fillers in PLA are often marginal and difficult to distinguish from experimental scatter, particularly for lignocellulosic and mineral waste fillers. The 40 wt. % loading was therefore selected to establish a clear reference baseline that highlights filler-specific characteristics—such as rigidity, porosity, and chemical composition—without the confounding contribution of fibers.

In contrast, hybrid composites S10–S12 were formulated with 7.5 wt. % biofiller combined with 32.5 wt. % flax fibers, resulting in a total reinforcement content of 40 wt. % that is identical to that of the filler-only systems. This design choice ensures that the overall reinforcement fraction remains constant, whereas the reinforcement architecture changes from a particle-dominated system (S3–S5) to a fiber-dominated hybrid system (S10–S12). Such an approach allows the effect of the reinforcement morphology (particulate versus fibrous) to be evaluated independently of the total reinforcement content.

The lower filler fraction (7.5 wt. %) in the hybrid systems was intentionally selected to avoid excessive particle agglomeration and processing-induced defects that are known to occur when particulate fillers are introduced at high loadings into fiber-reinforced thermoplastics. At this level, the biofillers primarily act as secondary modifiers, influencing interfacial bonding, stress transfer, pore structure, and functional behavior, whereas flax fibers remain the primary load-bearing phase. The observed performance enhancements in S10–S12 relative to both neat PLA and filler-only systems are therefore interpreted as arising from synergistic interactions between flax fibers and the respective biofillers rather than from filler loading alone.

Accordingly, the comparison between S3 and S5 and S10–S12 is not intended to imply equivalence between microfiller-only and hybrid micro–nanoscale structured systems, but rather to demonstrate how the same waste fillers behave when transitioning from a primary reinforcement role to a secondary, synergy-driven role within a fiber-reinforced PLA matrix.

2.1.4. Fabrication of Hybrid Composites

Flax/PLA hybrid composites were fabricated using dry premixing, followed by melt impregnation via compression molding. This processing route was selected to enable controlled melting of the PLA matrix, effective fiber wetting, and consolidation under applied pressure, while avoiding the use of extrusion or internal melt compounding. Although no continuous melt mixing was employed, this approach ensured that dispersion and impregnation occurred in the molten state of PLA during hot pressing. Prior to fabrication, all raw materials were dried in a vacuum oven (Model: NSW-144, Narendra Scientific Works, New Delhi, India) at 80 °C for 12 h to eliminate residual moisture and prevent the hydrolytic degradation of PLA during subsequent thermal processing. Moisture removal is critical because the presence of water can induce chain scission in PLA and promote void formation during melt impregnation. After drying, predetermined quantities of PLA pellets, flax fibers, and biofillers were weighed using a precision digital balance (accuracy ± 0.01 g) according to the formulations listed in Table 1.

The weighed constituents were manually dry premixed for approximately 10 min to achieve an initial macroscopically uniform distribution of the fibers and fillers. This premixed blend was then transferred into a preheated stainless-steel mold (300 mm $\times$ 300 mm $\times$ 3 mm) and subjected to compression molding using a hydraulic hot press (Model: HP-30, Technosearch Instruments, Mumbai, India). Molding was conducted at 190 °C under a constant pressure of 5 MPa. An initial preheating stage of 3 min was applied to fully melt the PLA matrix, thereby reducing its viscosity and enabling filler redistribution and effective wetting of the flax fibers within the molten polymer. A similar melt impregnation behavior under compression molding conditions has been reported to promote satisfactory dispersion and fiber–matrix interfacial contact when an appropriate temperature and pressure are applied [52,54–58].

Following preheating, the pressure was maintained for 10 min to facilitate complete impregnation of the flax fibers, expulsion of the entrapped air, and stabilization of the composite microstructure. The molded laminates were subsequently cooled at a controlled rate of 5–10 °C/min under sustained pressure to minimize residual stresses, prevent

warping, and ensure dimensional stability in accordance with established processing recommendations [59,60]. After cooling to room temperature, the composite sheets were removed from the mold and conditioned at 25 °C and 50% relative humidity for 24 h before machining and characterization.

Neat PLA (S1) was prepared as a control sample, while S9 contained flax fibers as the sole reinforcement. Samples S3–S5 consisted of single-filler PLA composites with 40 wt. % WPNS, WTLF, or WQES, respectively. The hybrid composites S10–S12 contained 32.5 wt. % flax fibers and 7.5 wt. % of each biofiller to enable comparative evaluation of flax–filler synergy. For each formulation, six replicate laminates were fabricated to ensure experimental reliability. The test specimens were machined from the molded sheets in accordance with the relevant ASTM standards for mechanical, thermal, and acoustic characterization.

2.2. Testing of Hybrid Flax-Based Composites

2.2.1. Apparent Density Measurement

The density of the fabricated composites was determined as the apparent (bulk) density, reflecting the consolidation quality of the compression-molded laminates. Rectangular specimens were cut from the molded sheets, and their mass was measured using a precision digital balance with an accuracy of ± 0.01 g. The specimen dimensions, as specified in ASTM D792 [61], were measured at multiple locations using a digital Vernier caliper (accuracy ± 0.01 mm), and the average values were used to calculate the external volume.

The apparent density (ρ_{app}) was calculated using Equation (1),

$$\rho = \frac{m}{v} \quad (1)$$

where m is the specimen mass and V is the external geometric volume. This method inherently captures the combined effects of the material composition, laminate compaction, and residual porosity introduced during compression molding. Accordingly, the reported values represent the laminate-level apparent density rather than the intrinsic material density of the constituent phases.

2.2.2. Mechanical Characterization

The tensile behavior was evaluated according to ASTM D638 [62]. Type-IV specimens (63.5 mm $\times$ 13 mm $\times$ 3 mm) were tested using a universal testing machine (Model: AIM-653-1, Aimil Ltd., New Delhi, India) at a crosshead speed of 5 mm/min. Five replicates were tested for each composite to ensure statistical reliability. The flexural performance was assessed using ASTM D790 [63] based on a three-point bending configuration with a span-to-depth ratio of 16:1. Rectangular samples (127 mm $\times$ 12.7 mm $\times$ 3 mm) were loaded at 2 mm/min until failure or a strain limit of 5% was reached. Impact resistance was quantified using the Izod impact tester (Model: IT 504, Tinius Olsen Ltd., Horsham, PA, USA) in accordance with ASTM D256 [64]. Notched specimens (63.5 mm $\times$ 12.7 mm $\times$ 3 mm) were tested with a 2.7 J hammer. The impact strength is reported in kJ/m². Hardness measurements were performed using a Shore-D durometer in accordance with ASTM D785 [65] (Model: DRHTD, Yuzuki Instruments, Tokyo, Japan). Six readings were obtained for each sample and averaged.

2.2.3. Thermal Conductivity

The thermal conductivity of the composite specimens was determined using a laser flash analyzer (Model: LFA-467, NETZSCH-Gerätebau GmbH, Selb, Germany) in accordance with ASTM E1461 [66]. Disk-shaped specimens with uniform thickness were prepared from compression-molded laminates. Prior to testing, both surfaces of each specimen were coated with a thin graphite layer to ensure uniform absorption of the laser pulse and

consistent emissivity during the measurement. The laser flash method directly measures the thermal diffusivity (α) by recording the transient temperature response of the specimen after laser heating. Thermal conductivity (k) was subsequently calculated using Equation (2),

$$k = \alpha \rho_{app} C_p \quad (2)$$

where ρ_{app} is the apparent density of the composite laminate determined experimentally, and C_p is the specific heat capacity obtained from literature values for PLA-based composites [22]. It is emphasized that the use of apparent density in the calculation reflects laminate-level consolidation effects, including packing efficiency and residual porosity, rather than the intrinsic material density.

2.2.4. Sound Absorption Testing

The sound absorption behavior was measured using the two-microphone impedance tube method according to ASTM E1050 [67]. Disk specimens (3 mm thickness; 99.5 mm and 29.5 mm diameter) were tested using an impedance tube setup (Model: Brüel & Kjær/Bruker, Nærum, Denmark). The sound absorption coefficient (SAC) and noise reduction coefficient (NRC) were derived using Equations (3) and (4), respectively:

$$SAC = 1 - \left(\frac{I_{ref}}{I_{inc}} \right) \quad (3)$$

where I_{ref} is the intensity of the reflected sound and I_{inc} is the intensity of the incident sound.

$$NRC = \frac{SAC_{250} + SAC_{500} + SAC_{1000} + SAC_{2000}}{4} \quad (4)$$

where SAC_{250} , SAC_{500} , SAC_{1000} , and SAC_{2000} are the sound absorption coefficients at 250, 500, 1000, and 2000 Hz, respectively.

2.2.5. Surface Roughness

Surface roughness (R_a) was analyzed using a contact profilometer (Model: ContourX-100, Bruker Nano Surfaces, Tucson, AZ, USA) according to ASTM D7127 [68]. Measurements were taken at three distinct locations for each specimen, utilizing a 5 mm evaluation length and a 0.8 mm cutoff wavelength.

2.2.6. Water Absorption and Retention Behavior

The moisture uptake was evaluated according to the ASTM D570 standard [69]. Rectangular specimens (50 mm × 50 mm × 3 mm) were dried at 60 °C for 24 h, weighed (W_{dry}), and immersed in distilled water at 23 °C for 24 h. After immersion, the samples were wiped dry and reweighed (W_{time}). Water absorption was calculated using Equation (5) as follows:

$$W_{abs}(\%) = \frac{W_{time} - W_{dry}}{W_{dry}} \times 100 \quad (5)$$

where W_{abs} is the weight percentage of water absorbed by the developed composite, W_{time} is the weight of the sample during water immersion, and W_{dry} is the initial weight of the dry sample.

2.2.7. Microstructural Characterization by SEM

The microstructural characteristics of the fractured composite specimens were examined using scanning electron microscopy (SEM) (Model: ContourX-100, Bruker Nano Surfaces, Tucson, AZ, USA) to elucidate the dispersion of the fillers, fiber–matrix interfacial

bonding, void distribution, and fracture mechanisms. Tensile-fractured specimens of hybrid composites S10, S11, and S12 were selected to directly correlate the microstructural features with the mechanical performance. The fractured surfaces were cut into small sections and sputter-coated with a thin layer of gold to prevent charging during imaging. SEM observations were performed at an accelerating voltage of 10–15 kV under high vacuum conditions. Micrographs were captured at multiple magnifications to examine the fiber pull-out, interfacial debonding, crack propagation paths, particle dispersion, and void morphology. Representative images were selected and annotated to highlight the dominant microstructural features governing the stress transfer and failure behavior.

3. Results and Discussions

3.1. Apparent Density

Figure 3 presents the apparent density values of the neat PLA, filler-only composites, flax-reinforced composites, and hybrid flax–filler systems. Neat PLA (S1) exhibited the lowest apparent density of 1.32 g/cm³, which is close to the intrinsic density of PLA reported in the literature. The relatively low value reflects the absence of reinforcing phases and limited resistance to volumetric shrinkage during cooling, leading to a comparatively lower consolidation efficiency in the compression-molded laminate. The flax-reinforced composite (S9) showed a substantially higher apparent density of 1.96 g/cm³. This increase is attributed to the presence of continuous flax fibers, which restrict matrix shrinkage, promote mechanical interlocking, and enhance compaction under the applied pressure. The fibrous architecture facilitated better stress transfer and void suppression, resulting in a more consolidated laminate structure than that of neat PLA.

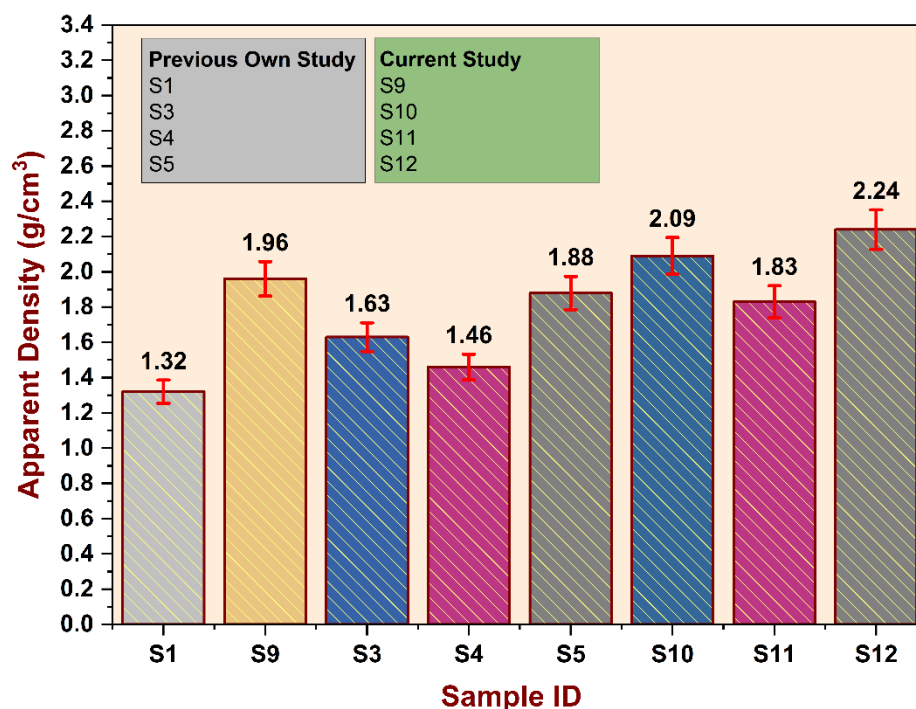


Figure 3. Apparent densities of PLA and flax–biofiller hybrid composites.

The WPNS-filled composite (S3) exhibited an apparent density of 1.63 g/cm³, indicating a moderate improvement in consolidation relative to neat PLA. The increase arises from the packing effect of lignocellulosic particles, which partially fill the free volume within the matrix. However, the irregular morphology and lower intrinsic density of WPNS limit further compaction, resulting in a density lower than that of fiber-reinforced systems. The

WTFLF-filled composite (S4) recorded an apparent density of 1.46 g/cm^3 , slightly lower than that of S3. This behavior is associated with the porous and fibrous nature of waste tea leaf particles, which tend to introduce microvoids and reduce the packing efficiency during molding. As a result, S4 exhibited higher internal porosity and lower laminate consolidation than the other filler-only composites. The WQES-filled composite (S5) showed a higher apparent density of 1.88 g/cm^3 , reflecting the influence of calcium carbonate-rich eggshell particles with higher intrinsic density. The finer particle size and higher surface area of WQES promoted improved packing and reduced void content under compression molding, leading to enhanced laminate consolidation relative to the WPNS- and WTFLF-based systems.

The hybrid composite containing flax fibers and WPNS (S10) exhibited an apparent density of 2.09 g/cm^3 . The significant increase compared to single-reinforcement systems indicates synergistic consolidation effects, where flax fibers provide structural integrity, while WPNS particles occupy interstitial spaces within the fiber network. This combined reinforcement architecture improves the compaction efficiency and reduces the void volume during melt impregnation. The flax–WTFLF hybrid composite (S11) showed an apparent density of 1.83 g/cm^3 , which is lower than that of S10 and S12. The reduced density is attributed to the inherently porous structure of the WTFLF, which limits its ability to fill voids effectively within the flax fiber network. Consequently, although fiber reinforcement enhances consolidation, the presence of the WTFLF maintains a relatively higher internal porosity. The highest apparent density of 2.24 g/cm^3 was observed for the flax–WQES hybrid composite (S12). This result highlights the strong synergistic effect between the flax fibers and fine CaCO_3 -rich WQES particles. The fibers act as the primary load-bearing phase, while the high-density, fine WQES particles efficiently fill voids and enhance the packing efficiency, resulting in superior laminate consolidation. The elevated apparent density indicates a minimal void content and a highly compact composite structure.

