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Sustainable composite foam development: Crosslinked tapioca starch with corn husk sheet reinforcement and chitosan biocoating

Manisara Phiriyawirut*, Kanitta Liangkee, Prueksa Pruangsillaparat and Suchada Tunnara
Department of Tool and Materials Engineering, Faculty of Engineering, King Mongkut's University of Technology Thonburi, Thungkru, Bangkok 10140, THAILAND
*Corresponding author: manisara.pee@kmutt.ac.th

ABSTRACT

Laminar composite foams were developed to create environmentally sustainable materials. Crosslinked tapioca starch was utilized as the foam matrix and reinforcement was provided by corn husk sheets, with a chitosan biocoating subsequently applied to modify the surface. These foams were produced through hot-pressing, with the number of corn husk sheet layers varied (1, 2, and 3) alongside chitosan coating concentrations (1% and 3% by weight). The resultant materials were subjected to comprehensive characterization, where morphology, density, crystallinity, flexural and impact strength, thermal stability, and water absorption properties were evaluated. Refined foam cell structure and reduced structural defects were observed upon incorporating corn husk sheets. Increases in density and mechanical strength, as determined through flexural and impact testing, were documented with higher fiber content. In contrast, thermal stability decreased, while water absorption was reduced. The chitosan biocoating further densified the foam cell structure, increasing density and mechanical strength with increasing concentration, though crystallinity showed no significant change. Notably, the chitosan coating improved water resistance. The composite foam with two corn husk layers and a 3% chitosan coating exhibited optimal performance, demonstrating a 57.32% reduction in water uptake after 20 minutes, a 251.61% increase in flexural strength, and a 195.23% rise in impact strength compared to the unmodified foam. These sustainable composite foams show significant potential for sustainable food packaging, highlighting the synergistic property enhancements achieved through this laminar composite architecture.

Keywords: Starch foam, Corn husk, Chitosan, Laminar composite, Water resistance

INTRODUCTION

Thailand has been increasingly confronted by a significant environmental challenge stemming from the rising volume of annual waste. This increase is largely attributed to greater consumer spending and rapid technological advancements, leading to shorter product lifespans. Notably, plastic materials, especially foam plastics, are widely used in packaging and containers due to their lightweight nature and ease of manufacture. However, the growing accumulation of plastic waste has exacerbated environmental problems [1]. In 2021, approximately 2.76 million tons of plastic waste were generated in Thailand, representing about 11% of the total waste. Most of this plastic was single-use. These synthetic, non-biodegradable plastics pose a major disposal issue. Incineration, a common waste management method, has resulted in environmental pollution. Specifically, the combustion of foam materials releases styrene gas, which is absorbed through the skin and lungs. Long-term styrene exposure has been linked to adverse effects on the central nervous system, including headaches, fatigue, and potential depression [2].

To address these environmental concerns, bio-based polymers have been investigated as sustainable substitutes for materials derived from fossil fuels, with their potential for reduced harmful emissions being a key advantage. Tapioca starch, a readily available bio-based polymer in Thailand, has been considered for food packaging applications under low moisture conditions. However, its inherent limitations, particularly brittleness, low mechanical strength, and poor water resistance, have restricted its widespread use compared to synthetic polystyrene foam. Starch, a renewable and biodegradable biopolymer obtained from abundant agricultural sources such as tapioca, corn, and rice, is a promising alternative to petroleum-based packaging materials [3-4]. Its easy availability, low cost, and biodegradability have aligned with the increasing demand for environmentally conscious materials.

Starch-based foams are recognized for their limited flexibility and high susceptibility to moisture

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[5]. To mitigate these inherent drawbacks, starch modification through crosslinking has been explored. Glyoxal, a documented crosslinking agent, effectively alters starch, improving water resistance [6-7]. Nevertheless, crosslinked starch foams still exhibit issues with brittleness and high water absorption. To address these remaining shortcomings, strategies involving blending with hydrophobic polymers, such as octanoyl starch [8] and PBS [9], or reinforcement with natural fibers like chitin [8], have been investigated. Natural fibers, including jute, corn husk, kraft pulp, sugarcane bagasse, and flax, have also been considered as reinforcement materials [10]. Studies on the mechanical properties of these natural fibers indicate that fiber length and orientation significantly influence their strength [11]. Consequently, research has increasingly focused on composite material development. Laminar composites, characterized by low density, lightweight, and versatile properties, have been explored as a potential solution. Material properties can be tailored in these structures by strategically aligning fibers or reinforcements along applied stress directions, optimizing performance for specific uses [12-13].

Often viewed as readily available agricultural residues, corn husks, the natural protective outer layers of corn cobs, have gained increasing attention as a sustainable and economical resource, traditionally used for food wrapping during cooking. A distinct color change is typically observed, from light green when fresh to cream or light brown upon drying. Fibers easily obtained from corn husks and stalks have been explored for textiles and composite reinforcement, and they are already being used in decorations and some food packaging due to their low cost [14]. The significant elongation exhibited by corn husks under tension, comparable to coconut and palm fibers, makes them interesting for reinforcing polymer matrices. It is generally accepted that a natural fiber's elongation at break is mainly determined by its cellulose content and the alignment of cellulose microfibrils along the fiber axis. Consequently, corn husk fibers have been investigated as reinforcement in numerous polymer composite studies [15-16]. Based on these promising characteristics, corn husk sheets are particularly suitable for laminar reinforcement within crosslinked starch foam structures.

To specifically address the inherent susceptibility to water often associated with starch foam packaging, crosslinked starch laminate composite foams, reinforced through the strategic incorporation of corn husks in a layered structure, were developed as a central focus of this research. Corn husks were utilized explicitly as a reinforcing phase within these composite structures to improve the overall mechanical integrity and reduce water uptake. However, it is well-established that natural cellulose fibers, including those abundantly present in corn husks, are known to exhibit a degree of interaction with water molecules due to the presence of numerous hydroxyl groups (-OH) within their fundamental chemical structure, which readily attract and bind with water molecules through hydrogen bonding [17]. This inherent characteristic has been observed to result in limitations such as poor overall water resistance of the resulting composite.

Chitosan, a linear polysaccharide copolymer derived from chitin, has garnered significant interest for its ability to impart enhanced water resistance and antifungal properties, as widely explored in scientific literature [18-22]. Previous studies have demonstrated that chitosan coatings can slightly increase density and reduce water and moisture absorption in starch-based materials [23]. Its capacity to form films that significantly enhance barrier properties, particularly water resistance due to its polar nature, makes it a promising candidate. Moreover, chitosan is recognized for its potential to improve mechanical strength and rigidity when applied as a coating, a characteristic expected to densify the surface of our starch/corn husk foams and provide additional structural integrity. Its inherent antimicrobial properties also offer a potential benefit for food packaging applications.