It is emphasized that the apparent density values reported here do not represent true intrinsic material densities, but rather serve as a comparative indicator of consolidation quality and internal porosity among the different composite systems. A higher apparent density corresponds to improved compaction and reduced void fraction, which directly influences mechanical stiffness, thermal transport, and acoustic behavior. Accordingly, the density results are interpreted comparatively to elucidate the structure–property relationships rather than as absolute material constants.

Theoretical Density Validation Using the Rule of Mixtures

To evaluate the physical plausibility of the experimentally measured apparent density values, the theoretical densities of all the composite systems were calculated using the rule of mixtures (ROM) based on the intrinsic constituent densities reported in the literature [23,24,26]. The ROM calculation provides a reference benchmark for the intrinsic material density of each composite and enables the assessment of deviations arising from laminate consolidation and porosity effects. The intrinsic densities used for the calculation were approximately 1.25 g/cm^3 for PLA, 1.45 g/cm^3 for flax fibers, 1.30 g/cm^3 for lignocellulosic fillers (WPNS and WTFLF), and 2.70 g/cm^3 for CaCO_3 -rich waste quail eggshell (WQES). The theoretical density (ρ_{ROM}) was calculated using Equation (6),

$$\rho_{\text{ROM}} = \left(\sum \frac{\omega_i}{\rho_i} \right)^{-1} \quad (6)$$

where ω_i and ρ_i represent the weight fraction and intrinsic density of each constituent, respectively.

As shown in Table 2, the ROM-predicted densities of all composite systems lie within a physically realistic range of approximately $1.25\text{--}1.55 \text{ g/cm}^3$, depending on the reinforce-

ment type and composition. These values represent the intrinsic material densities expected based solely on the constituent properties and composition. The higher experimentally measured values therefore reflect the apparent laminate density, which is influenced by consolidation quality, packing efficiency, and residual porosity introduced during compression molding, rather than unit inconsistencies or calculation errors. Accordingly, the ROM analysis confirms that the density discrepancies arise from processing-related effects and supports the interpretation of experimental density values as comparative indicators of consolidation quality rather than absolute material constants.

Table 2. Theoretical densities of composite systems calculated using the rule of mixtures.

Sample ID	Composite Formulation (wt. %)	ρ_{ROM} (g/cm ³)
S1	100% PLA	1.25
S9	60% PLA + 40% flax fiber	1.33
S3	60% PLA + 40% WPNS	1.27
S4	60% PLA + 40% WTLF	1.27
S5	60% PLA + 40% WQES	1.55
S10	60% PLA + 32.5% flax fiber + 7.5% WPNS	1.31
S11	60% PLA + 32.5% flax fiber + 7.5% WTLF	1.31
S12	60% PLA + 32.5% flax fiber + 7.5% WQES	1.37

3.2. Tensile Strength Analysis

The tensile strength of the flax/PLA composites (Figure 4) exhibited a clear improvement trend with the incorporation of fillers, with values ranging from 52.36 to 60.41 MPa. Pure PLA (S1) exhibited the lowest strength at 21.74 MPa, primarily owing to its inherently brittle nature and limited strain accommodation. The addition of 40 wt. % flax fiber (S9) significantly increased the tensile strength to 48.25 MPa, confirming the superior reinforcing capability of flax, whose high cellulose crystallinity and low microfibril angle facilitate efficient stress transfer within the PLA matrix. When individual fillers were incorporated with flax, distinct strengthening behaviors were observed. The flax–WPNS–PLA composite (S10) attained the highest tensile strength of 60.41 MPa, followed by flax–WTLF–PLA (S11) at 56.27 MPa and flax–WQES–PLA (S12) at 52.36 MPa, respectively. The superior performance of S10 was attributed to the rigid lignocellulosic structure and high hardness of the pistachio shell particles, which promoted better load-sharing by restricting local plastic deformation. The enhanced bonding between the alkali-treated WPNS and the flax/PLA interface minimized voids and filler pullout, thereby improving crack resistance. S11 also exhibited substantial reinforcement owing to the polyphenolic constituents of the tea-leaf particles, which improved the interfacial adhesion through hydrogen bonding and constrained the polymer chain mobility. Although S12 also improved the strength, its slightly lower values compared to S10 and S11 resulted from the brittle nature of the CaCO₃-rich quail eggshell particles, which increased the stiffness but provided limited energy dissipation during loading.

A comparative assessment with previous jute-based hybrid composites (S6–S8) revealed important material–structure interactions. In an earlier study [36], jute–WPNS–PLA (S6) achieved 57.38 MPa, jute–WTLF–PLA (S7) reached 51.82 MPa, and jute–WQES–PLA (S8) recorded 48.27 MPa. In all three formulations, the flax-based hybrids (S10–S12) outperformed their jute-based counterparts. For instance, the tensile strength increased from 57.38 MPa in S6 to 60.41 MPa in S10, from 51.82 MPa in S7 to 56.27 MPa in S11, and from 48.27 MPa in S8 to 52.36 MPa in S12, respectively. This consistent improvement is attributed to the intrinsic microstructural differences between flax and jute. While both are lignocellulosic, flax possesses a higher cellulose content (up to 70–80%) and lower lumen volume, providing a denser and more uniform fiber structure that better resists

tensile deformation. The reduced hemicellulose and pectin content in flax lowers moisture absorption, thereby minimizing fiber swelling and interfacial weakening during composite processing. In addition, the lower microfibrillar angle of flax promotes more effective tensile load transfer when aligned within the matrix. These characteristics resulted in improved stress distribution at the fiber–filler–PLA interface, enabling superior strengthening even when fillers were added at the same 7.5 wt. % level.

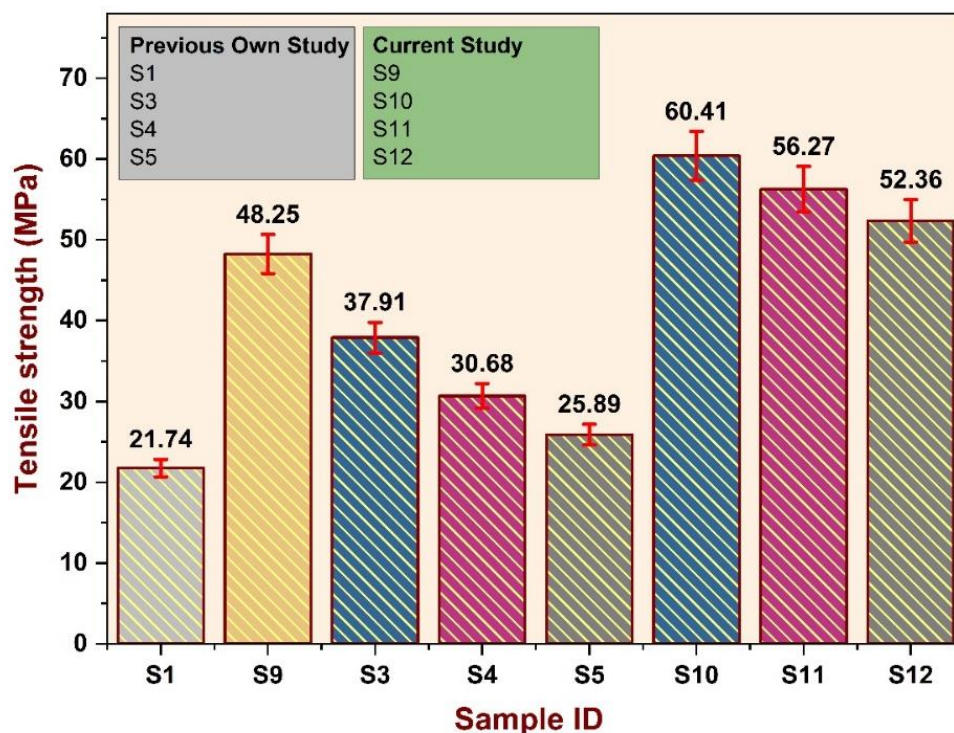


Figure 4. Tensile strength of PLA and flax–biofiller hybrid composites.

Mechanistically, tensile strengthening in flax hybrids arises from three synergistic factors: (i) stronger interfacial adhesion owing to the hydroxyl-rich flax surface promoting hydrogen bonding with PLA and filler functional groups; (ii) reduced interfacial defects because flax fibers collapse less during compression molding than jute fibers, which exhibit higher lumen collapse; and (iii) superior crack-bridging capability, where flax fibers delay crack propagation more effectively, increasing the composite’s resistance to premature failure. In contrast, the jute hybrids displayed a lower tensile strength because jute contains a higher hemicellulose content and bundles with larger lumens, which introduce microvoids and weaken the adhesion during loading. Furthermore, the higher moisture affinity of jute often leads to localized debonding, especially when hard particulates such as WPNS and WQES are present, resulting in the initiation of microcracks. The tensile results clearly indicate that replacing jute with flax significantly enhanced the composite strength across all filler categories. This improvement highlights the superior reinforcing efficiency of flax and its more stable interfacial behavior within the PLA matrix. The trends observed for both the jute and flax systems affirm that WPNS remains the most effective filler owing to its rigidity and surface chemistry, followed by WTLF and WQES. However, the magnitude of improvement was consistently greater in the flax-based composites, demonstrating that flax offers a more compatible and mechanically robust reinforcement platform for PLA-based hybrid biocomposites.

3.3. Flexural Strength Analysis

The flexural strength results (Figure 5) showed a clear enhancement in the bending performance when flax fibers and biofillers were incorporated into the PLA matrix. Pure PLA (S1) exhibited a baseline flexural strength of 48.76 MPa, which is consistent with its brittle behavior and limited resistance to bending stresses. Incorporating flax fiber alone (S9) significantly increased the strength to 89.23 MPa, owing to flax's higher cellulose content, lower microfibril angle, and stronger intrinsic rigidity than jute. These characteristics promote more efficient stress transfer during flexural loading, resulting in superior stiffness and crack-propagation resistance. When individual fillers were added to PLA without fibers (previous work: S3–S5), the flexural strengths were 77.29 MPa for WPNS, 60.24 MPa for WTLF, and 54.89 MPa for WQES, respectively. These moderate enhancements reflected the filler–matrix interfacial interactions and inherent stiffness differences between lignocellulosic (WPNS and WTLF) and CaCO₃-based (WQES) particulates.

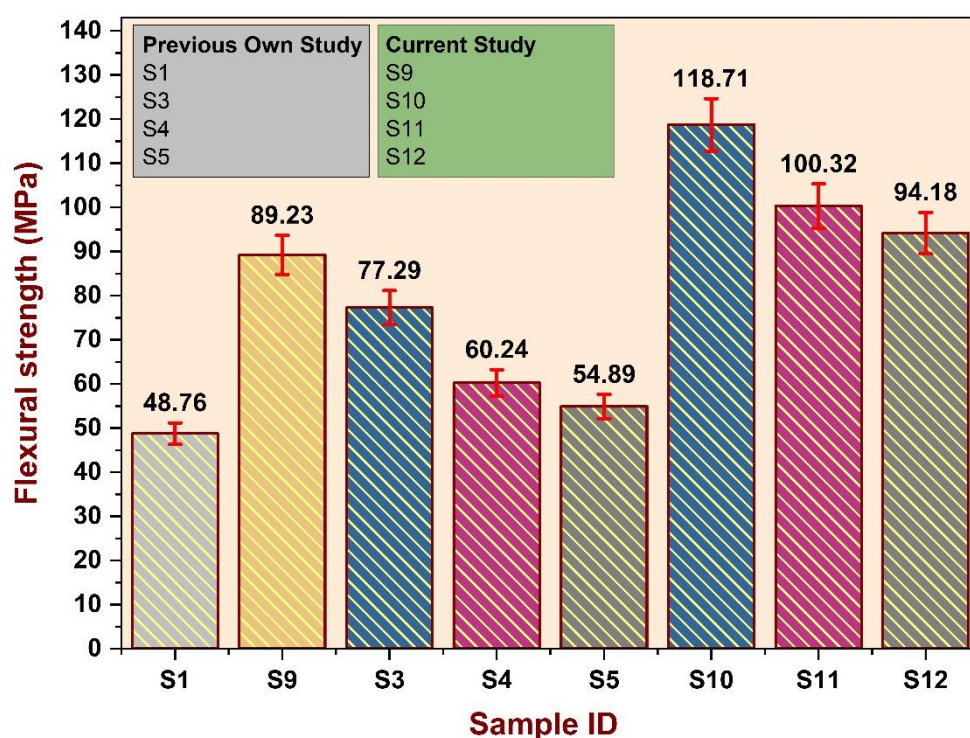


Figure 5. Flexural strength comparison of PLA and hybrid composites.

In the current flax-based hybrid composites, a notable improvement was observed across all three systems. The flax–WPNS hybrid (S10) exhibited the highest flexural strength of 118.71 MPa, surpassing not only its individual components (S9 and S3) but also the jute-based hybrid counterpart S6 (114.67 MPa). This improvement is attributed to the synergistic interaction between the high modulus of flax and the rigid lignocellulosic pistachio shell particles, which enhances the load transfer efficiency and restricts shear deformation under bending. The superior packing density achieved by the WPNS within the flax-reinforced matrix likely reduced the void content and increased the composite stiffness, further increasing the flexural resistance. Similarly, the flax–WTLF composite (S11) exhibited a strength of 100.32 MPa, which was higher than that of flax alone (S9) and the previous jute–WTLF hybrid (S7:95.59 MPa). Tea leaf particulates contain polyphenolic compounds and exhibit surface roughness, which improves β -relaxation damping and frictional resistance at the interface. When combined with flax fibers that possess higher tensile stiffness and

reduced hydrophilicity (post-alkali) [36], the composite experienced a more stable stress redistribution during bending, resulting in improved flexural performance.

For the CaCO_3 -rich quail eggshell filler, the flax–WQES hybrid composite (S12) reached 94.18 MPa, exceeding both S9 and the jute-based WQES hybrid (S8:88.27 MPa). This enhancement, although less pronounced than that in the WPNS and WTLF systems, can be explained by the rigid, mineral-rich structure of the eggshell particles, which improves the compressive stiffness during bending. However, owing to the brittle nature and angular morphology of CaCO_3 particles, stress concentrations tend to form at higher load levels, slightly limiting the overall increment compared to lignocellulosic fillers. Nonetheless, flax fibers compensate for this by offering higher interfacial adhesion and reduced microcrack initiation, thereby enabling S12 to outperform the corresponding jute-based formulation.

A comparison of the flax and jute hybrid systems distinctly highlights the superiority of flax as a reinforcement under flexural loading. For all three filler combinations, flax-based hybrids (S10–S12) consistently exhibited flexural strengths that were 4–7% higher than those of their jute counterparts (S6–S8). This improvement is attributed to the higher tensile stiffness, finer fiber structure, and better surface activation of flax after alkaline treatment, which collectively enhance the fiber–matrix bonding and reduce interfacial slippage. Jute fibers, which are coarser and more lignin-rich, introduce slightly more internal voids and microstructural heterogeneity, which weaken the bending resistance compared to flax. Thus, the results confirm that replacing jute with flax significantly improves the mechanical performance of PLA-based biofiller composites under flexural stress, demonstrating the potential of flax for stiffer and more structurally reliable biodegradable composite applications.