This study introduces a novel approach to address limitations in existing starch-based biocomposites by developing a unique laminar composite architecture. This innovative design integrates a crosslinked tapioca starch foam matrix with distinct corn husk sheet reinforcing layers, a structural configuration that is less explored in sustainable materials. We aim for a synergistic enhancement of properties through this specific layered design. A chitosan biocoating is then applied to the composite surface. This coating's primary goal is to enhance water resistance, while its known antimicrobial properties offer a potential secondary benefit for food packaging applications. Our research focuses explicitly on simultaneously improving critical properties like water resistance and mechanical strength, which remain significant hurdles for many current starch-based materials, especially for sustainable food packaging. We investigate how varying the number of corn husk sheet layers and the chitosan coating concentration influences these crosslinked tapioca starch laminate composite foams' morphology, density, thermal properties, flexural/impact strength, and water absorption. This detailed investigation aims to clearly articulate the scientific gap our work addresses and its unique contribution to the field of sustainable biocomposites.

MATERIALS AND METHODS

Materials

Tapioca starch, the foundational matrix material, was obtained from commercial suppliers in Thailand. Processing aids, including guar gum and magnesium

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stearate, were sourced from Chemipan Corporation Co., Ltd. (Thailand). To modify the tapioca starch matrix, glyoxal (40% solution, Merck) was employed as the crosslinking agent. Acetic acid (analytical reagent grade, RCI Labscan Co. Ltd, Thailand) was utilized in pH adjustments and chitosan processing. Corn husk sheets, representing the sustainable reinforcement component, were procured from a local farm within Thailand. The corn husk sheets used were carefully selected to maintain consistent dimensions, specifically 20 mm in width, 150 mm in length, and approximately 0.2 mm in thickness. The control over thickness was primarily achieved through a meticulous manual selection process of individual corn husk sheets, ensuring uniformity for the fabrication of the composite foams. Chitosan was acquired from Marine Bio Resources Co. Ltd. (Thailand) to apply biocoating, which is characterized by a degree of deacetylation (DD) of 72%.

Preparation of chitosan biocoated corn husk sheets

A chitosan biocoating procedure was implemented to enhance interfacial compatibility and reduce the moisture sensitivity of the corn husk sheet reinforcement. Chitosan solutions, at 1% and 3% (w/v) concentrations, were formulated by dissolving chitosan powder in a 1% (v/v) aqueous acetic acid solution. The resulting solutions were magnetically stirred at room temperature (25°C) until complete dissolution and homogeneity were achieved.

Next, pre-cut corn husk layers were immersed in the prepared chitosan solutions at 25°C for uniform coating. After immersion, these biocoated layers were oven-dried at 50°C for 10 minutes to remove residual solvent and fix the chitosan. The resulting chitosan-modified reinforcement was then stored in a desiccator until incorporated into the crosslinked tapioca starch foam matrix.

Crosslinked tapioca starch preparation

An initial pre-drying step was performed on 80 g of tapioca starch at 110°C for 24 hours to remove residual moisture and prepare the crosslinked tapioca starch. The dried starch was then dispersed in 120 mL of deionized water. Glyoxal, employed as the crosslinking agent, was added at a concentration of 0.0375 g per 100 g of starch [7-8]. The mixture was then mechanically stirred at 25°C for 20 minutes to ensure uniform crosslinking was achieved. Subsequently, the resulting starch-glyoxal mixture was vacuum-filtered using an aspirator pump. Unreacted glyoxal was removed by thoroughly rinsing the filtered mixture three times with deionized water. The filtered crosslinked starch was then dried at 50°C for 24 hours. Finally, the dried material was ground into a fine powder using a mortar and pestle and then sieved through a 60-mesh sieve to obtain a consistent particle size.

Composite foam formulation

A uniform dry blend was formulated using 100 g of crosslinked tapioca starch powder. To enhance binding and lubrication, 1 wt% guar gum and 2 wt% magnesium stearate were incorporated, respectively [9]. These components were thoroughly mixed using a kitchen-aid mixer. Subsequently, 120 g of distilled water was gradually introduced into the dry mixture. The resulting mixture was subjected to an additional 20 minutes of mixing to ensure complete dispersion and hydration.

Fabrication of crosslinked starch/corn husk laminate composite foams

Composite foam fabrication utilized a compression molding technique. Molds, 15 mm wide by 120 mm long, adhered to ASTM D5943-5996 flexural and impact testing standards. Starch mixture and reinforcing corn husk layers (either uncoated or chitosan-coated) were integrated following specific patterns (Figure 1). For the single-layer design (Figure 1a), the process began with a starch mixture base, followed by a corn husk layer centered in the mold, then topped with more starch mixture. In the two-layer configuration (Figure 1b), two husk layers were placed at the mold's bottom and top, with starch mixture introduced between them. For the three-layer setup (Figure 1c), three husk layers alternated with the starch mixture, with a husk layer forming the final surface. The loaded molds were then subjected to 160°C and 50 bar pressure for 4 minutes to facilitate crosslinking and lamination. The resulting crosslinked starch/husk laminate composite foams were subsequently cooled to ambient temperature.

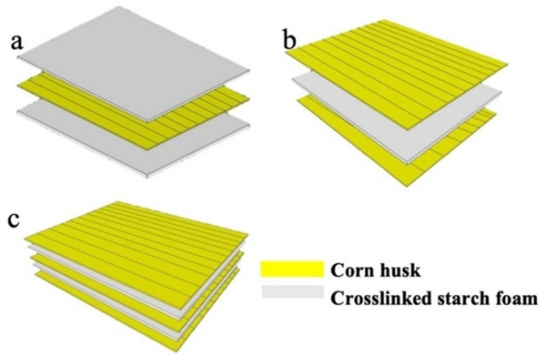

Figure 1 The configuration of crosslinked starch/corn husk laminate composite foams with varying numbers of corn husk layers: (a) single-layer, (b) two-layer, and (c) three-layer.

Characterization

Morphology: The microstructure of corn husk sheets, as well as crosslinked tapioca starch/corn husk laminate composite foams, both with and without chitosan biocoating, was investigated using scanning electron microscopy (SEM, JEOL JSM-6610 LV). Fracture surfaces resulting from flexural testing were sputter-

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coated with gold to improve electrical conductivity. SEM images were obtained at an accelerating voltage of 10 kV, allowing for visualization of the foam morphology and interfacial interactions.

Density: The density of the composite foams was determined at room temperature using a density determination kit, with chloroform employed as the displacement fluid.

Crystal structure: The crystalline structure of the composite foams was analyzed using X-ray diffraction (XRD). Measurements were performed at room temperature on both chitosan-coated and uncoated samples.

Thermal stability: The thermal degradation behavior of the composite foams was evaluated using thermogravimetric analysis (TGA, NETZSCH TG 209 F3 Tarsus). Before TGA, samples were dried at 80°C for 72 hours to remove residual moisture. TGA was conducted under a nitrogen atmosphere at a heating rate of 10°C/min, from 30°C to 500°C. Weight loss data was subsequently used to assess the thermal stability of the materials.

Mechanical properties: The flexural strength of the composite foams was determined through a three-point bending test conducted on a LLOYD Instruments LR50K universal testing machine, following ASTM D790-03. The test was performed with a maximum load of 5 kN and a 2 mm/min crosshead speed. The impact strength of notched composite foams was evaluated using a cantilever beam (Izod-type) impact test, per ASTM D256. Notches, 4.8 mm in depth and 0.25 mm in radius, were introduced into the samples using a milling machine, as specified by ASTM D256. Three impact tests were performed per sample, and the average impact strength was subsequently reported.

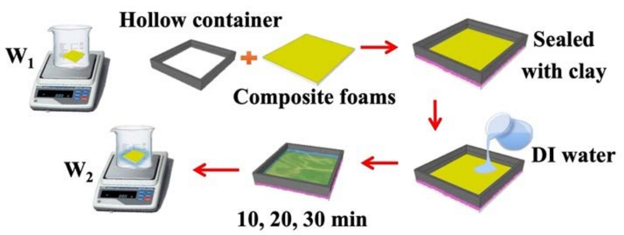

Figure 2 Experimental setup for water absorption crosslinked starch/corn husk laminate composite foams.

Water absorption and weight loss: To evaluate the water absorption behavior of the fabricated sustainable composite foams, square specimens of crosslinked tapioca starch/corn husk laminate composites (15.0 mm x 15.0 mm), both with and without chitosan biocoating, were prepared. Water absorption capacity was determined using a custom-designed apparatus, as shown in Figure 2. A plastic hollow rectangular container with dimensions of 15.0 × 15.0 mm, sized to fit the test samples precisely, was placed on each foam specimen and sealed with modeling clay to prevent leakage. Deionized water was then carefully added to the container. Water uptake was measured at predetermined time intervals of 10, 20, and 30 minutes. After each interval, any surface water was gently removed using absorbent paper, and the specimens were immediately reweighed. All tests were performed under controlled conditions at 25 °C. The percentage of water absorption was calculated using Equation 1:

$$\text{Water Absorption (\%)} = [(W_2 - W_1) / W_1] \times 100 \quad (1)$$

where W_1 represents the initial weight of the specimen, and W_2 represents the weight of the specimen after water absorption.

In order to evaluate the potential for degradation or leaching of constituent components from the composite foams, weight loss measurements were conducted. Specimens, sized 15.0 mm x 15.0 mm, were immersed in deionized water for 24 hours. Following this immersion, specimens were carefully removed and placed within a forced-air convection oven, maintained at a temperature of 60°C. Drying was continued for 24 hours, or until a consistent weight was attained, indicating the removal of all absorbed moisture. The weight loss percentage was then determined by applying Equation 2.

$$\text{Weight Loss (\%)} = [(W_1 - W_3) / W_1] \times 100 \quad (2)$$

where W_1 represents the initial weight of the specimen, and W_3 represents the weight of the specimen after immersion and drying.

RESULTS AND DISCUSSION

Crosslinked starch/corn husk laminate composite foams

Crosslinked tapioca starch composite foams were reinforced with corn husk sheets, selected for their established mechanical strength and inherent water resistance. To evaluate the influence of reinforcement layers, single-layer, two-layer, and three-layer configurations of corn husk were incorporated into the starch foam matrix.

The morphology of the resulting crosslinked tapioca starch/corn husk laminate composite foams was examined through scanning electron microscopy (SEM). Representative SEM images are presented in Table 1. For pure crosslinked tapioca starch foam, a dense outer layer with small foam cells was observed, while the inner core exhibited larger cells and evidence of fracture, indicative of inherent brittleness. However, upon the introduction of a single corn husk sheet as a central reinforcement layer, a more uniform foam cell structure was achieved. This structural modification was accompanied by an increase in overall density and a reduction in average cell size. Furthermore, the void size within the central core was also observed to decrease.

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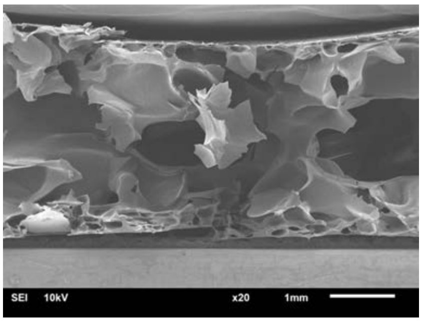
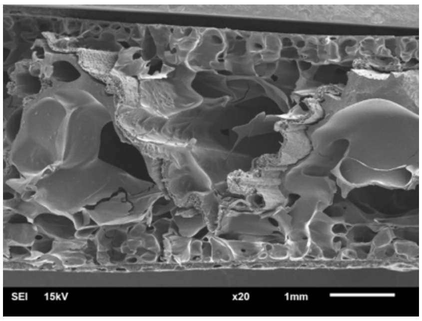
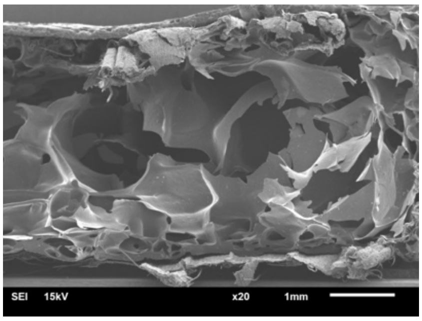
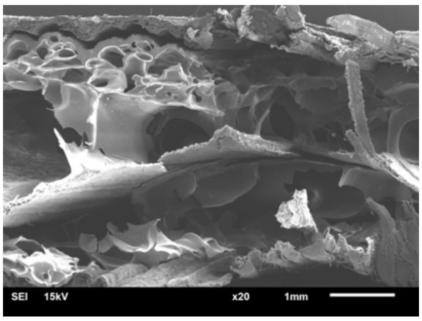
A notable increase in foam cell density and a concurrent decrease in cell size were observed upon incorporating two corn husk sheets, strategically positioned on the upper and lower surfaces of the tapioca starch foam. Conversely, including three corn husk sheet layers resulted in the formation of larger voids within the foam structure. It is hypothesized that the increased confinement of the starch matrix between three corn husk sheet layers restricted its expansion during the foaming process, leading to the development of larger, fewer foam cells. Consequently, the three-layer composite exhibited a lower overall foam density when compared to the composites reinforced with one and two layers. These findings highlight the significant influence of corn husk sheet distribution on the resulting foam morphology and density.