3.4. Impact Strength Analysis

The impact strength results (Figure 6) for both the current flax-reinforced PLA composites (S10–S12) and the earlier jute-reinforced systems (S6–S8) revealed clear differences in the energy absorption capability influenced by the fiber morphology, interfacial adhesion, and filler–matrix compatibility. In the present study, the incorporation of flax fibers significantly enhanced the resistance to crack propagation compared to neat PLA (1.12 kJ/m², S1), with the flax/PLA control (S9) displaying an impact strength of 3.56 kJ/m² due to flax's higher strain-to-failure, finer fibrillation tendency, and superior fiber pullout characteristics relative to jute. When biofillers were incorporated, the flax-based hybrid composites exhibited further improvement: S10 (flax + WPNS + PLA) achieved 4.75 kJ/m², S11 (flax + WTLF + PLA) registered the highest value of 5.04 kJ/m², and S12 (flax + WQES + PLA) attained 4.13 kJ/m². This enhancement arises from the improved stress delocalization caused by the synergistic effects between the surface-treated flax fibers and treated fillers, which together create microcrack deflection pathways, controlled fiber pull-out, and increased interfacial friction—mechanisms known to dissipate impact energy efficiently.

Compared with earlier jute-based hybrid systems from a previous study [36], similar trends were observed in filler ranking, but with consistently lower absolute values. The jute-based hybrids S6 (WPNS), S7 (WTLF), and S8 (WQES) recorded impact strengths of 4.02, 4.35, and 3.74 kJ/m², respectively. Although WTLF provided the highest impact response among all the fillers, the magnitude of improvement in the jute systems was noticeably lower than that in the corresponding flax-based composites. This difference stems from the inherent structural and mechanical disparities between the two fibers. Jute fibers possess a higher lignin content (~12–14%) and a greater microfibrillar angle, rendering them stiffer and more brittle under impact loading. Consequently, the jute tends to fracture abruptly rather than undergo fiber pullout, thereby reducing the energy absorption. In contrast, flax fibers contain a higher cellulose content (>70%) and a lower microfibril angle, enabling better elastic deformation and improved crack bridging during impact events. These

characteristics allow the flax fibers to delay crack propagation more effectively, resulting in superior toughness in the present study.

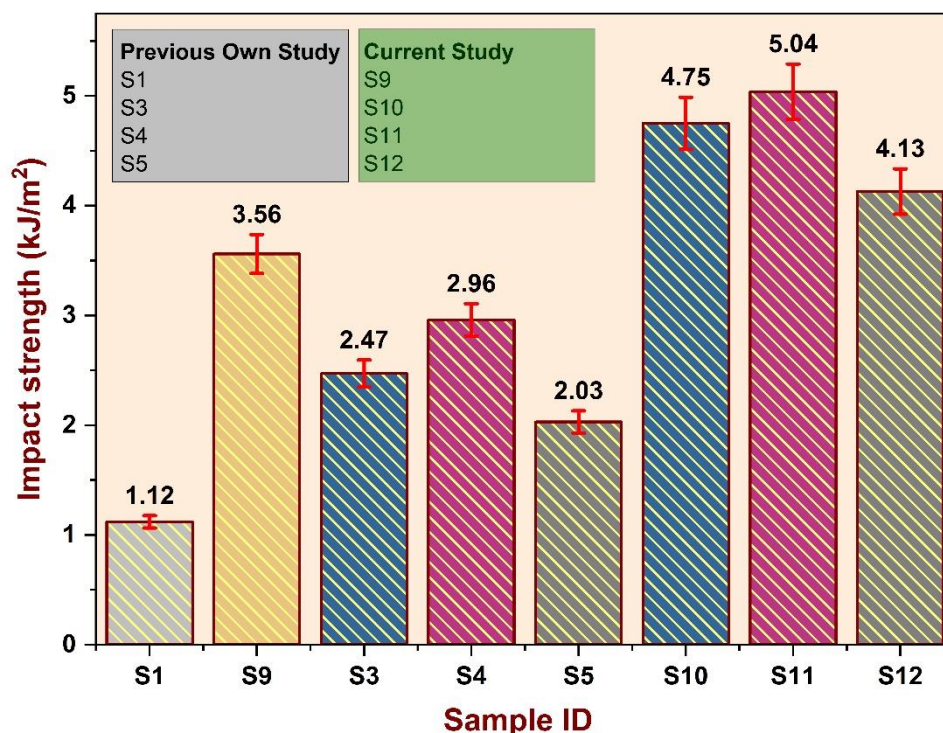


Figure 6. Impact strength of flax/PLA composites with various biofillers.

The influence of each filler was consistent across both studies, with the WTLF exhibiting the highest energy-absorbing potential owing to its porous, polyphenol-rich morphology, which creates microcavities within the matrix, promoting crack deviation and plastic deformation. The WPNS offers moderate improvement owing to its rigid lignocellulosic shell structure, which enables stress transfer but limits ductility. WQES, dominated by CaCO_3 , enhances stiffness but restricts high-impact absorption owing to its brittle mineral nature, explaining the lower values in S12 and S8 compared with organic fillers. A direct comparison demonstrated that flax-based hybrids exhibited 15–20% higher impact strength than their jute-based counterparts for the same filler type. This superiority is attributed to the more compliant fiber structure of flax, stronger fiber–matrix interlocking resulting from the alkali treatment, and better stress redistribution facilitated by finer fibril bundles. Thus, the present study confirms that replacing jute with flax not only improves the intrinsic toughness but also maximizes the reinforcement potential of agricultural waste fillers within a PLA matrix.

3.5. Shore D Hardness

The Shore D hardness values of the developed composites (Figure 7) revealed a consistent improvement upon the incorporation of flax fibers and biofills into the PLA matrix. Pure PLA (S1) exhibited a hardness of 78.83 °ShD, whereas the addition of flax fiber to S9 increased the hardness to 81.15 °ShD. This can be attributed to the inherent stiffness of flax and its higher cellulose content, which promotes better stress transfer at the surface. When the individual fillers were blended with PLA (S3–S5), the hardness values increased to 83.71 °ShD for WPNS (S3), 80.35 °ShD for WTLF (S4), and 86.82 °ShD for WQES (S5). These enhancements are primarily linked to the rigid lignocellulosic structure of WPNS, moderate stiffness contributed by phenolic-rich WTLF, and high mineral (CaCO_3) content of WQES, which increases the surface resistance under indentation.

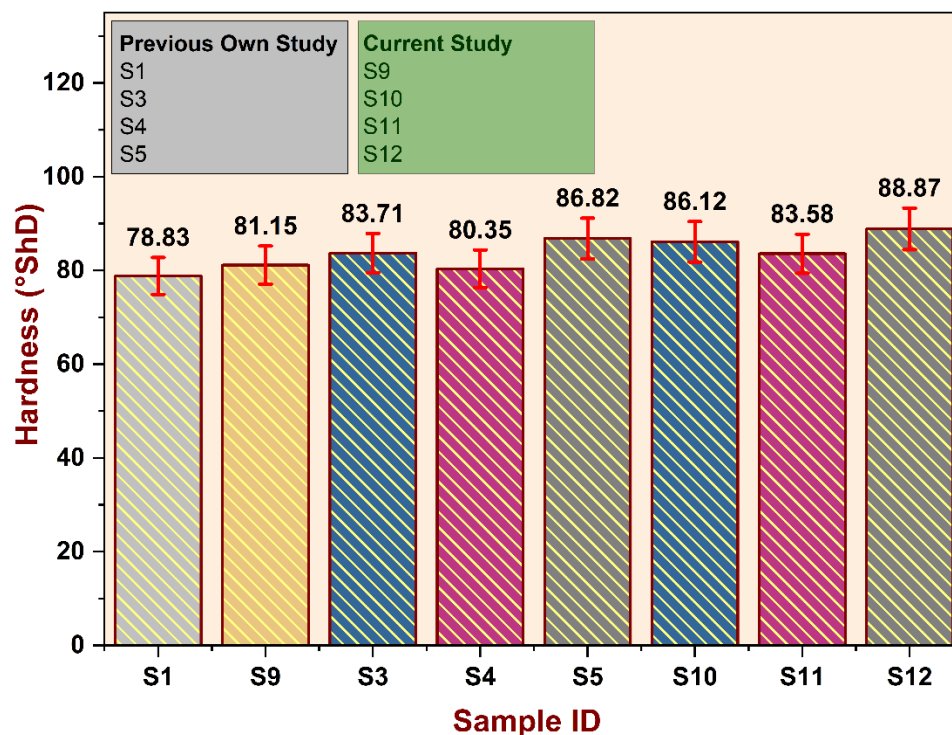


Figure 7. Shore-D hardness of PLA and flax-biofiller hybrid composites.

In the current flax-based hybrid composites (S10–S12), the hardness increased further, with WPNS–flax–PLA (S10) reaching 86.12 °ShD, WTLF–flax–PLA (S11) achieving 83.58 °ShD, and the WQES–flax–PLA hybrid (S12) exhibiting the highest value of 88.87 °ShD. The additional increase in hardness compared to that of S9 was primarily governed by the synergistic action between the flax fibers and fillers. Flax possesses a higher tensile modulus and lower microfibril angle than jute, enabling superior interfacial packing and matrix stiffening, which facilitates better constraint of polymer chain mobility near the surface, as reflected by higher indentation resistance. The fillers further enhanced the superficial rigidity by occupying microvoids and increasing the local stiffness, particularly in the case of WQES, where CaCO_3 micro-nanoscale domains acted as micro-reinforcements that resisted localized deformation.

Comparing these values with the hardness values of previous jute-based hybrids (S6–S8) [36], which were 84.43 °ShD for WPNS, 81.94 °ShD for WTLF, and 87.12 °ShD for WQES, it is evident that flax-based hybrids consistently show superior performance. The improvement ranged from approximately 1–2 °ShD for the WQES system and 2–3 °ShD for the WPNS and WTLF systems. This enhancement is mechanistically linked to the higher stiffness of flax fibers relative to jute fibers, as flax fibers possess higher crystallinity and a more compact microstructure, enabling superior filler-locking effects within the PLA matrix compared to jute fibers. In jute-based composites, residual lumen voids and higher hemicellulose content promote microcompression during indentation, causing slightly lower hardness. Conversely, flax fibers, with their smoother fibrillar alignment and reduced internal porosity, improve the packing density and reduce polymer chain relaxation during indentation, thereby increasing the shore hardness. Additionally, flax fibers form more uniform filler–fiber–matrix networks owing to their smaller diameter variation and better surface compatibility with PLA after alkali treatment, further enhancing the resistance to surface deformation. The hardness results clearly show that flax-based composites outperform the corresponding jute-based systems for all three fillers, confirming that flax is a more effective reinforcing phase for improving the surface integrity and resistance

to localized mechanical damage. The S12 (flax + WQES + PLA) composite exhibited the highest hardness among all the materials, underscoring the combined strengthening effect of the CaCO_3 -rich fillers and high-modulus flax fibers.

3.6. Thermal Conductivity Analysis

Figure 8 illustrates the thermal conductivities of the neat PLA, filler-only composites, flax-reinforced composites, and hybrid flax–filler systems. Neat PLA (S1) exhibited the lowest thermal conductivity of $0.163 \text{ W}/(\text{m}\cdot\text{K})$, which is consistent with the inherently low thermal transport capability of amorphous biodegradable polymers due to strong phonon scattering within the polymer chains. The incorporation of flax fibers (S9) increased the thermal conductivity to $0.265 \text{ W}/(\text{m}\cdot\text{K})$, reflecting the contribution of fibrous reinforcement in forming more continuous heat-transfer pathways within the matrix. The aligned and interconnected nature of flax fibers reduces phonon scattering compared to neat PLA, while also enhancing laminate consolidation under compression molding.

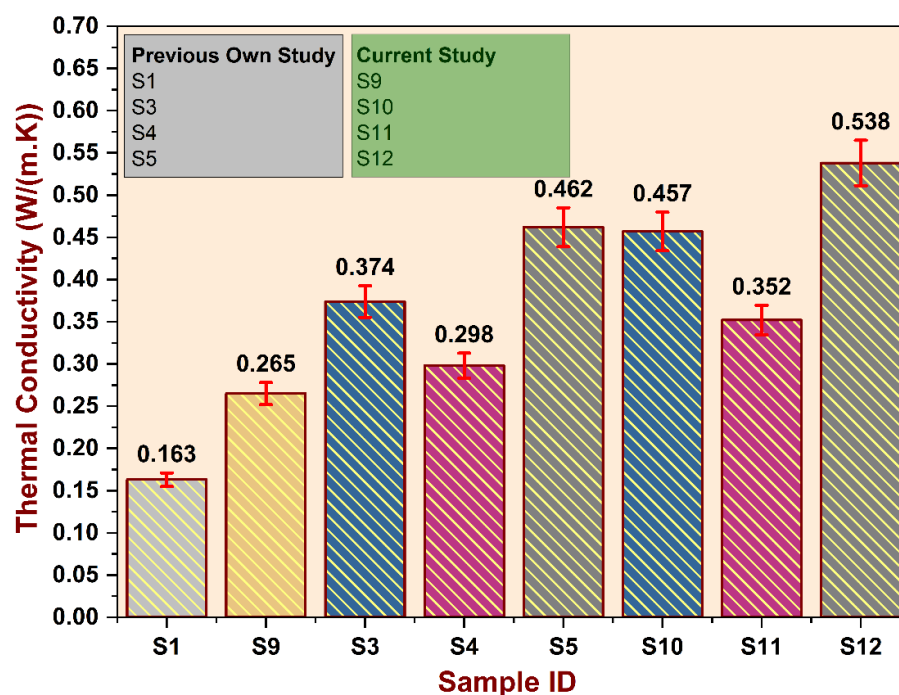


Figure 8. Thermal conductivity of PLA and flax-based hybrid composites.

Among the filler-only composites, the WPNS- and WTLF-filled systems (S3 and S4) showed moderate increases in thermal conductivity, reaching $0.374 \text{ W}/(\text{m}\cdot\text{K})$ and $0.298 \text{ W}/(\text{m}\cdot\text{K})$, respectively. The enhancement observed in S3 is attributed to the improved packing efficiency and reduced void content relative to S4, where the inherently porous structure of the WTLF introduces additional phonon-scattering sites that limit heat transfer. In contrast, the WQES-filled composite (S5) exhibits a higher thermal conductivity of $0.462 \text{ W}/(\text{m}\cdot\text{K})$, owing to the CaCO_3 -rich composition of eggshell particles, which possess higher intrinsic thermal conductivity than lignocellulosic fillers and promote more efficient heat conduction through the composite.

The hybrid composites demonstrated distinct thermal transport behavior arising from the combined presence of flax fibers and biofillers. The flax–WPNS hybrid (S10) showed a thermal conductivity of $0.457 \text{ W}/(\text{m}\cdot\text{K})$, comparable to that of S5, indicating that the fibrous network provided by flax fibers, together with particulate packing by WPNS, facilitates efficient heat-transfer pathways. The flax–WTLF hybrid (S11) exhibited a lower value of $0.352 \text{ W}/(\text{m}\cdot\text{K})$, which is consistent with the higher internal porosity and reduced packing

efficiency associated with the WTLF particles, leading to increased phonon scattering and thermal resistance. The highest thermal conductivity was observed for the flax–WQES hybrid composite (S12), reaching 0.538 W/(m·K). This enhancement is attributed to the synergistic interaction between the flax fibers and fine, CaCO₃-rich WQES particles. Flax fibers act as the primary heat-conducting skeleton, while WQES particles efficiently occupy interstitial regions, reducing the void content and interfacial thermal resistance. The resulting compact microstructure provides continuous and efficient heat-transfer pathways, thereby maximizing the thermal conductivity.

3.7. Sound Absorption Properties

The sound absorption behavior of the developed flax/PLA composites (Figure 9) showed a marked improvement over neat PLA and demonstrated clear filler-dependent trends associated with the intrinsic pore structure, lignocellulosic content, and CaCO₃-rich particulates. Neat PLA (S1) exhibited the lowest sound absorption coefficient (SAC = 0.283) and noise reduction coefficient (NRC = 0.207), consistent with its dense and nonporous morphology, which restricts the viscous and thermal dissipation of the incident acoustic energy. The incorporation of flax fibers in S9 increased the SAC to 0.403 and NRC to 0.346, largely because the hollow lumen structure, rough surfaces, and fibrillar arrangement of flax introduced tortuous pathways and microcavities that enhanced frictional losses in the acoustic boundary layer. This initial improvement confirmed that flax is an effective acoustic modifier compared to the relatively smooth PLA matrix.

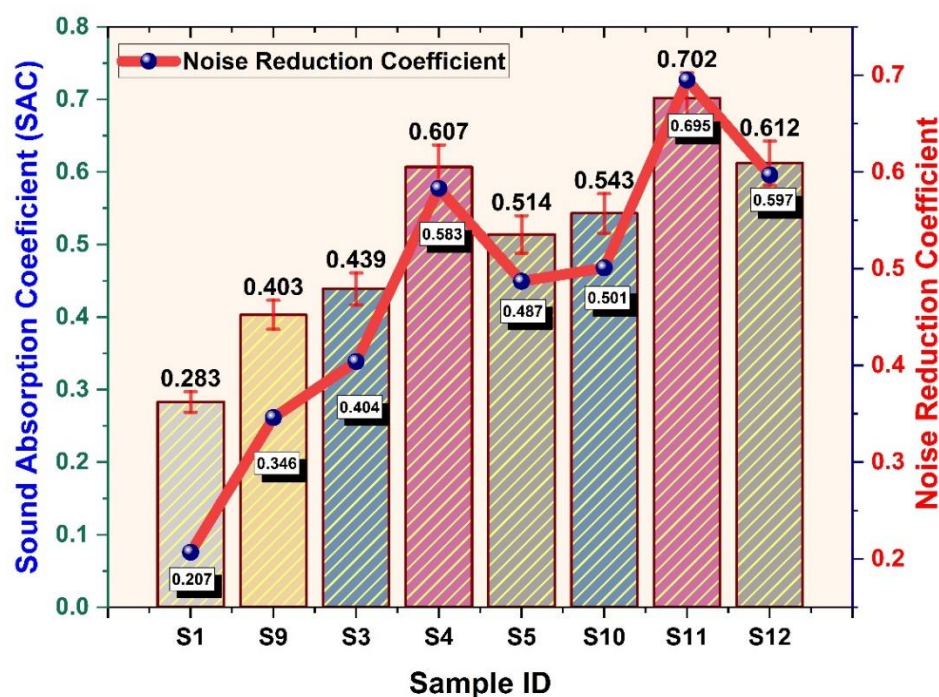


Figure 9. Sound absorption performance of flax/PLA biofilled composites.

The introduction of individual fillers further increased the sound absorption performance, with WPNS-filled PLA (S3) recording SAC = 0.439 and NRC = 0.404, WPNS-filled PLA (S4) achieving SAC = 0.607 and NRC = 0.583, and WTLF-filled PLA (S5) showing SAC = 0.514 and NRC = 0.487. The superior absorption of the WTLF can be attributed to its highly porous microstructure and the natural presence of phenolic compounds and entrapped air voids, which increase the acoustic impedance mismatch of the composite and facilitate multifrequency attenuation. WPNS, being lignocellulosic with hard granular particles, enhances absorption through internal friction and microscattering, whereas WQES,

dominated by CaCO_3 , offers controlled stiffness increments that shift the peak absorption toward mid-frequency ranges.

A more pronounced enhancement was observed for the flax hybrid composites. The flax–WPNS hybrid (S10) achieved $\text{SAC} = 0.543$ and $\text{NRC} = 0.501$, whereas the flax–WTLF hybrid (S11) reached the maximum values among all the samples, with $\text{SAC} = 0.702$ and $\text{NRC} = 0.695$. The flax–WQES system (S12) also exhibited high acoustic performance, with $\text{SAC} = 0.612$ and $\text{NRC} = 0.597$. These improvements arise from the synergistic interaction between the flax fiber networks and filler-induced microporosity. Flax fibers contribute to longitudinal channels and transverse lumens that trap and dissipate acoustic waves, whereas fillers create discontinuities and microgaps that intensify scattering, viscous damping, and thermoelastic loss mechanisms. Among the fillers, the WTLF exhibited the most effective sound absorption, reflecting the combined influence of high porosity, low density, and irregular particle morphology that encourages multidirectional wave attenuation.

Compared with previous jute-based hybrid composites (S6–S8) [36], the flax-based systems exhibited a consistent performance advantage. Jute–WPNS (S6) recorded $\text{SAC} = 0.485$ and $\text{NRC} = 0.462$, jute–WTLF (S7) achieved $\text{SAC} = 0.643$ and $\text{NRC} = 0.618$, and jute–WQES (S8) achieved $\text{SAC} = 0.578$ and $\text{NRC} = 0.539$. Although jute fibers also contain lumens and can support acoustic damping, flax fibers generally have finer fibrils, greater flexibility, and higher surface roughness, all of which promote enhanced sound energy dissipation. Furthermore, flax forms a more interconnected network when embedded in PLA, leading to superior micro-resonance and viscoelastic dissipation compared with the relatively coarse and stiffer jute fibers. This structural difference explains why the flax–WTLF composite (S11) consistently outperformed its jute counterpart (S7), with an NRC improvement from 0.618 to 0.695. Similarly, flax–WPNS (S10) and flax–WQES (S12) exhibited higher acoustic efficiencies than jute–WPNS (S6) and jute–WQES (S8), confirming that flax is inherently more effective in creating tortuous acoustic pathways and microvoids that are conducive to sound absorption.

Across all formulations, the improvements in SAC and NRC can be mechanistically attributed to three dominant phenomena: (i) viscous damping, where fillers and fibers introduce frictional resistance to air particle movement; (ii) thermal dissipation, where porous regions convert sound energy into heat; and (iii) multiscale scattering, which is enhanced by heterogeneities created by filler particles and the flax-fiber network. Notably, the CaCO_3 -rich WQES increased the composite stiffness, shifting the absorption peaks while maintaining substantial dissipation at mid-frequencies. In contrast, the WTLF maximizes broadband absorption owing to its inherently porous cellular architecture and low acoustic impedance. The results confirmed that the flax-based hybrid composites (S10–S12) provided superior acoustic performance compared to both neat PLA and previous jute-based hybrids (S6–S8). The enhanced performance is driven by finer, more compliant flax fibers that interact more effectively with the filler-derived porosity, making flax a more suitable reinforcement for multifunctional acoustic biocomposites.

3.8. Surface Roughness Analysis

The surface roughness (R_a) of the developed composites (Figure 10) exhibited distinct trends depending on the fiber type, filler characteristics, and filler–matrix compatibility. Pure PLA (S1) exhibited the lowest roughness (0.021 mm) because of its homogeneous and continuous polymer structure. The incorporation of flax fibers (S9) slightly increased R_a to 0.028 mm, reflecting the inherently uneven microtexture and lumen-based morphology of the flax fibers, which introduced micro-asperities on the composite surface even after alkali treatment. Among the single-filler systems, WPNS/PLA (S3), WTLF/PLA (S4), and WQES/PLA (S5) exhibited higher roughness values (0.044, 0.076, and 0.065 mm,

respectively). This is directly attributed to the particle morphology of the fillers: WPNS, being lignocellulosic with angular edges, moderately disrupts the surface smoothness; WTLF contains porous, irregular polyphenolic structures that protrude at the surface, causing the highest roughness; and WQES, rich in CaCO_3 , produces fine but rigid particles that generate a comparatively smoother but still textured surface. These microstructural contributions mirror those observed in earlier jute-based studies, confirming that the filler geometry strongly dictates R_a .

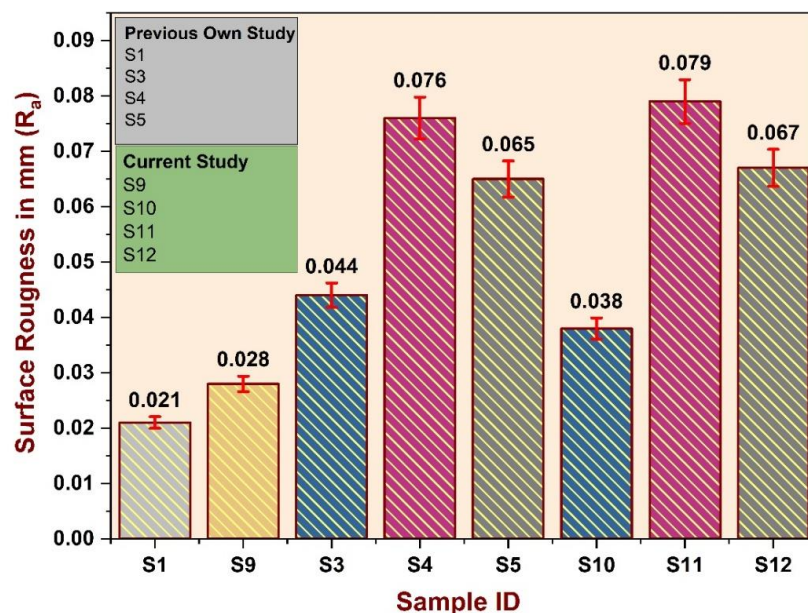


Figure 10. Surface roughness of PLA and flax-reinforced hybrid composites.

In the hybrid flax composites, the surface roughness decreased significantly for all three formulations, with R_a values of 0.038 mm (S10), 0.079 mm (S11), and 0.067 mm (S12). The notable reduction in R_a for the WPNS–flax composite (S10) compared to WTLF–flax (S11) and WQES–flax (S12) is attributed to the improved interfacial wetting provided by the flax fibers, whose finer fibrillation and greater surface accessibility promote better PLA flow around the WPNS particles during compression molding. In contrast, WTLF–flax (S11) exhibited the highest R_a among the flax hybrids owing to the porous and flaky nature of the tea residues, which tend to migrate toward the surface during melt consolidation. The moderate roughness of WQES–flax (S12) (0.067 mm) indicates that the nano–micro CaCO_3 particles disperse well within the flax–PLA matrix but still create localized protrusions because of their rigid inorganic nature.

When comparing the current flax-based hybrids (S10–S12) with previous jute-based hybrids (S6–S8) [36], a consistent reduction in roughness was observed. The jute hybrid series exhibited R_a values of 0.032 (S6), 0.072 (S7), and 0.051 mm (S8), which are typically higher than those of their flax counterparts. This improvement arises from the finer diameter, better fibrillation behavior, and smoother longitudinal surfaces of flax fibers compared to those of jute fibers, which reduce the number of fiber pull-out sites and provide more uniform filler anchoring. Flax also promotes superior melt infiltration of PLA because of its lower hemicellulose content and more efficient alkali-induced defibrillation, thus minimizing the voids that would otherwise manifest as surface asperities. Consequently, the flax–PLA matrix formed a more continuous outer layer, even in the presence of WPNS, WTLF, or WQES fillers, yielding smoother surfaces than the equivalent jute–PLA composites. This demonstrates that flax-based hybrids are more effective in achieving a consistent

surface topology, which is advantageous for tribological performance, coating adhesion, and aesthetic applications.

3.9. Water Absorption Properties

The water absorption behavior (Figure 11) of the PLA-based composites (S1, S9, S10–S12) revealed clear differences arising from the hydrophilic nature of the natural fibers and the microstructural attributes introduced by the biofillers. Pure PLA (S1) exhibited the lowest uptake ($0.92 \pm 0.06\%$), consistent with its low polarity and limited free-volume pathways for moisture diffusion. The incorporation of flax fiber in S9 increased the uptake to $1.24 \pm 0.16\%$, reflecting the inherently high cellulose content of flax and the abundant hydroxyl groups that promote hydrogen bonding with water molecules. When WPNS, WTLF, and WQES were incorporated along with flax (S10–S12), the maximum moisture intake increased further to $1.81\% \pm 0.17\%$, $2.12\% \pm 0.18\%$, and $1.12\% \pm 0.05\%$, respectively. The elevated absorption in S10 and S11 can be attributed to the lignocellulosic structure and internal porosity of the WPNS and WTLF, which provide capillary channels that facilitate Fickian diffusion. In contrast, the relatively low uptake in S12 arose from the dense microstructure and low porosity of the CaCO_3 -rich WQES, which restricted water penetration and reduced the number of available hydrophilic sites. The progressive increase in absorption with immersion time initially follows a steep gradient owing to rapid diffusion into voids and microgaps at the fiber–matrix interface, eventually plateauing as saturation limits are approached.

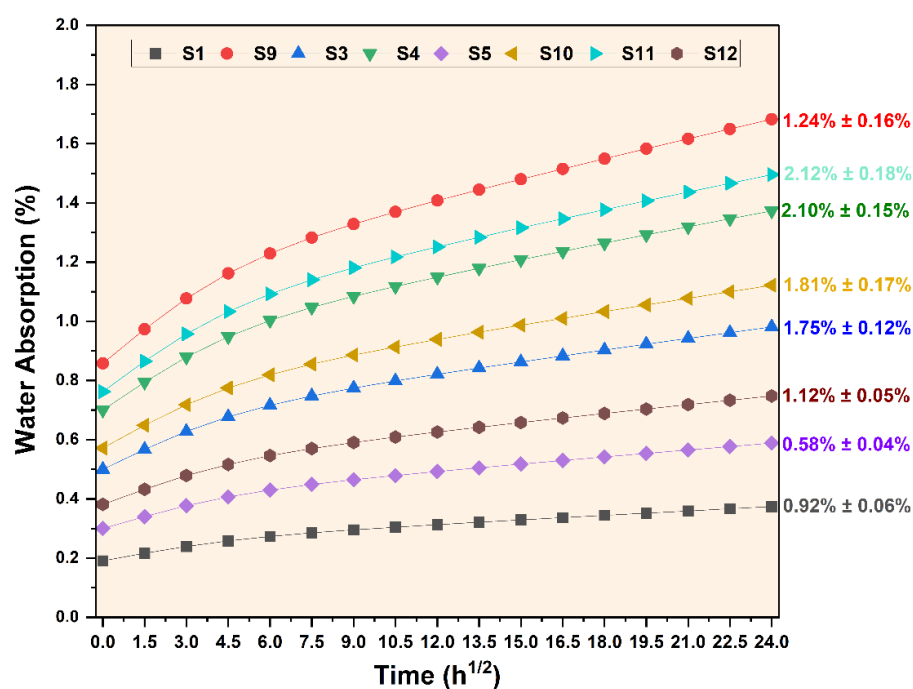


Figure 11. Water absorption of flax/PLA composites with different fillers.

A comparison with jute-based composites from a previous study demonstrated that flax-containing systems generally absorbed less moisture than their jute counterparts. The jute composites S6, S7, and S8 exhibited higher moisture uptakes of $1.90 \pm 0.13\%$, $2.35 \pm 0.16\%$, and $1.20 \pm 0.08\%$, respectively. This reduction in the flax systems (S10–S12) can be mechanistically explained by the finer fibrillar structure of flax, which undergoes more effective alkali modification, producing improved fiber–matrix compatibility and lower interfacial void content. Jute, which is coarser and contains higher hemicellulose content, retains more amorphous hydrophilic regions even after alkali treatment, resulting

in a greater propensity for moisture uptake and swelling. The relatively lower absorption in the flax–WQES (S12) compared to the jute–WQES (S8) also highlights the contribution of the smoother flax fiber topology, which facilitates better encapsulation by PLA and reduces water-accessible pathways.

Retention rate analysis further validated the impact of moisture on composite durability. The flax-based hybrid systems (S10–S12) retained a greater proportion of their tensile, flexural, and impact strengths after conditioning than the jute hybrids did, indicating that the flax fibers experienced less interfacial degradation under wet conditions. The lower mechanical loss in S12 particularly reflects the barrier effect of micro–nanoscale CaCO_3 domains in the WQES, which minimizes hydrolytic chain scission in the PLA matrix and limits microcrack propagation. Conversely, the higher performance drop observed in jute-based hybrids (especially S7) originates from the swelling-induced debonding at the fiber/matrix interface and the presence of lumen cavities that act as moisture reservoirs. The superior moisture resistance and structural stability of flax-based systems highlight the advantages of flax over jute in applications in which dimensional stability and long-term mechanical retention in humid environments are critical factors.