Table 1 Fracture surface analysis (visual inspection and SEM micrograph) and density of crosslinked starch/corn husk laminate composite foams with varying numbers of corn husk layers.

Numbers of corn husk layers	Fracture surface		Density (g/cm^3)
	Visual inspection	SEM micrograph (20X)	
0	-		0.29 ± 0.01
1			0.41 ± 0.13
2			0.41 ± 0.15
3			0.60 ± 0.32

The density of the crosslinked tapioca starch/corn husk laminate composite foams, incorporating one, two, and three layers of corn husk sheets, was measured and is presented in Table 1. A consistent increase in density was observed with each additional corn husk sheet layer. This increase can be attributed

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to the inherent density of the corn husk sheets, determined to be 0.35 ± 0.45 g/cm³. As the number of corn husk sheet layers integrated into the composite structure was increased, a corresponding rise in the overall density of the resulting foam material was noted. This finding is consistent with previous research [8, 24]. This observation highlights the effective contribution of corn husk sheets as a reinforcing component in enhancing the bulk density of the tapioca starch-based composite foam.

X-ray diffraction (XRD) patterns, illustrating the crystalline structures of crosslinked tapioca starch foam and a composite foam reinforced with two corn husk sheet layers, are presented in Figure 3. A characteristic peak at 17.1°, indicative of the semi-crystalline nature of the starch, was observed in the XRD pattern of the crosslinked tapioca starch foam [25]. Upon incorporating two corn husk sheet layers, a new peak was detected at 22.6°, attributed to crystalline cellulose within the corn husk [26]. This cellulose-specific peak, absent in the pure starch foam pattern, confirms the successful integration of corn husk sheets. While the overall pattern of the laminate composite foam appeared sharper than that of the pure starch foam, this primarily indicates the crystallinity contributed by the corn husk component itself, rather than a significant increase in the crystallinity of the starch matrix.

Table 2 presents the flexural strength and elongation at break of the crosslinked tapioca starch/corn husk laminate composite foams. A notable increase in flexural strength was observed as the corn husk sheet content was increased from one to two layers. This improvement is attributed to the laminar reinforcement mechanism, which effectively enhanced the material's capacity to bear load during three-point bending tests. It is theorized that this type of reinforcement facilitates a more efficient distribution of both compressive and tensile stresses experienced by the material, a phenomenon supported by previous studies' findings [27]. However, a subsequent increase in the corn husk sheet content to three layers decreased flexural strength. This reduction is likely due to a relative decrease in the proportion of the continuous starch matrix within the composite structure, leading to a reduction in the number of cohesive foam cells and a concurrent increase in overall porosity. Consequently, the efficiency of interfacial stress transfer between the distinct phases of the composite was compromised, ultimately resulting in a lower measured flexural strength. Moreover, while corn husk provides reinforcement, the interface between the corn husk and the starch matrix might not be perfectly bonded without adhesion promoters. At three layers, the increased number of interfaces could lead to more potential sites for interfacial debonding or delamination under flexural stress, thereby lowering the strength.

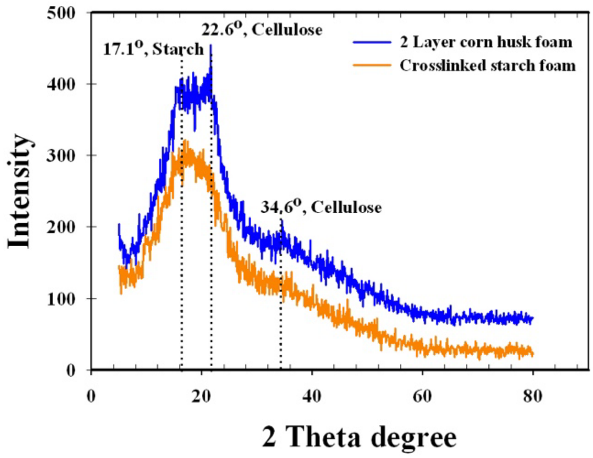

Figure 3 XRD patterns of crosslinked starch foam and crosslinked starch laminate composite foams with 2 layers of corn husk sheet.

Table 2 Flexural properties and impact strength of crosslinked starch/corn husk laminate composite foams with varying numbers of corn husk layers.

Numbers of corn husk layers	Flexural properties			Impact strength (J/m²)
	Strength (MPa)	Strain (%)	Modulus (MPa)	
0	3.1 ± 0.4	4.1 ± 0.0	278 ± 0.1	56.6 ± 0.5
1	7.5 ± 0.2	4.8 ± 0.1	566 ± 0.0	96.9 ± 0.2
2	10.3 ± 0.1	4.6 ± 0.8	796 ± 0.2	139.0 ± 0.3
3	8.2 ± 0.3	7.0 ± 0.2	278 ± 0.1	117.7 ± 0.4

Interestingly, the observed increase in flexural strain for the three-layer composite, notably higher than that of the one- and two-layer samples, can be attributed to several toughening mechanisms inherent in multi-layered laminar structures. The increased number of reinforcing corn husk layers likely facilitates improved stress distribution across the composite, allowing individual layers to deform more effectively before catastrophic failure [28]. This multi-layered configuration can also promote crack deflection at the interfaces, forcing propagating cracks to follow a more tortuous path and thereby absorbing more energy [29]. Furthermore, the overall enhanced deformability of the structure, despite the presence

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of rigid reinforcing layers, may stem from the intricate interplay between the flexible starch foam matrix and the multiple corn husk layers, enabling greater strain accommodation before fracture [30]. These combined effects contribute to the superior ductility observed in the three-layer composite.

Impact resistance of the crosslinked tapioca starch laminate composite foams, reinforced with varying corn husk sheet layers (1, 2, and 3 layers), was evaluated, and the results are presented in Table 2. A noticeable increase in impact resistance was recorded when the corn husk sheet content was raised from 1 to 2 layers. This improvement is believed to be a direct result of the inherent strength of the corn husk sheets, which effectively reinforced the composite foam against impact. Furthermore, the increased density and reduced foam cell size observed in the 2-layer samples facilitated efficient stress transfer within the composite, contributing to the enhanced impact performance. However, a subsequent decrease in impact resistance was seen upon further increasing the corn husk sheet content to 3 layers. This reduction is associated with a corresponding decrease in foam density and an increase in foam cell size, leading to larger voids. These voids are hypothesized to have interfered with effective stress transfer, thereby diminishing the overall impact resistance of the composite foam.

The thermal stability of corn husk sheets, crosslinked tapioca starch foam, and the resulting crosslinked tapioca starch/corn husk laminate composite foams was evaluated using thermogravimetric analysis (TGA). Distinct thermal degradation profiles for each material were observed, as illustrated in Figure 4 and summarized in Table 3.