3.10. Microstructural Characteristics of Hybrid Composites (S10–S12)

Scanning electron microscopy (SEM) analysis of the hybrid composites S10–S12 provides direct microstructural evidence supporting the distinct mechanical, thermal, acoustic, surface, and moisture-related behaviors observed in this study. The three systems exhibited markedly different fracture morphologies and interfacial architectures owing to the contrasting morphology, chemistry, and packing characteristics of the WPNS, WTLF, and WQES fillers when combined with flax fibers in a PLA matrix.

3.10.1. Scheme S10: Flax–WPNS Hybrid Composite

The fracture surface of the flax–WPNS hybrid composite (S10) revealed a moderately compact microstructure characterized by crack deflection around the flax fibers, partial fiber pull-out, and localized WPNS particle agglomeration (Figure 12a–d). The deflection of cracks along the flax fibers indicates effective engagement of the fibers in load transfer, whereas the presence of fiber pull-out rather than clean fiber fracture suggests progressive interfacial debonding accompanied by frictional energy dissipation. These features collectively explain how S10 exhibited the highest tensile and flexural strengths among the hybrid composites. However, the SEM images also revealed interfacial gaps and particle agglomerates, particularly in the WPNS-rich regions. These heterogeneities act as localized stress concentrators, limiting further improvements in impact resistance and thermal transport. From a surface perspective, the relatively good wetting of the WPNS by PLA, assisted by the flax fiber network, reduces extreme surface asperities, resulting in a moderate surface roughness compared with S11. Acoustically, the reduced porosity and tighter packing limit viscous and thermal losses, explaining why S10 exhibits lower sound absorption than S11, despite its superior mechanical performance. S10 represents a mechanically optimized hybrid, where the balance between flax reinforcement and rigid lignocellulosic WPNS particles increases the load-bearing efficiency over multifunctional damping or insulation.

3.10.2. S11: Flax–WTLF Hybrid Composite

In contrast, the flax–WTLF hybrid composite (S11) displayed a distinctly porous and defect-rich microstructure, as evidenced by large interconnected voids, porous WTLF particles, smooth fiber pull-out surfaces, and crack propagation through pore networks (Figure 13a–d). The extensive interfacial voids and limited fiber bridging indicate weaker

fiber–matrix bonding and reduced stress transfer efficiency, which explains the comparatively lower tensile and flexural strengths of S11.

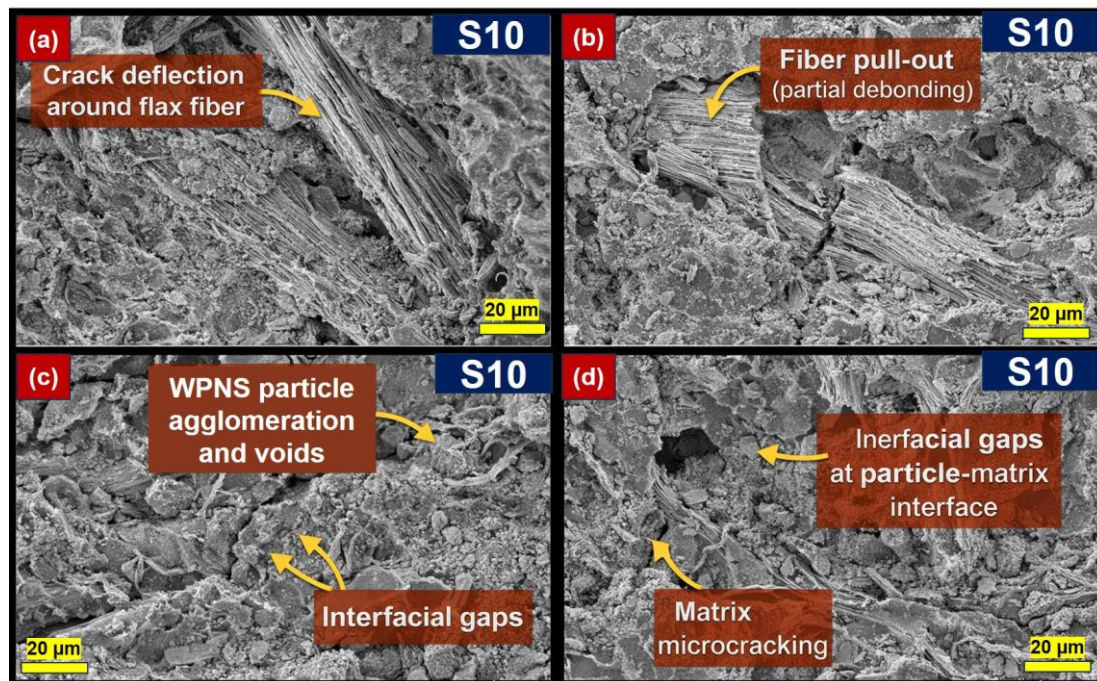


Figure 12. (a–d). SEM micrographs of the flax fiber and WPNS-reinforced PLA hybrid composite (Sample S10).

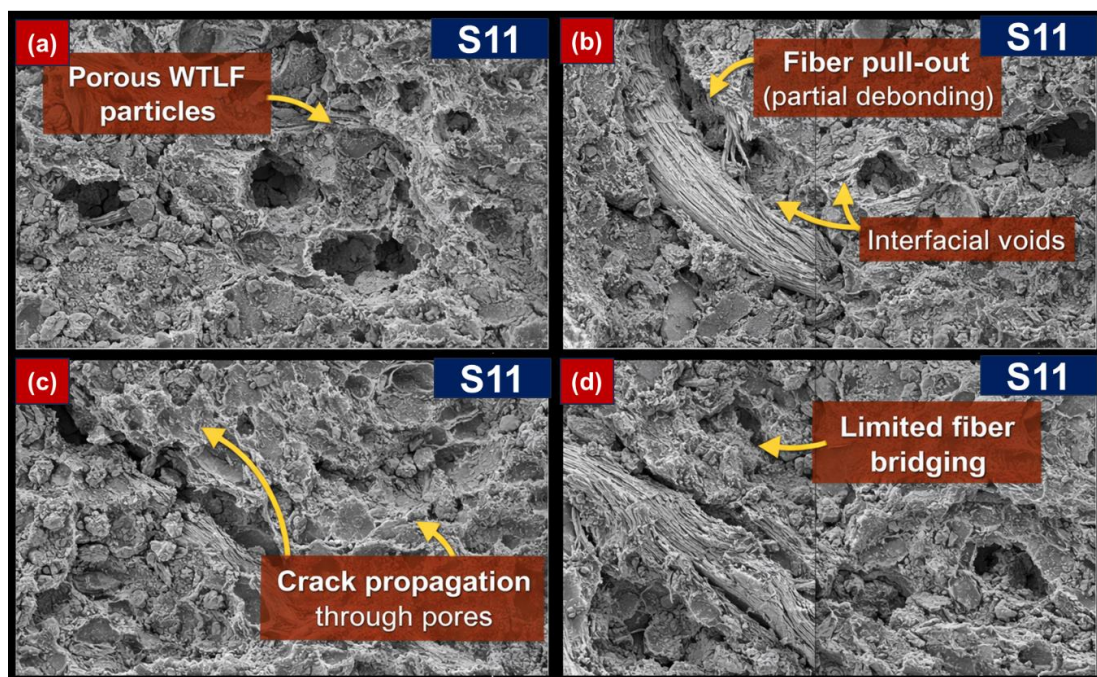


Figure 13. (a–d). SEM micrographs of the flax fiber and WTLF-reinforced PLA hybrid composite (Sample S11).

Despite this mechanical limitation, the same microstructural features are highly advantageous for acoustic performance. The porous architecture and interconnected voids act as effective acoustic dissipation zones, promoting viscous damping, thermal losses, and multipath scattering of sound waves. SEM-confirmed pore connectivity directly explains

why S11 exhibited the highest sound absorption coefficient (SAC) and noise reduction coefficient (NRC) among all composites. From a thermal standpoint, the porous microstructure introduces numerous phonon-scattering sites, reducing effective heat-transfer pathways and resulting in lower thermal conductivity compared to S10 and S12. The rough and irregular surface topology produced by the WTLF particle protrusion is consistent with the highest surface roughness values observed experimentally. Furthermore, the open pore structure provided moisture-accessible pathways, explaining the higher water absorption of S11. Thus, S11 is best described as an acoustically optimized composite, where enhanced damping and absorption arise at the expense of mechanical stiffness and thermal transport efficiency.

3.10.3. S12: Flax–WQES Hybrid Composite

The flax–WQES hybrid composite (S12) exhibited the most compact and homogeneous microstructure among the hybrids. SEM images (Figure 14a–d) reveal strong fiber–matrix interfacial bonding, extensive crack deflection and fiber bridging, and uniformly distributed CaCO_3 particle pull-out cavities. The presence of matrix tearing around the fibers indicates progressive interfacial failure rather than brittle debonding, confirming efficient stress transfer and controlled energy dissipation. The fine, CaCO_3 -rich WQES particles effectively occupied interstitial spaces within the flax fiber network, significantly reducing the void content and enhancing the packing efficiency. This compact microstructure directly explains the highest apparent density and thermal conductivity observed for S12, as the reduced porosity minimizes phonon scattering and lowers the interfacial thermal resistance. The mineral-rich particles formed continuous microscale heat conduction pathways when coupled with the flax fiber skeleton, making S12 a thermally optimized system.

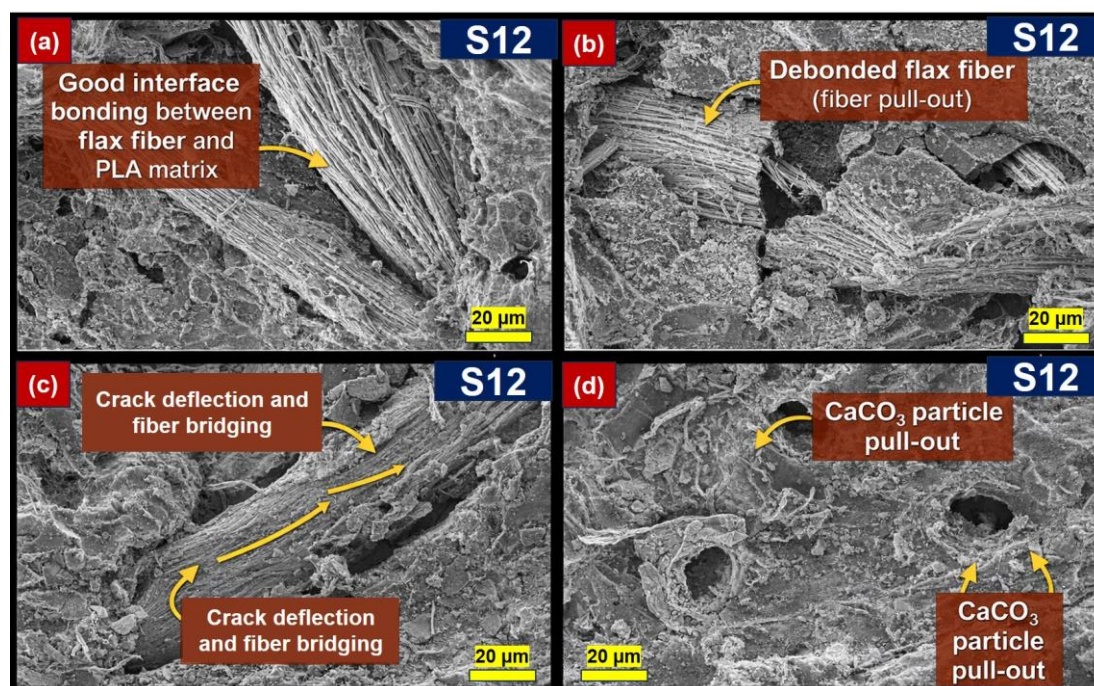


Figure 14. (a–d). SEM micrographs of the flax fiber and WQES-reinforced PLA hybrid composite (Sample S12).

From the perspective of surface and durability, the dense microstructure and inorganic nature of WQES limit surface irregularities and water-accessible pathways, resulting in moderate surface roughness and low water absorption among the hybrids. Acoustically, the reduced porosity restricts viscous dissipation, leading to lower sound absorption than S11, but still higher than that of neat PLA owing to fiber-induced scattering. S12 represents

a structurally compact and thermally efficient hybrid composite, where superior packing and interfacial integrity dominate the acoustic damping.

4. An Integrated Fuzzy Critic-Copras Approach

The evaluation of multi-constituent biocomposite systems, particularly those incorporating natural fibers and heterogeneous agro-waste fillers, requires decision-making tools capable of handling conflicting criteria and inherent uncertainties. Traditional deterministic optimization techniques often fall short when the criteria influencing composite performance, such as tensile strength, flexural modulus, impact toughness, thermal stability, and acoustic damping, are interdependent and exhibit nonlinear relationships. Moreover, expert evaluations of the relative importance of mechanical, thermal, and functional attributes frequently contain vagueness and ambiguity, which cannot be adequately captured using classical crisp-weight assignment methods. To overcome these limitations, fuzzy multi-criteria decision-making (MCDM) models, particularly those integrating objective weighting schemes, have gained prominence in material selection and composite optimization studies.

In this study, a Triangular Fuzzy Weighted Bonferroni Mean Operator (TFWBMO) is employed in conjunction with the Criteria Importance Through Intercriteria Correlation (CRITIC) method to derive objective fuzzy weights that account for both the contrast intensity and the degree of conflict among evaluation metrics. The Bonferroni mean operator provides a flexible aggregation framework capable of modeling the interactions among criteria, enabling the weighting process to account for synergistic or compensatory relationships between the mechanical, thermal, and acoustic properties. The use of triangular fuzzy numbers further enhances the robustness of the model by representing expert judgments with a range rather than a single crisp value, thereby reflecting the uncertainty in composite property assessments.

Following the weighting stage, the Complex Proportional Assessment (COPRAS) method, extended with fuzzy logic, was applied to compute the relative significance of each composite formulation. COPRAS is particularly suitable for material ranking because it simultaneously incorporates both maximizing criteria (e.g., tensile strength, flexural strength, thermal conductivity, and sound absorption) and minimizing criteria (e.g., water absorption and surface roughness) while maintaining proportional relationships among alternatives. Its hybridization with the TFWBMO–CRITIC approach yielded a mathematically rigorous and logically transparent decision-making system capable of ranking flax/PLA composites reinforced with WPNS, WTLF, and WQES.

Thus, this integrated fuzzy CRITIC–COPRAS framework was introduced to address the complex, multiperformance nature of hybrid biocomposites. This ensures the objective weighting of criteria, accommodates uncertainty in expert evaluations, and enables a systematic comparison between the newly developed flax-reinforced composites and previously reported jute-reinforced systems. By capturing the interactions among the properties and reflecting the true performance variations, the proposed mathematical model provides a scientifically grounded tool for identifying the optimal filler configuration and guiding future material designs.

Triangular fuzzy number (TFN)

A TFN $\tilde{a}$ can be illustrated using a triplet (a^l, a^m, a^u) . The membership function $\mu_{\tilde{a}}(\chi)$ is defined in Equation (7),

$$\mu_{\tilde{a}}(x) = \begin{cases} 0, & x < a^l \\ \frac{x-a^l}{a^m-a^l}, & a^l \leq x \leq a^m \\ \frac{x-a^u}{a^m-a^u}, & a^l \leq x \leq a^m \\ 0, & x \geq a^u \end{cases} \quad (7)$$

where $0 < a^l \leq a^m \leq a^u$, a^l and a^u denote the lowest and highest values of the support of, respectively, and a^m symbolizes the mean value.