Thermal decomposition of corn husk sheets was observed to occur in two stages. An initial 5.67% weight loss was noted between 36°C and 162°C, attributed to moisture evaporation. A subsequent substantial weight loss of 38.87% began at 188°C, corresponding to cellulose and hemicellulose degradation. Similarly, crosslinked tapioca starch foam exhibited a two-stage degradation profile. The first stage, between 36°C and 248°C, involved 7.16% weight loss due to moisture evaporation. At 269°C, significant degradation (79.21% weight loss) was observed, associated with the breakdown of α-1,4 glycosidic linkages within the starch polymer.

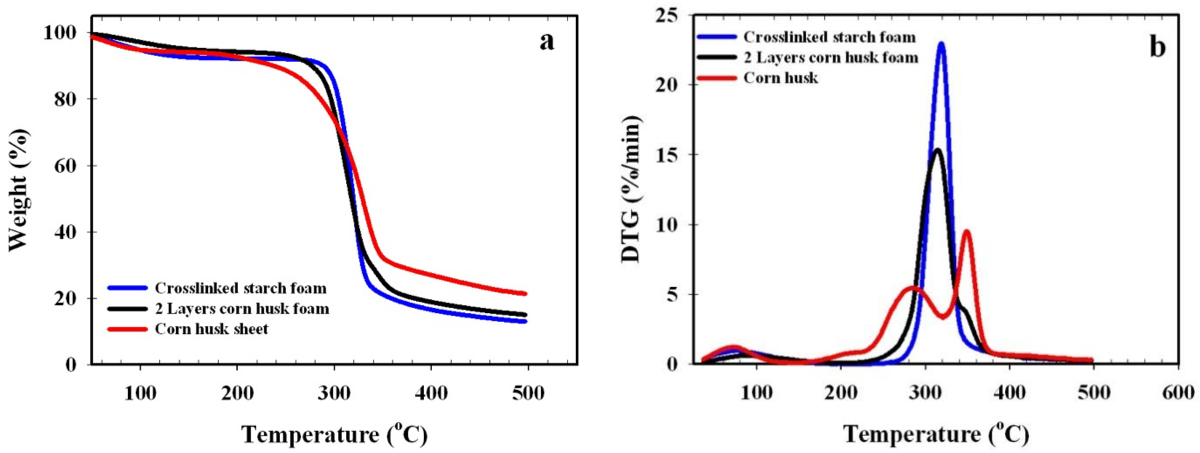

Figure 4 Thermal degradation behavior of corn husk sheet, crosslinked starch foam and crosslinked starch/corn husk laminate composite foams (a) TGA thermogram and (b) DTG thermogram.

Table 3 Thermal degradation of corn husk sheet, crosslinked starch foam and crosslinked starch/corn husk laminate composite foams.

Sample	1st Degradation		2nd Degradation	
	T (°C)	W (%)	T (°C)	W (%)
Corn husk sheet	36-162	5.67	188	72.76
Crosslinked starch foams	36-248	7.16	269	79.21
Crosslinked starch laminate composite foams with 2 layers of corn husk sheet	36-117	5.70	247	79.14

It was observed that the crosslinked tapioca starch/corn husk laminate composite foams demonstrated a reduced thermal stability when compared to the crosslinked tapioca starch foam. This reduction can be attributed to corn husk sheets containing β-1,4 glycosidic linkages. These linkages are known to degrade at a lower temperature (188°C) than the α-1,4 linkages found in tapioca starch (degrading between 260-320°C). Consequently, the composite foams showed an earlier onset of thermal degradation [31], indicating that the corn husk

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component's thermal stability dictates the overall composite's initial degradation temperature.

The water absorption behavior of crosslinked tapioca starch foam and its corn husk sheet reinforced composites was analyzed over a 10 to 30 minutes period (Figure 5). An increase in water absorption with time was observed for both materials. However, water uptake was significantly reduced upon incorporating corn husk sheets.

The reduction in water absorption is primarily attributed to the barrier effect of the layered corn husk sheets within the composite structure. While cellulose, the main component of corn husk [14], exhibits an inherent lower affinity for water than the highly hydrophilic starch matrix, its role extends beyond mere hydrophobicity. The dense, layered arrangement of the corn husk sheets effectively impedes the diffusion of water molecules into the underlying starch foam matrix, thereby limiting direct and prolonged contact between water and the more absorbent starch component. This physical impediment significantly contributes to the observed decrease in overall water uptake.

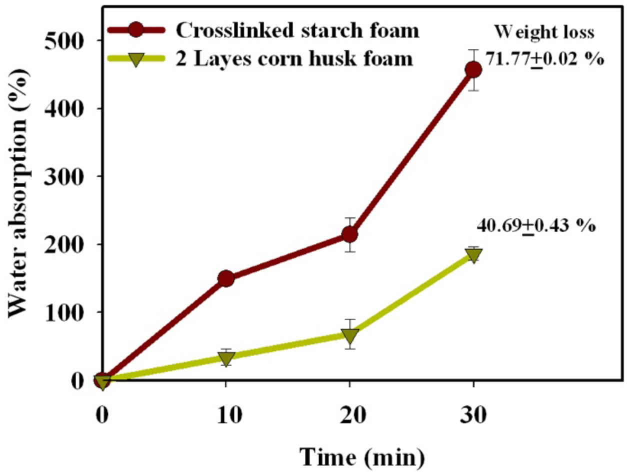

Figure 5 Water absorption and weight loss of crosslinked starch foam and crosslinked starch laminate composite foams with 2 layers of corn husk sheet.

Further evidence supporting the enhanced water resistance was obtained through weight loss measurements. Following a 24-hour water immersion and subsequent 24-hour drying at 60°C, the neat crosslinked tapioca starch foam exhibited a substantial weight loss of 71.77 ± 0.02%, indicating significant water-induced degradation. In contrast, the composite reinforced with two layers of corn husk sheet demonstrated a significantly lower weight loss of 40.69 ± 0.43%. This represents a 43.3% reduction in weight loss compared to the neat crosslinked tapioca starch foam. This decrease in weight loss is attributed to the protective effect of the corn husk sheets, which effectively encapsulate the starch foam, thereby minimizing direct water contact and reducing the leaching of starch components into the aqueous medium.

The placement of corn husk sheet layers significantly influences the composite's overall performance in practical applications, particularly concerning mechanical integrity, barrier properties, and material efficiency. A well-distributed, multi-layered structure enhances flexural and impact strength, which is crucial for durable packaging during handling and transport, by effectively distributing stress and resisting crack propagation. Furthermore, the strategic layering of corn husk within the hydrophilic starch foam creates tortuous pathways for water molecules, significantly improving water resistance-a vital property for preserving food freshness. This allows for tailoring packaging solutions to specific performance needs, optimizing material usage, and contributing to the sustainability and cost-effectiveness of potential mass production.