Basic operational laws related to TFNs

Let $\tilde{a}$ and $\tilde{b}$ be two TFNs. The fundamental expression is as follows:

- (i) $\tilde{a} \oplus \tilde{b} = (a^l, a^m, a^u) \oplus (b^l, b^m, b^u) = (a^l + b^l, a^m + b^m, a^u + b^u)$
- (ii) $\tilde{a} \otimes \tilde{b} = (a^l, a^m, a^u) \otimes (b^l, b^m, b^u) = (a^l b^l, a^m b^m, a^u b^u)$
- (iii) $\lambda \otimes \tilde{a} = \lambda \otimes (a^l, a^m, a^u) = (\lambda a^l, \lambda a^m, \lambda a^u), \lambda > 0$
- (iv) Defuzzification: Center of Area (COA) = $\frac{a^l + a^m + a^u}{3}$
- (v) $\tilde{a}^{-1} = (a^l, a^m, a^u)^{-1} = \left(\frac{1}{a^l}, \frac{1}{a^m}, \frac{1}{a^u}\right)$

Bonferroni Mean Operator (BMO)

The Bonferroni mean operator was developed using Bonferroni. It can provide an aggregation lying among the min, max operators and the logical “or” and “and” operators, and can be defined as in Equation (8),

$$BM^{p,q}(a_1, a_2, \dots, a_n) = \left(\frac{1}{n(n-1)} \sum_{\substack{i,j=1 \\ i \neq j}}^n a_i^p a_j^q \right)^{\frac{1}{p+q}} \quad (8)$$

Let $p, q \geq 0$ be the parameters and a_i ($i = 1, 2, \dots, n$) be a collection of non-negative real numbers. Then, the aggregation function—namely, the Weighted Bonferroni Mean (WBM) Operator—is defined as shown in Equation (9),

$$WBM^{p,q}(a_1, \dots, a_n) = \left(\frac{1}{n(n-1)} \sum_{\substack{i,j=1 \\ i \neq j}}^n (w_i a_i)^p (w_j a_j)^q \right)^{\frac{1}{p+q}} \quad (9)$$

Let $p, q \geq 0$ and a_i be the collection of non-negative numbers. w_i ($i = 1, 2, \dots, n$) is the weight vector of a_i , where w_i denotes the degree of importance of a_i , satisfying $0 < w_i < 1$, ($i = 1, 2, \dots, n$), and $\sum_{i=1}^n w_i = 1$.

Triangular fuzzy Bonferroni mean operator (TFBMO)

BM operators can be adapted to situations involving TFN inputs. The $TFBM^{p,q}$ operator is expressed as shown in Equation (10),

$$TFBM^{p,q}(a_1, a_2, \dots, a_n) = \left[\begin{array}{l} \left(\frac{1}{n(n-1)} \sum_{\substack{i,j=1 \\ i \neq j}}^n (a_i^l)^p (a_j^l)^q \right)^{\frac{1}{p+q}}, \\ \left(\frac{1}{n(n-1)} \sum_{\substack{i,j=1 \\ i \neq j}}^n (a_i^m)^p (a_j^m)^q \right)^{\frac{1}{p+q}}, \\ \left(\frac{1}{n(n-1)} \sum_{\substack{i,j=1 \\ i \neq j}}^n (a_i^u)^p (a_j^u)^q \right)^{\frac{1}{p+q}} \end{array} \right] \quad (10)$$

Let $\tilde{a}_i = (a_i^l, a_i^m, a_i^u)$ ($i = 1, 2, \dots, n$) be a set of TFN and let $p, q > 0$.

Triangular Fuzzy Weighted Bonferroni Mean Operator (TFWBM)

$$TFWBM_{w_i}^{p,q}(\tilde{a}_1, \tilde{a}_2, \dots, \tilde{a}_n) = \left(\frac{1}{n(n-1)} \sum_{\substack{i,j=1 \\ i \neq j}}^n (w_i \tilde{a}_i)^p (w_j \tilde{a}_j)^q \right)^{\frac{1}{p+q}} \quad (11)$$

The TFWBM can be defined as shown in Equation (11).

Let $\tilde{a}_i = [a_i^l, a_i^m, a_i^u]$ ($i = 1, 2, \dots, n$) be a set of sets of TFNs and $p, q > 0$, $w = (w_1, w_2, \dots, w_n)^T$ is the weight vector of $\tilde{a}_i = [a_i^l, a_i^m, a_i^u]$ ($i = 1, 2, \dots, n$), where w_i indicates the importance degree of $\tilde{a}_i$, satisfying $w_i > 0$ ($i = 1, 2, \dots, n$) and $\sum_{i=1}^n w_i = 1$.

4.1. Proposed TFWBMAO-Based Fuzzy CRITIC

CRITIC was proposed by Diakoulaki, Mavrotas, and Papayannakis in 1995 and is a popular method for criteria weighting that eliminates the subjective bias in determining the weights. The criteria weights depend on the data in the decision matrix. This technique maintains conflicts among the criteria and provides unique information. This method is helpful in separating subjective bias through weight determination and capturing the critical aspects of the criteria. This method requires the prior conversion of linguistic data into numerical data; moreover, the weight is affected by the error in the decision matrix. The method is sensitive to data, and errors in the decision matrix can influence the criteria weight. The algorithm of the proposed TFWBMAO-based Fuzzy CRITIC method is as follows:

Suppose there are l DMs symbolized by DM_r , ($r = 1, 2, 3, \dots, l$), m alternatives A_i ($i = 1, 2, 3, \dots, m$) and n evaluation criteria C_j ($j = 1, 2, 3, \dots, n$). The algorithm for the anticipated TFWBMAO-based CRITIC is as follows:

Step 1: Construct the decision matrix

The decision matrix involving the alternatives and attributes as shown in Equation (12),

$$\begin{bmatrix} r_{11} & \dots & r_{1j} & r_{1,j+1} & \dots & r_{1n} \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\ r_{ii} & \dots & r_{ij} & r_{i,j+1} & \dots & r_{in} \\ \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\ r_{m1} & \dots & r_{mj} & r_{j,m+1} & \dots & r_{mn} \end{bmatrix} \quad (12)$$

where $i = 1, 2, \dots, m$; $j = 1, 2, \dots, n$ and $\{A_1, A_1, \dots, A_m\}$ and r_{ij} is the element of the decision matrix for i th alternative and j th attribute.

Step 2: Normalize the decision matrix

Equations (13) and (14) are used to normalize the positive (benefit) and negative (cost) attributes of the decision matrix, respectively.

$$x_{ij} = \frac{r_{ij} - r_i^-}{r_i^+ - r_i^-} \quad i = 1, 2, \dots, m; j = 1, 2, \dots, n \quad (13)$$

$$x_{ij} = \frac{r_{ij} - r_i^+}{r_i^- - r_i^+} \quad i = 1, 2, \dots, m; j = 1, 2, \dots, n \quad (14)$$

where x_{ij} represents the normalized values of the decision matrix for the i th alternative with respect to the j th attribute and $r_i^+ = \max(r_{i1}, r_{i2}, \dots, r_{in})$, $r_i^- = \min(r_{i1}, r_{i2}, \dots, r_{in})$.

Step 3: Obtain the correlation coefficients

The correlation coefficient ρ_{jk} between attributes is determined by Equation (15),

$$\rho_{jk} = \frac{\sum_{i=1}^m (x_{ij} - \bar{x}_j)(x_{ik} - \bar{x}_k)}{\sqrt{\sum_{i=1}^m (x_{ij} - \bar{x}_j)^2 \sum_{i=1}^m (x_{ik} - \bar{x}_k)^2}} \quad (15)$$

Step 4: Calculate the sample standard deviation

The sample standard deviation of each attribute is calculated using Equation (17),

$$\sigma_j = \sqrt{\frac{1}{n-1} \sum_{i=1}^n (x_{ij} - \bar{x}_j)^2}, i = 1, 2, \dots, m \quad (17)$$

Step 5: Obtain the index (C_j)

Then, the index (C_j) is calculated using Equation (18),

$$C_j = \sigma_j \sum_{k=1}^n (1 - \rho_{jk}), j = 1, 2, \dots, n \quad (18)$$

Step 6: Calculate the weights of attributes w_j

The weights of the attributes are calculated using Equation (19),

$$w_j = \frac{C_j}{\sum_{j=1}^n C_j} \quad (19)$$

4.2. TFWBMO Based Fuzzy COPRAS

The COPRAS method was introduced by Zavadskas and Kaklauskas. COPRAS is a prominent multi-criteria decision-making technique that can effectively rank alternatives. This method is suitable for comparing different criteria by selecting them on a similar scale. This method depends on assessing the superiority of a single alternative over other alternatives through the rating and evaluation processes. It follows a systematic approach, such as matrix creation, conversion in a single scale, multiplication of the scaled value with the assigned weight, calculation of the summed weighted normalized values for useful and non-useful criteria, and ranking of alternatives. This method can be used for various applications, such as project evaluation, material selection, and environmental management. The COPRAS method considers both beneficial and non-beneficial aspects and is easily applicable. This method reduces subjectivity in the decision-making process. It is a versatile method applicable to different domains such as engineering, business, and environmental evaluation. The method has the limitation of changing the ranking with input data; in addition, the output depends on the assignment of weight to criteria that need expert opinion and can introduce some subjectivity. The implementation of triangular fuzzy numbers allows the incorporation of linguistic terms and uncertainties into the decision-making process. The algorithm of the proposed TFWBMO-based fuzzy COPRAS is described as follows:

Step 1: Formulate the triangular fuzzy decision matrix $\tilde{R}^r$, proposed by the DMs, as shown in Equation (20),

$$\tilde{R}^r = \begin{bmatrix} A_1^r & C_1^r & C_2^r & \cdots & C_n^r \\ \tilde{a}_{11}^r & \tilde{a}_{12}^r & \ddots & \tilde{a}_{1n}^r \\ \cdots & \cdots & \cdots & \cdots \\ A_m^r & \tilde{a}_{m1}^r & \tilde{a}_{m2}^r & \cdots & \tilde{a}_{mn}^r \end{bmatrix} \quad (20)$$

Step 2: Find the aggregated decision matrix using the TFWBM operator by allocating different weights to each DM as shown in Equation (21),

$$\tilde{R} = \begin{bmatrix} C_1 & C_2 & \cdots & C_n \\ A_1 & \tilde{a}_{11} & \tilde{a}_{12} & \ddots & \tilde{a}_{1n} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ A_m & \tilde{a}_{m1} & \tilde{a}_{m2} & \cdots & \tilde{a}_{mn} \end{bmatrix} \quad (21)$$

Step 3: Normalization of the fuzzy decision matrix using Equation (22),

$$\bar{R}_{ij} = \frac{R_{ij}}{\sum_{i=1}^m R_{ij}}; i = 1, 2, 3, \dots, m; j = 1, 2, 3, \dots, n \quad (22)$$

Step 4: Obtain the weighted normalized decision-making matrix $\hat{R}$ by multiplying the weights to the normalized decision matrix using Equation (23),

$$\hat{R}_{ij} = \bar{R}_{ij} \cdot w_j; i = 1, 2, 3, \dots, m; j = 1, 2, 3, \dots, n \quad (23)$$

Step 5: Compute the sum of criteria for that the optimum value is most preferable for each alternative using Equation (24),

$$\tilde{P}_j = \sum_{i=1}^m \tilde{x}_{ij} \quad (24)$$

Step 6: Calculate the sum of the criteria for the minimum value is most preferable for each alternative using Equation (25),

$$\tilde{R}_j = \sum_{i=k+1}^m \tilde{x}_{ij} \quad (25)$$

Step 7: Evaluate the minimum value of $\tilde{R}_j$ and $\tilde{R}_{\min}$ using Equation (26),

$$\tilde{R}_{\min} = \min_j \tilde{R}_j; j = 1, 2, \dots, n \quad (26)$$

Step 8: Obtain the relative significance of each alternative using Equation (27),

$$\tilde{Q}_j = \tilde{P}_j + \frac{\tilde{R}_{\min} \sum_{j=1}^n \tilde{R}_j}{\tilde{R}_j \sum_{j=1}^n \tilde{R}_{\min}}; j = 1, 2, \dots, n \quad (27)$$

Step 9: Determine the value of the optimality criterion K using Equation (28),

$$K = \max_j \tilde{Q}_j; j = 1, 2, \dots, n \quad (28)$$

Step 10: Determine the priority. Greater value of Q_j for the alternative j indicates a higher priority of the alternatives.

Step 11: Estimate the utility degree of the alternatives using Equation (29),

$$N_j = \frac{Q_j}{Q_{\max}} \times 100; j = 1, 2, \dots, n \quad (29)$$

4.3. Application of the Proposed TFWBMAO-Based Fuzzy CRITIC-COPRAS Approach

In this study, the Triangular Fuzzy Weighted Bonferroni Mean Aggregation Operator (TFWBMAO) was integrated with the CRITIC–COPRAS multi-criteria decision-making framework to systematically rank the 12 composite alternatives developed for structural and acoustical applications. The complete sequence of the adopted methodology is shown in Figure 15.

Expert judgments were obtained from three decision makers (DM1, DM2, and DM3), all of whom possessed substantial research experience in natural fiber-polymer composites and the functional roles of biofillers. Their evaluations were gathered through a structured questionnaire designed to identify the most suitable composite formulation

from the 12 alternatives (S1–S12) based on eight performance criteria, as summarized in Tables 3 and 4. Figure 16 shows the decision sequence for the material selection problem.

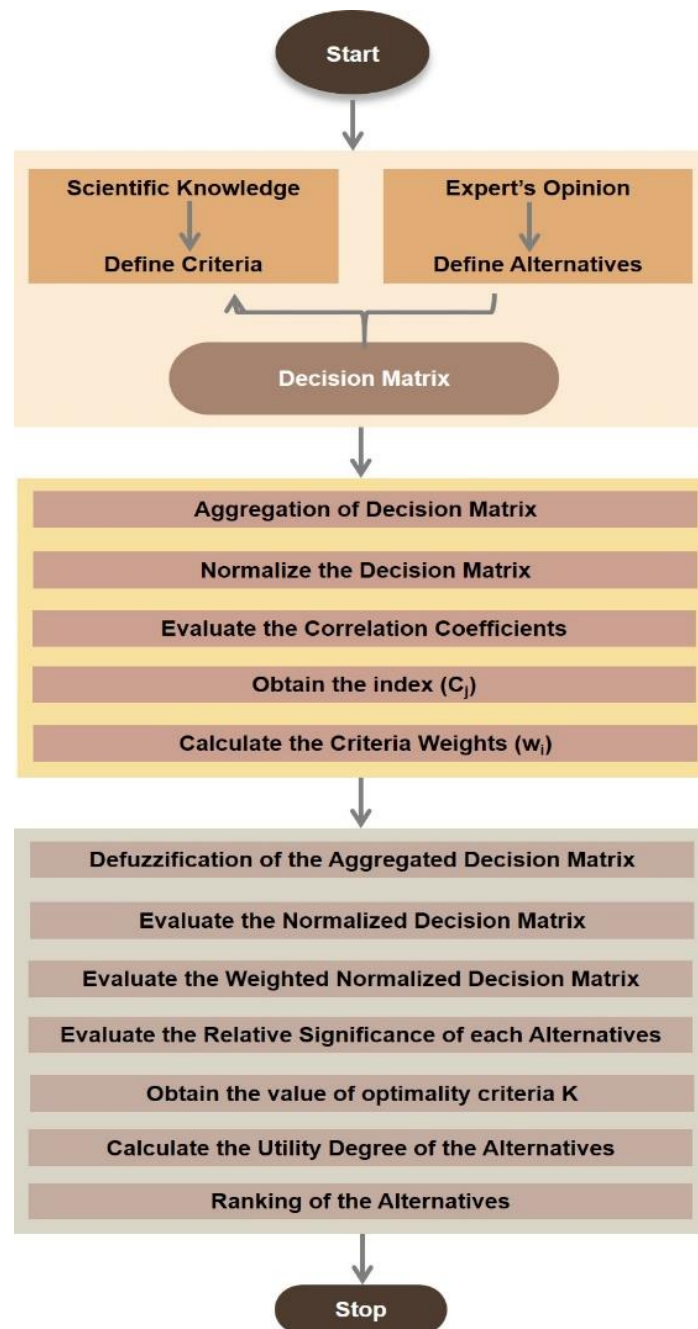


Figure 15. Fuzzy CRITIC–COPRAS model for composite ranking.

To ensure fairness and avoid bias, equal importance was assigned to all decision makers, considering their similar technical expertise. Each DM expressed their evaluation using linguistic rating terms, reflecting the relative significance of each criterion in the context of the structural strength, durability, sound absorption, and thermal characteristics of the flax- and jute-reinforced PLA systems. These linguistic inputs were then translated into triangular fuzzy numbers (TFNs) for further processing. Table 5 presents the triangular fuzzy number based linguistic term, and Table 6 shows the linguistic valuation of the DMs for alternatives.

Table 3. Compositions and properties of the alternatives.

Alternative	Density (g/cm ³)	Tensile Strength (MPa)	Flexural Strength (MPa)	Impact Strength (kJ/m ²)	Hardness (Shore D)	Noise Reduction Coefficient (NRC)	Thermal Conductivity (W/(m.K))	Surface Roughness (mm)
S1	1.32	21.74	48.76	1.12	77.28	0.207	0.163	0.021
S2	1.39	42.13	86.04	3.23	79.55	0.315	0.247	0.025
S3	1.63	37.91	77.29	2.47	83.71	0.404	0.374	0.044
S4	1.46	30.68	60.24	2.96	80.35	0.583	0.298	0.076
S5	1.88	25.89	54.89	2.03	86.82	0.487	0.462	0.065
S6	1.73	57.38	114.67	4.02	84.43	0.462	0.418	0.032
S7	1.57	51.82	95.59	4.35	81.94	0.618	0.315	0.072
S8	2.09	48.27	88.27	3.74	87.12	0.539	0.509	0.051
S9	1.86	48.25	89.23	3.56	81.15	0.346	0.265	0.028
S10	2.09	60.41	118.71	4.75	86.12	0.501	0.457	0.038
S11	1.83	56.27	100.32	5.04	83.58	0.695	0.352	0.079
S12	2.24	52.36	94.18	4.13	88.87	0.597	0.538	0.067

Table 4. Details of the criteria.

Criteria	Property	Details	Attributes
C1	Density	Influences the weight of the structural materials and stability	Beneficial
C2	Tensile strength	Determine the maximum stress against pulling or stretching	Beneficial
C3	Flexural strength	Determine the capacity to withstand external bending load	Beneficial
C4	Impact strength	Determine the capacity to resist sudden loading	Beneficial
C5	Hardness	Determine the long-term durability and structural integrity	Beneficial
C6	Noise Reduction Coefficient	Determine the capacity of the materials to absorb sound	Beneficial
C7	Thermal Conductivity	Determine the ability of the material to withstand the heat	Non-Beneficial
C8	Surface Roughness (μm)	Influence the adhesion/bonding nature of the filler and fiber in composites	Non-Beneficial

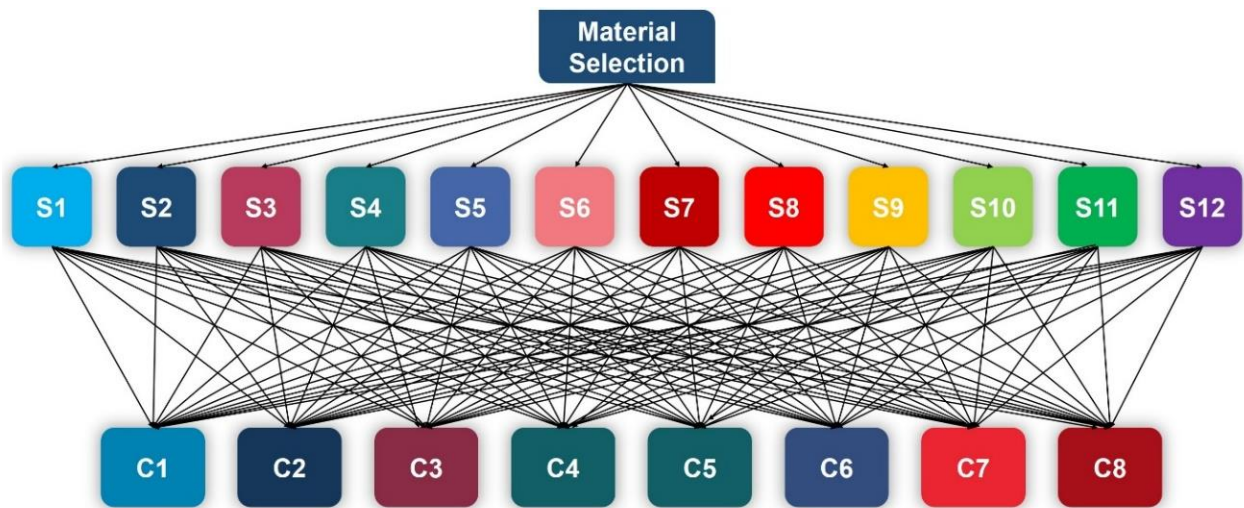


Figure 16. Material selection network showing relationships between samples and criteria.

Table 5. Triangular fuzzy number based linguistic term.

Linguistic Assessment	Linguistic Value	Triangular Fuzzy Set
Very low	VL	(2,3,4)
Low	L	(3,4,5)
Moderate	M	(6,7,8)
High	H	(7,8,9)
Very high	VH	(8,9,9)

Table 6. Linguistic valuation of the DMs for alternatives.

Alternates	DMs	C1	C2	C3	C4	C5	C6	C7	C8
S1	DM1	(3,4,5)	(2,3,4)	(2,3,4)	(2,3,4)	(3,4,5)	(3,4,5)	(3,4,5)	(2,3,4)
	DM2	(3,4,5)	(2,3,4)	(2,3,4)	(2,3,4)	(3,4,5)	(3,4,5)	(3,4,5)	(2,3,4)
	DM3	(3,4,5)	(2,3,4)	(2,3,4)	(2,3,4)	(3,4,5)	(2,3,4)	(3,4,5)	(2,3,4)
S2	DM1	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(3,4,5)
	DM2	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(3,4,5)	(3,4,5)	(3,4,5)
	DM3	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(3,4,5)	(6,7,8)	(3,4,5)	(3,4,5)
S3	DM1	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(7,8,9)	(6,7,8)	(6,7,8)	(6,7,8)
	DM2	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(7,8,9)	(3,4,5)	(6,7,8)	(6,7,8)
	DM3	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)
S4	DM1	(6,7,8)	(3,4,5)	(3,4,5)	(6,7,8)	(6,7,8)	(7,8,9)	(6,7,8)	(7,8,9)
	DM2	(6,7,8)	(3,4,5)	(3,4,5)	(6,7,8)	(6,7,8)	(7,8,9)	(3,4,5)	(7,8,9)
	DM3	(6,7,8)	(3,4,5)	(3,4,5)	(6,7,8)	(3,4,5)	(7,8,9)	(3,4,5)	(7,8,9)
S5	DM1	(7,8,9)	(3,4,5)	(3,4,5)	(3,4,5)	(7,8,9)	(6,7,8)	(7,8,9)	(6,7,8)
	DM2	(7,8,9)	(3,4,5)	(3,4,5)	(3,4,5)	(7,8,9)	(6,7,8)	(7,8,9)	(6,7,8)
	DM3	(7,8,9)	(3,4,5)	(3,4,5)	(3,4,5)	(6,7,8)	(6,7,8)	(7,8,9)	(6,7,8)
S6	DM1	(7,8,9)	(7,8,9)	(8,9,9)	(7,8,9)	(7,8,9)	(7,8,9)	(7,8,9)	(6,7,8)
	DM2	(7,8,9)	(7,8,9)	(8,9,9)	(7,8,9)	(7,8,9)	(6,7,8)	(7,8,9)	(6,7,8)
	DM3	(7,8,9)	(7,8,9)	(8,9,9)	(7,8,9)	(6,7,8)	(7,8,9)	(7,8,9)	(6,7,8)
S7	DM1	(6,7,8)	(7,8,9)	(7,8,9)	(8,9,9)	(7,8,9)	(8,9,9)	(6,7,8)	(7,8,9)
	DM2	(6,7,8)	(7,8,9)	(7,8,9)	(8,9,9)	(7,8,9)	(7,8,9)	(6,7,8)	(7,8,9)
	DM3	(6,7,8)	(7,8,9)	(7,8,9)	(8,9,9)	(6,7,8)	(8,9,9)	(6,7,8)	(7,8,9)
S8	DM1	(7,8,9)	(6,7,8)	(7,8,9)	(7,8,9)	(8,9,9)	(7,8,9)	(8,9,9)	(6,7,8)
	DM2	(7,8,9)	(6,7,8)	(7,8,9)	(7,8,9)	(8,9,9)	(6,7,8)	(8,9,9)	(6,7,8)
	DM3	(7,8,9)	(6,7,8)	(7,8,9)	(7,8,9)	(7,8,9)	(7,8,9)	(8,9,9)	(6,7,8)
S9	DM1	(7,8,9)	(6,7,8)	(7,8,9)	(6,7,8)	(7,8,9)	(6,7,8)	(6,7,8)	(3,4,5)
	DM2	(7,8,9)	(6,7,8)	(7,8,9)	(6,7,8)	(7,8,9)	(6,7,8)	(3,4,5)	(3,4,5)
	DM3	(7,8,9)	(6,7,8)	(7,8,9)	(6,7,8)	(6,7,8)	(6,7,8)	(6,7,8)	(3,4,5)
S10	DM1	(8,9,9)	(8,9,9)	(8,9,9)	(8,9,9)	(7,8,9)	(7,8,9)	(7,8,9)	(6,7,8)
	DM2	(8,9,9)	(8,9,9)	(8,9,9)	(8,9,9)	(7,8,9)	(6,7,8)	(7,8,9)	(6,7,8)
	DM3	(8,9,9)	(8,9,9)	(8,9,9)	(8,9,9)	(7,8,9)	(7,8,9)	(7,8,9)	(6,7,8)
S11	DM1	(7,8,9)	(7,8,9)	(7,8,9)	(8,9,9)	(7,8,9)	(8,9,9)	(6,7,8)	(7,8,9)
	DM2	(7,8,9)	(7,8,9)	(7,8,9)	(8,9,9)	(7,8,9)	(7,8,9)	(6,7,8)	(7,8,9)
	DM3	(7,8,9)	(7,8,9)	(7,8,9)	(8,9,9)	(6,7,8)	(8,9,9)	(6,7,8)	(7,8,9)
S12	DM1	(8,9,9)	(7,8,9)	(7,8,9)	(7,8,9)	(8,9,9)	(7,8,9)	(8,9,9)	(6,7,8)
	DM2	(8,9,9)	(7,8,9)	(7,8,9)	(7,8,9)	(8,9,9)	(6,7,8)	(8,9,9)	(6,7,8)
	DM3	(8,9,9)	(7,8,9)	(7,8,9)	(7,8,9)	(7,8,9)	(7,8,9)	(8,9,9)	(6,7,8)

The combined linguistic assessments for all composite alternatives, expressed as TFNs, are presented in Table 7. This structured fuzzy decision-making approach ensures that subjective assessments are rigorously quantified, thus enabling the accurate extraction of objective criteria weights through CRITIC and the robust ranking of composite alternatives via COPRAS.

Table 7. Assessment of the DMs for the criteria.

Criteria	DM1 (TFN)	DM2 (TFN)	DM3 (TFN)
C1—Density	(6,7,8)	(7,8,9)	(6,7,8)
C2—Tensile Strength	(7,8,9)	(8,9,9)	(7,8,9)
C3—Flexural Strength	(7,8,9)	(7,8,9)	(8,9,9)
C4—Impact Strength	(6,7,8)	(7,8,9)	(7,8,9)
C5—Hardness	(6,7,8)	(6,7,8)	(7,8,9)
C6—Noise Reduction Coefficient	(7,8,9)	(8,9,9)	(7,8,9)

Table 7. Cont.

Criteria	DM1 (TFN)	DM2 (TFN)	DM3 (TFN)
C7—Thermal Conductivity	(3,4,5)	(2,3,4)	(3,4,5)
C8—Surface Roughness	(3,4,5)	(3,4,5)	(2,3,4)

4.4. Algorithm of the Proposed TFWBMO-Based CRITIC–COPRAS Method

The procedural workflow of the Triangular Fuzzy Weighted Bonferroni Mean Operator (TFWBMO)-integrated CRITIC–COPRAS model used for ranking the flax- and jute-based PLA biocomposites is outlined below:

Step 1: Construct the aggregated fuzzy decision matrix using TFWBMO, which synthesizes the individual evaluations of all decision makers into a consolidated triangular fuzzy matrix. The resulting aggregated assessments are listed in Table 7.

Step 2: Normalize the aggregated matrix to enable objective comparison among the criteria. The beneficial and non-beneficial criteria were normalized using Equations (13) and (14), respectively, and the resulting normalized values are listed in Table 8.

Table 8. Normalized Decision matrix for criteria weights.

Alt	Density (C1)	Tensile (C2)	Flexural (C3)	Impact (C4)	Hardness (C5)	NRC (C6)	Thermal (C7)	Roughness (C8)
S1	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
S2	0.034	0.527	0.533	0.538	0.196	0.221	0.224	0.069
S3	0.151	0.418	0.408	0.344	0.555	0.404	0.563	0.397
S4	0.068	0.231	0.164	0.469	0.265	0.771	0.360	0.948
S5	0.273	0.107	0.088	0.232	0.823	0.574	0.797	0.759
S6	0.200	0.922	0.943	0.740	0.617	0.522	0.680	0.190
S7	0.122	0.778	0.669	0.824	0.402	0.842	0.405	0.879
S8	0.376	0.686	0.565	0.668	0.849	0.681	0.923	0.517
S9	0.264	0.685	0.579	0.622	0.334	0.285	0.272	0.121
S10	0.776	1.000	1.000	0.926	0.763	0.602	0.784	0.293
S11	0.605	0.893	0.737	1.000	0.543	1.000	0.504	1.000
S12	1.000	0.792	0.649	0.768	1.000	0.799	1.000	0.793

Step 3: Determine the correlation coefficients (ρ_{jk}) between each pair of criteria by using Equation (15). These coefficients, reported in Table 9, quantify the degree of interdependence between the performance indicators.

Table 9. Correlations coefficients between the attributes.

Criteria	C1	C2	C3	C4	C5	C6	C7	C8
C1—Density	1	0.95	0.96	0.89	0.84	0.56	0.98	0.72
C2—Tensile Strength	0.95	1	0.99	0.90	0.82	0.45	0.94	0.69
C3—Flexural Strength	0.96	0.99	1	0.93	0.89	0.48	0.96	0.71
C4—Impact Strength	0.89	0.90	0.93	1	0.75	0.62	0.88	0.81
C5—Hardness	0.84	0.82	0.89	0.75	1	0.40	0.87	0.77
C6—NRC	0.56	0.45	0.48	0.62	0.40	1	0.55	0.92
C7—Thermal Conductivity	0.98	0.94	0.96	0.88	0.87	0.55	1	0.74
C8—Surface Roughness	0.72	0.69	0.71	0.81	0.77	0.92	0.74	1

Step 4: Compute the standard deviation (σ_j) for each criterion using Equation (17). These values, listed in Table 10, represent the contrast intensities of each attribute.