Chitosan-coated crosslinked starch/corn husk laminate composite foams

The crosslinked tapioca starch laminate composite foams reinforced with corn husk sheets exhibited enhanced mechanical properties and improved, albeit still limited, water resistance. Furthermore, the susceptibility of corn husk sheets to fungal growth under high humidity conditions was identified as a challenge. To address these limitations, a chitosan solution was applied as a biocoating to the corn husk sheets further to enhance the water resistance of the composite foams. Chitosan coatings with concentrations of 1% and 3% (w/v) were investigated. The inherent hydrophilicity of both starch and corn husk contributed to the observed water absorption and fungal susceptibility. Chitosan, recognized for its antimicrobial and hydrophobic characteristics, was employed to establish a barrier against water uptake and fungal proliferation [22].

The cross-sectional morphology of composite foams, reinforced with two layers of corn husk sheets and subjected to chitosan biocoatings (0-3 wt%), was examined via scanning electron microscopy (SEM), with representative micrographs presented in Table 4. SEM analysis revealed a significant influence of the chitosan biocoating on the resultant foam morphology. Specifically, increased foam cell density was observed with increasing chitosan concentrations. This densification is attributed to forming a chitosan film that effectively occluded pores within the corn husk sheets. Consequently, water vapor diffusion during the foam formation process was hindered, leading to restricted foam cell expansion and a denser overall microstructure. The chitosan biocoating, therefore, plays a key role in modifying the cellular architecture of these sustainable composite foams, resulting in enhanced structural integrity and potentially improved

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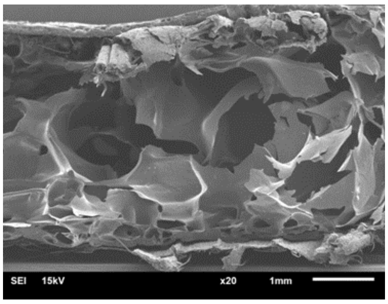
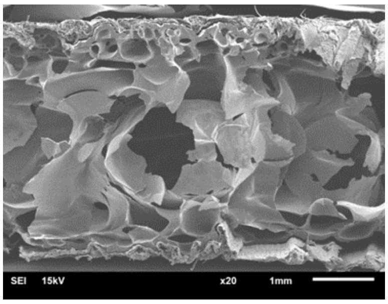
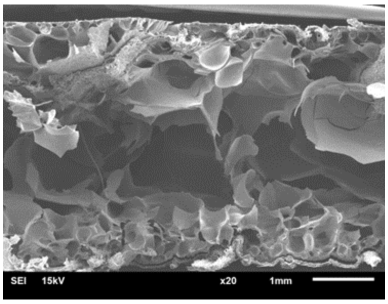
barrier properties against moisture and other environmental factors.

A direct correlation was observed between chitosan coating concentration and the density of crosslinked tapioca starch laminate composite foams reinforced with two layers of corn husk sheets. As detailed in Table 4, foam density increased proportionally with higher chitosan concentrations. This increase is attributed to the deposition of chitosan, a relatively dense polymer, onto corn husk fibers and within the foam's internal structure. The inherent high density of corn husk sheets (1.16 ± 0.40 g/cm³) also contributed to the composite's overall density. Chitosan application effectively filled voids and interstitial spaces within the foam, yielding a more compact material. This density enhancement is considered a key factor in the improved mechanical properties of the composite foams, as load-bearing capacity and structural integrity are directly influenced.

Table 4 Fracture surface analysis (SEM) and density of two-layered corn husk/crosslinked starch composite foams with varying chitosan concentrations.

	Chitosan concentration (%)		
	0	**1**	**3**
SEM (20X)			
Density (g/cm^3)	0.41 ± 0.15	0.43 ± 0.29	0.50 ± 0.20

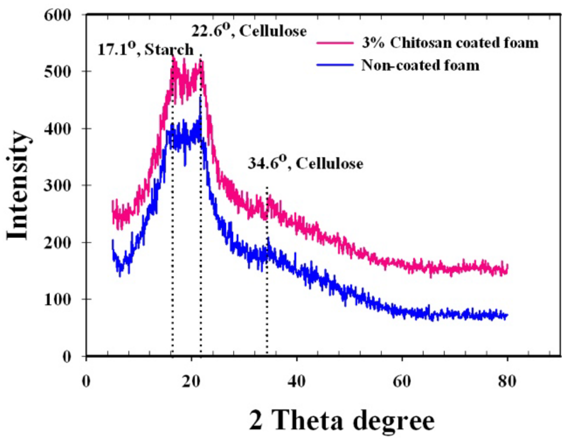


Figure 6 XRD patterns of crosslinked starch laminate composite foams with 2 layers of corn husk sheet with and without 3% chitosan coating.

Figure 6 displays the X-ray diffraction (XRD) patterns obtained for the crosslinked tapioca starch/corn husk laminate composite foams, with a direct comparison made between the uncoated sample and the sample coated with 3 wt% chitosan. Typical chitosan diffraction peaks, usually found around 9.35°, 19.42°, and 21.14° [32], were not visible in the pattern of the chitosan-coated composite foam. This absence is likely because the chitosan concentration used was too low for the XRD technique to detect. No significant change in the crystallinity of the composite foam was observed with the chitosan coating, suggesting a minimal impact on the material's structural order. This indicates that while chitosan may interact with the corn husk/tapioca starch matrix, it does not substantially alter the overall crystallinity under these conditions.

The thermal resistance properties of crosslinked tapioca starch laminate composite foams, reinforced with corn husk sheets with and without 3% chitosan coating, were compared using thermogravimetric analysis (TGA). As depicted in Figure 7 and summarized in Table 5, the results revealed that the composite foams containing 2% corn husk sheets, both uncoated and coated with 3% chitosan, exhibited a two-stage thermal degradation process.

The initial stage of thermal degradation, associated with the evaporation of absorbed moisture and volatile components, was observed in the temperature range of 36-117°C for the uncoated composite, resulting in a 5.7% weight loss. This initial stage occurred between 36-190°C for the chitosan-coated composite, with a corresponding weight loss of 6.81%. The second major degradation stage, attributed to the decomposition of α 1-4 glycosidic linkages and chitosan, commenced at 247°C for the uncoated sample and a slightly lower temperature of 238°C for the chitosan-coated sample. This second stage resulted in substantial weight

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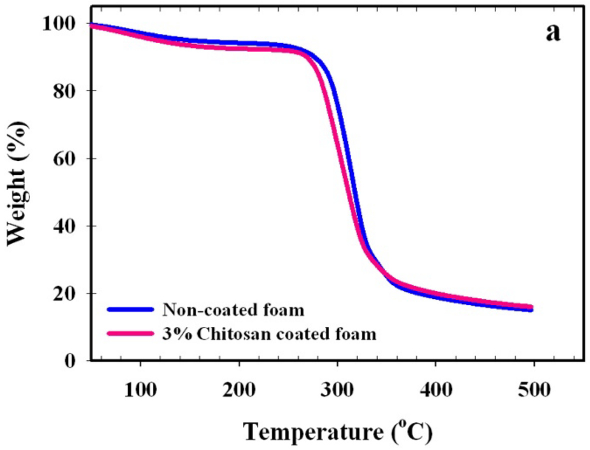
losses of 79.14% and 79.37% for the uncoated and coated samples, respectively.