Table 10. Standard deviations of the normalized attributes.

Criteria	Standard Deviation (σ)
C1—Density	0.311
C2—Tensile Strength	0.242
C3—Flexural Strength	0.255
C4—Impact Strength	0.262
C5—Hardness	0.106
C6—NRC	0.210
C7—Thermal Conductivity	0.236
C8—Surface Roughness	0.247

Step 5: Calculate the CRITIC-derived informational index (C_j) for each criterion using Equation (18), which combines the standard deviation and correlation structure.

Step 6: Derive the objective weights (w_j) for all criteria using Equation (19). The resulting criteria weights, presented in Table 11, reflect the relative significance of each performance indicator based solely on the data structure.

Table 11. Weights of the attributes.

Criteria	Weight (W_j)
C1—Density	0.102
C2—Tensile Strength	0.158
C3—Flexural Strength	0.173
C4—Impact Strength	0.162
C5—Hardness	0.121
C6—NRC	0.134
C7—Thermal Conductivity	0.083
C8—Surface Roughness	0.067

Step 7: Defuzzify the aggregated fuzzy matrix to obtain crisp numerical scores for all alternatives using relevant defuzzification rules. Table 12 presents the defuzzified decision matrix.

Table 12. Defuzzified Aggregated Decision matrix.

Alt	C1	C2	C3	C4	C5	C6	C7	C8
S1	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
S2	0.0075	0.0970	0.1248	0.0810	0.0086	0.0215	0.0165	0.0036
S3	0.0263	0.0661	0.0973	0.0474	0.0155	0.0456	0.0461	0.0190
S4	0.0113	0.0291	0.0337	0.0664	0.0058	0.1004	0.0291	0.0385
S5	0.0452	0.0139	0.0199	0.0299	0.0130	0.0716	0.0717	0.0282
S6	0.0344	0.1580	0.1567	0.1267	0.0166	0.0570	0.0536	0.0086
S7	0.0204	0.1247	0.1143	0.1550	0.0068	0.1390	0.0307	0.0355
S8	0.0584	0.1109	0.1068	0.1123	0.0186	0.1293	0.0829	0.0224
S9	0.0433	0.1108	0.1095	0.1009	0.0043	0.0532	0.0133	0.0055
S10	0.1790	0.1776	0.1703	0.1620	0.0140	0.0665	0.0664	0.0137
S11	0.1313	0.1516	0.1278	0.1760	0.0153	0.1502	0.0385	0.0420
S12	0.2220	0.1317	0.1112	0.1329	0.0192	0.1260	0.0864	0.0318

Step 8: Normalize the defuzzified decision matrix according to Equation (22) to form the COPRAS-ready normalized matrix reported in Table 13.

Table 13. Normalized Decision matrix.

Alt	C1	C2	C3	C4	C5	C6	C7	C8
S1	0.392	0.360	0.411	0.222	0.870	0.298	0.303	0.266
S2	0.412	0.697	0.725	0.641	0.895	0.453	0.459	0.316

Table 13. Cont.

Alt	C1	C2	C3	C4	C5	C6	C7	C8
S3	0.484	0.628	0.651	0.490	0.942	0.581	0.695	0.557
S4	0.433	0.508	0.508	0.587	0.904	0.839	0.554	0.962
S5	0.558	0.428	0.463	0.403	0.977	0.701	0.858	0.823
S6	0.513	0.949	0.966	0.798	0.950	0.665	0.777	0.405
S7	0.466	0.858	0.806	0.863	0.922	0.889	0.586	0.911
S8	0.620	0.799	0.744	0.742	0.980	0.776	0.946	0.646
S9	0.552	0.798	0.752	0.706	0.914	0.498	0.492	0.354
S10	0.863	1.000	1.000	0.942	0.969	0.721	0.849	0.481
S11	0.760	0.931	0.845	1.000	0.940	1.000	0.654	1.000
S12	1.000	0.867	0.793	0.819	1.000	0.860	1.000	0.848

Step 9: Develop the weighted normalized matrix by multiplying the normalized values with their respective objective weights using Equation (23). The resulting matrices are listed in Table 14.

Table 14. Weighted Normalized Decision matrix.

Alt	C1	C2	C3	C4	C5	C6	C7	C8
S1	0.102	0.158	0.173	0.162	0.121	0.134	0.083	0.067
S2	0.1074	0.3062	0.3052	0.4671	0.1246	0.2040	0.1260	0.0798
S3	0.1259	0.2754	0.2743	0.3578	0.1310	0.2614	0.1903	0.1404
S4	0.1128	0.2220	0.2139	0.4286	0.1256	0.3771	0.1517	0.2427
S5	0.1454	0.1874	0.1950	0.2936	0.1359	0.3154	0.2350	0.2076
S6	0.1337	0.4149	0.4070	0.5829	0.1322	0.2990	0.2114	0.1022
S7	0.1212	0.3746	0.3392	0.6300	0.1283	0.4001	0.1593	0.2300
S8	0.1616	0.3492	0.3129	0.5417	0.1367	0.4089	0.2570	0.1630
S9	0.1439	0.3490	0.3165	0.5153	0.1271	0.2776	0.1338	0.0895
S10	0.2250	0.4374	0.4217	0.6875	0.1350	0.4027	0.2307	0.1216
S11	0.1977	0.4073	0.3565	0.7281	0.1313	0.5588	0.1778	0.2527
S12	0.2596	0.3780	0.3347	0.5962	0.1396	0.4801	0.2716	0.2143

Step 10: Determine the relative priority of each composite sample by calculating the significance index using Equation (27), which aggregates the contributions of the beneficial and non-beneficial criteria.

Step 11: Compute the utility degree of each alternative using Equation (29). The utility scores and final rankings of all filler-based natural fiber composite formulations are summarized in Table 15.

Table 15. Ranking of the alternatives.

Alt	Utility Degree N_j	Rank
S10	1.000	1
S11	0.957	2
S6	0.912	3
S12	0.893	4
S7	0.871	5
S8	0.828	6
S9	0.799	7
S3	0.768	8
S4	0.731	9
S5	0.708	10
S2	0.665	11
S1	0.583	12

Table 16 presents a comparative ranking of the newly developed flax-reinforced composites (S10–S12) against previously reported jute-based hybrids (S6–S8). The results

clearly show that flax-based systems outperform their jute counterparts in terms of overall performance. Among all hybrids, S10 (Flax + WPNS) achieved the highest rank, followed by S11 (Flax + WTLF), reflecting the high reinforcing efficiency of flax when combined with lignocellulosic filler. Although S12 (Flax + WQES) was ranked fourth, it surpassed the corresponding jute-based formulation. In contrast, the jute hybrids S6–S8 ranked lower positions in the ranking, indicating comparatively reduced mechanical, thermal, and acoustical properties. This step confirmed that flax is the superior primary reinforcement for optimizing the PLA-based hybrid composites.

Table 16. Comparison with Jute-based composites (S6–S8).

Hybrid	Fiber	Rank
S10	Flax + WPNS	1
S11	Flax + WTLF	2
S12	Flax + WQES	4
S6	Jute + WPNS	3
S7	Jute + WTLF	5
S8	Jute + WQES	6

4.5. Comparative Study of Jute-Based (S6–S8) and Flax-Based (S10–S12) Hybrid Composites

A comparative assessment of the jute-based hybrid composites (S6–S8) and the newly developed flax-based hybrids (S10–S12) clearly demonstrated the superior reinforcing capability of flax for all the three filler systems. When WPNS was used as the secondary reinforcement, the transition from jute (S6) to flax (S10) resulted in a 5.28% increase in tensile strength, a 3.52% improvement in flexural strength, and a substantial 18.16% increase in impact strength. The hardness increased by 2%, and the noise reduction coefficient and thermal conductivity improved by 8.44% and 9.33%, respectively, highlighting the enhanced structural compactness and acoustic damping of the flax–WPNS hybrids. Similarly, for the WTLF-filled systems, replacing jute (S7) with flax (S11) yielded notable gains, including an 8.59% increase in tensile strength, a 4.95% improvement in flexural strength, and a significant 15.86% increase in impact strength. In addition, the NRC and thermal conductivity improved by 12.46% and 11.75%, respectively, further reinforcing the ability of flax to form more efficient energy-dissipating and thermally conductive networks within the PLA matrix. For the WQES composites, the flax-based system (S12) outperformed its jute counterpart (S8) with increases of 8.47% in tensile strength, 6.69% in flexural strength, and 10.43% in impact strength, accompanied by a 10.76% improvement in NRC and a 5.69% increase in thermal conductivity. Across all comparisons, the hardness improved consistently by approximately 2%, whereas the surface roughness showed filler-dependent increases owing to differences in the fiber–matrix interfacial behavior. The percentage improvements achieved by S10–S12 confirm that flax provides a more robust reinforcement platform than jute, enhancing the mechanical performance, acoustic efficiency, and thermal responsiveness, establishing flax as a superior fiber for developing high-performance PLA-based hybrid biocomposites.

4.6. Comparative Analysis with Reported PLA Composites Reinforced with Waste-Derived and Natural Fillers

To contextualize the mechanical performance of flax-reinforced PLA hybrid composites, a comparative analysis with recently reported PLA composites reinforced with natural fibers and waste-derived fillers is presented in Table 17. This comparison highlights the performance gains achieved through fiber–filler hybridization and clarifies the advantages of the proposed systems over filler-only and single-fiber-reinforced PLA composites reported in the literature. Compared with recent PLA composites reinforced solely with agro-waste

or animal-waste fillers, the present flax-based hybrid systems achieve substantially higher tensile and flexural performance while enabling multifunctional behavior. Filler-only systems typically show modest strength gains ($\leq 15\%$) or even mechanical degradation at higher loadings, whereas the introduction of flax fibers enables efficient stress transfer and crack deflection. The results demonstrate that hybrid fiber–filler architectures are essential for overcoming the intrinsic limitations of waste-derived particulate reinforcements, particularly when mechanical integrity is required alongside sustainability.

Table 17. Comparison of mechanical properties of the present flax/PLA hybrid composites with reported PLA composites.

Reinforcement/Filler	Processing Route	Tensile Strength (MPa)	Flexural Strength (MPa)	Key Outcome	Advancement Highlighted	References
Flax fiber + WPNS/WTLF/WQES (hybrid)	Compression molding	65–70 (S10)	120–125 (S10)	Multifunctional optimization	Highest tensile–flexural balance with waste fillers; property-specific optimization (mechanical, thermal, acoustic)	Present study
Jute fiber + WPNS/WTLF/WQES		57	115	Mechanical improvement vs. PLA	Flax substitution yields higher strength and better consolidation	Previous study [36]
Chicken feathers/fish scales/seashells		+4.1% (CF)	+10.2% (FS)	Waste valorization	Present work achieves much larger strength gains via fiber–filler synergy	[70]
Pomegranate peel biofiller (15 wt. %)		+13.6%	–	Biofiller efficiency	Hybridization with flax provides far greater mechanical enhancement	[71]
Quinoa husk (0–30 wt. %)	Melt compounding	19 MPa (20 wt%)	27 MPa	Cost reduction, sustainability	Flax hybrids overcome strength degradation common in filler-only systems	[72]
Rapeseed straw	Extrusion + injection	45–50	90–100	Modulus increase	Hybrid fiber–filler architecture mitigates strength loss	[73]
Rapeseed straw/flaxseed meal		48	95	Sustainability	SEM-supported superior interfacial bonding in present study	[74]
Eggshell/walnut shell/coffee waste		+72% vs. PLA	110	Aging-sensitive	Fiber reinforcement stabilizes performance across properties	[75]

5. Conclusions

This study examined flax fiber–reinforced PLA hybrid biocomposites containing WPNS, WTLF, and WQES, with the explicit aim of assessing whether flax-based systems offer performance advantages over previously reported jute-based composites. These results confirm that flax fibers provide a more effective reinforcement platform for PLA when combined with appropriately selected waste-derived fillers. Among the hybrid systems, the flax–WPNS composite (S10) exhibited the most favorable mechanical performance, achieving tensile and flexural strength improvements of approximately 30–40% relative to neat PLA. The flax–WTLF composite (S11) demonstrated the highest acoustic performance, reaching a maximum sound absorption coefficient of approximately 0.65–0.70 due to its porous structure. In contrast, the flax–WQES composite (S12) showed the highest apparent density (2.24 g/cm^3) and thermal conductivity (0.54 W/(mK)), reflecting enhanced consolidation and mineral-rich filler effects. The surface roughness and water

absorption trends were similarly governed by the filler morphology, with WTLF increasing the surface irregularity and moisture uptake, whereas WQES improved the dimensional stability. SEM analysis revealed that the observed property differences were controlled by distinct microstructural mechanisms rather than the reinforcement content alone. In S10, crack deflection, partial fiber pull-out, and moderate particle dispersion promoted efficient stress transfer, explaining its superior tensile behavior. In S11, interconnected porosity, interfacial voids, and limited fiber bridging enhance acoustic energy dissipation but reduce mechanical stiffness. In S12, strong fiber–matrix adhesion, homogeneous dispersion of fine CaCO₃-rich particles, reduced void content, and efficient packing enable improved thermal transport and structural compactness. These observations clarify that claims such as “good interfacial bonding” and “reduced voids” are property-specific outcomes rather than universal characteristics. The fuzzy CRITIC–COPRAS approach was employed as a decision-support tool to balance the competing mechanical, thermal, acoustic, and physical properties. Rather than serving as an independent objective, the model provides practical guidance for material selection, identifying the flax–WPNS hybrid as the most balanced formulation when a multifunctional performance is required. Based on their performance profiles, these composites are suitable for targeted applications. Acoustically optimized S11 is well suited for automotive interior panels, headliners, and noise-damping components. Thermally efficient and dimensionally stable S12 is promising for building interiors, thermal management panels, and partition boards. The mechanically balanced S10 is appropriate for consumer products, furniture elements, and semi-structural interior components, where strength and sustainability must be combined. Future work should focus on the quantitative evaluation of intraphase formation using pycnometry and spectroscopy, long-term durability under environmental exposure, and scaling the hybrid architectures [76] to industrial manufacturing routes such as extrusion and additive manufacturing.

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Abbreviations

α	Thermal diffusivity
ASTM	American Society for Testing and Materials
BET	Brunauer–Emmett–Teller

BJH	Barrett–Joyner–Halenda
BM	Bonferroni Mean
CaCO ₃	Calcium carbonate
COPRAS	Complex Proportional Assessment
C _p	Specific heat capacity
CRITIC	Criteria Importance Through Intercriteria Correlation
FFT	Fast Fourier transform
g/cm ³	Gram per cubic centimeter
ISO	International Organization for Standardization
k	Thermal conductivity
LFA	Laser flash analyzer
MCDM	Multi-criteria decision-making
Mpa	Megapascal
nm	Nanometer
NRC	Noise reduction coefficient
PLA	Poly(lactic acid)
PMMA	Poly(methyl methacrylate)
RH	Relative humidity
ROM	Rule of mixtures
ρ _{app}	Apparent density
ρ _{ROM}	Theoretical density calculated using rule of mixtures
SAC	Sound absorption coefficient
SEM	Scanning electron microscopy
TFBM	Triangular Fuzzy Bonferroni Mean
TFN	Triangular Fuzzy Number
UTM	Universal testing machine
W/(mK)	Watt per meter kelvin
WPNS	Waste pistachio nut shells
WQES	Waste quail eggshell
WTLF	Waste tea leaf fiber
μm	Micrometer
ShD	Shore-D Hardness

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