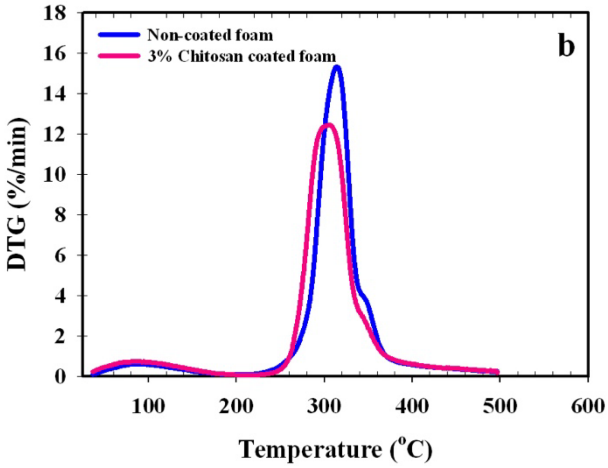


Figure 7 Thermal degradation behavior crosslinked starch laminate composite foams with 2 layers of corn husk sheet with and without 3% chitosan coating. (a) TGA thermogram and (b) DTG thermogram.

Table 5 Thermal degradation of two-layered corn husk/crosslinked starch composite foams with varying chitosan concentrations.

Chitosan conc. (%)	1st Degradation		2nd Degradation	
	T (°C)	W (%)	T (°C)	W (%)
0	36-117	5.7	247	79.14
3	36-190	6.81	238	79.37

It was observed that the chitosan-coated corn husk sheet reinforced crosslinked tapioca starch composite foam exhibited slightly faster thermal degradation compared to the uncoated counterpart. This accelerated degradation is believed to be due to the earlier decomposition of chitosan, which occurs around 200°C, a temperature lower than that of corn husk sheets and crosslinked tapioca starch. The earlier decomposition of chitosan likely initiates the degradation process at a lower temperature, leading to a slightly faster overall degradation rate for the coated composite.

The flexural and impact properties of crosslinked tapioca starch composite foams, reinforced with two corn husk sheet layers and a chitosan biocoating, were evaluated, and the results are detailed in Table 6. A slight enhancement in both flexural strength and elongation was observed with increasing chitosan concentration. This improvement is attributed to the chitosan coating, which reduced foam cell size and increased density, leading to higher structural rigidity. Furthermore, the crosslinked tapioca starch composite foams upon chitosan coating noted a marginal increase in impact resistance. This observation is believed to be due to the influence of chitosan on the foam structure, where smaller and denser foam cells are thought to contribute to improved impact energy absorption.

Table 6 Flexural properties and impact strength of two-layered corn husk/crosslinked starch composite foams with varying chitosan concentrations.

Chitosan conc. (%)	**Flexural properties**			**Impact strength (J/m^2)**
	Strength (MPa)	**Strain (%)**	**Modulus (MPa)**	
0	10.3 ± 0.1	4.6 ± 0.8	796.0 ± 0.2	139.0 ± 0.3
1	10.7 ± 0.1	3.9 ± 0.7	982.2 ± 0.2	160.8 ± 0.0
3	10.9 ± 0.2	2.7 ± 0.0	989.8 ± 0.0	167.1 ± 0.2

The water absorption of composite laminate foams over 10-30 minutes was investigated, with a comparison made between crosslinked tapioca starch/corn husk foams and chitosan-coated variants (Figure 8). A reduction in water penetration was observed following chitosan application onto the corn husk sheets. This decrease is attributed to chitosan's hydrophobic nature, effectively sealing the pores of the exterior corn husk sheets and limiting water uptake. Notably, the 3 wt% chitosan coating exhibited lower water absorption than the 1 wt% coating, likely due to the more substantial barrier hindering water molecule diffusion.

Weight loss in crosslinked tapioca starch/corn husk laminate composite foams, with and without chitosan biocoating, is presented in Figure 8. A reduction in weight loss was observed upon chitosan application to the corn husk sheets. This phenomenon is attributed to the hydrophobic nature of chitosan, leading to diminished dissolution or leaching of materials due to reduced water interaction. Furthermore, a progressive decrease in weight loss was recorded

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with increasing chitosan concentrations on the corn husk sheet surfaces, suggesting the formation of a more substantial barrier against water interaction with a denser chitosan layer.

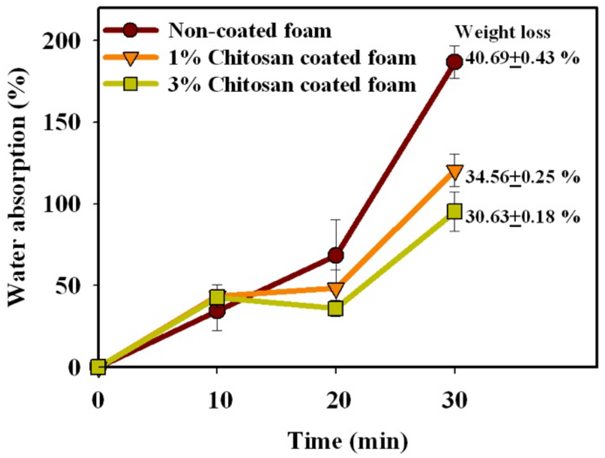

Figure 8 Water absorption and weight loss of crosslinked starch laminate composite foams with 2 layers of corn husk sheet with varying chitosan concentrations.

CONCLUSIONS

This study investigated the morphology, density, crystal structure, flexural and impact strength, thermal stability, and water absorption of crosslinked tapioca starch laminate composite foams, reinforced with corn husk sheets, with and without chitosan coating. Key findings are summarized:

Crosslinked tapioca starch composite foams reinforced with corn husk sheets exhibited an open-cell structure. Smaller, denser foam cells at the edges resulted in a higher density than crosslinked tapioca starch foam. Flexural and impact strength increased with more corn husk layers, attributed to corn husk's good elongation. However, thermal stability was lower than that of crosslinked tapioca starch foam. Corn husk sheets' top and bottom placement restricted foam cell expansion, leading to smaller, denser cells and improved flexural strength and water resistance.

Chitosan coating on corn husk sheets sealed surface pores, impeding water vapor evaporation. This resulted in smaller, denser foam cells, increased flexural and impact strength, and improved water resistance, which was further enhanced by increasing chitosan concentration.

In summary, the structural and functional properties of crosslinked tapioca starch composite foams were significantly influenced by corn husk sheet incorporation and chitosan biocoating. An optimized formulation of two corn husk sheet layers and a 3% chitosan coating demonstrated substantial improvements This particular composition showed a 57.32% reduction in water uptake and marked enhancements in mechanical strength, with a 251.61% increase in flexural strength and a 195.23% rise in impact strength compared to the unmodified foam, highlighting the strong potential of these sustainable materials for various applications, especially sustainable food packaging.

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REFERENCES

1. Jambeck JR, Geyer R, Wilcox C, Siegler TR, Perryman M, Andrady A, et al. Plastic waste inputs from land into the ocean. Science [Internet]. 2015;347(6223): 768-71. Available from: http://doi.org/10.1126/science.1260352.
2. Rosemond Z, Chou S, Wilson J, Schwartz M, Tomei-Torres F, Ingerman L, et al. Toxicological profile for styrene. Atlanta, Georgia: Agency for Toxic Substances and Disease Registry; 2010.
3. Tapia-Blácido DR, Aguilar GJ, Teixeira de Andrade M, Rodrigues-Júnior MF, Guareschi-Martins FC. Trends and challenges of starch-based foams for use as food packaging and food container. Trends Food Sci [Internet]. 2022;119:257-71. Available from: https://doi.org/10.1016/j.tifs.2021.12.005.
4. Jiang T, Duan Q, Zhu J, Liu H, Yu L. Starch-based biodegradable materials: Challenges and opportunities. Adv Ind Eng Polym Res [Internet]. 2020;3:8-18. Available from: https://doi.org/10.1016/j.aiepr.2019.11.003.
5. Bénézet JC, Stanojlovic-Davidovic A, Bergeret A, Ferry L, Crespy A. Mechanical and physical properties of expanded starch, reinforced by natural fibres. Ind Crop Prod [Internet]. 2012;37(1):435-40. Available from: https://doi.org/10.1016/j.indcrop.2011.07.001.
6. Kumar Y, Singh S, Saxena DC. A comprehensive review on methods, mechanisms, properties, and emerging applications of crosslinked starches. Int J Biol Macromol [Internet]. 2025;306(2):141526. Available from: https://doi.org/10.1016/j.ijbiomac.2025.141526.
7. Uslu MK, Polat S. Effects of glyoxal cross-linking on baked starch foam. Carbohydr Polym [Internet]. 2012;87(3):1994-99. Available from: https://doi.org/10.1016/j.carbpol.2011.10.008.

8. Phiriyawirut M, Hankham P, Butsukhon R, Pongvichai U. Biomass-based composite foam from tapioca starch/octenyl succinate starch blended with alpha-chitin. Open J Compos Mater [Internet]. 2019;9:355-64. Available from: https://doi.org/10.4236/ojcm.2019.94022.

9. Phiriyawirut M, Mekaroonluck J, Hauyam T, Kittilaksanon A. Biomass-based foam from crosslinked tapioca starch/polybutylene succinate blend. J Renew Mater [Internet]. 2016;4:185-89. Available from: https://doi.org/10.7569/JRM.2015.634121.

10. Bénézet JC, Stanojlovic-Davidovic A, Bergeret A, Ferry L, Crespy A. Mechanical and physical properties of expanded starch, reinforced by natural fibres. Ind Crop Prod [Internet]. 2012;37(1):435-40. Available from: https://doi.org/10.1016/j.indcrop.2011.07.001.

11. Dungani R, Karina M, Subyakto, Sulaeman A, Hermawan De, Hadiyane A. Agricultural waste fibers towards sustainability and advanced utilization: A review. Asian J Plant Sci [Internet]. 2016;15:42-55. Available from: https://doi.org/10.3923/ajps.2016.42.55.

12. Liew KM, Pan ZZ, Zhang LW. An overview of layerwise theories for composite laminates and structures: Development, numerical implementation and application. Compos Struct [Internet]. 2019;216:240-59. Available from: https://doi.org/10.1016/j.compstruct.2019.02.074.

13. Phiriyawirut M, Rodprasert P, Kulvorakulpitak P, Cothsila R, Kengkla N. Pushing the boundaries of starch foams: Novel laminar composites with paper reinforcement. Renew Mater [Internet]. 2025;13(1):101-14. Available from: https://doi.org/10.32604/jrm.2024.056830.

14. Reddy N, YongY. Natural cellulose fibers from corn stover. In: Innovative Biofibers from Renewable Resources [Internet]. Springer Berlin: Heidelberg; 2015. Available from: https://doi.org/10.1007/978-3-662-45136-6.

15. Ibrahim MIJ, Sapuan SM, Zainudin ES, Zuhri MYM. Potential of using multiscale corn husk fiber as reinforcing filler in cornstarch-based biocomposites. Int J Biol Macromol [Internet]. 2019;139:596-604. Available from: https://doi.org/10.1016/j.ijbiomac.2019.08.015.

16. Hazrol MD, Sapuan SM, Ilyas RA, Zainudin ES, Zuhri MYM, Abdul NI. Effect of corn husk fibre loading on thermal and biodegradable properties of kenaf/cornhusk fibre reinforced corn starch-based hybrid composites. Heliyon [Internet]. 2023;9(4):e15153. Available from: https://doi.org/10.1016/j.heliyon.2023.e15153.

17. Natural Fibers, Biopolymers, and Biocomposites [Internet]. Mohanty AK, Misra M, Drzal LT. 1st ed. Boca Raton: CRC Press; 2005. Available from: https://doi.org/10.1201/9780203508206.

18. Ben Seghir B, Benhamza MH. Preparation, optimization and characterization of chitosan polymer from shrimp shells. J Food Meas Charact [Internet]. 2017;11:1137-47. Available from: https://doi.org/10.1007/s11694-017-9490-9.

19. Aguilar R, Nakamatsu J, Ramírez E, Elgegren M, Ayarza J, Kim S, et al. The potential use of chitosan as a biopolymer additive for enhanced mechanical properties and water resistance of earthen construction. Constr Build Mater [Internet]. 2016;114:625-37. Available from: https://doi.org/10.1016/j.conbuildmat.2016.03.218.

20. Inthamat P, Karbowiak T, Tongdeesoontorn W, Siripatrawan U. Biodegradable active coating from chitosan/astaxanthin crosslinked with genipin to improve water resistance, moisture and oxygen barrier and mechanical properties of Kraft paper. Int J Biol Macromol [Internet]. 2024;254(2):127816. Available from: https://doi.org/10.1016/j.ijbiomac.2023.127816.

21. Tanpichai S, Witayakran S, Wootthikanokkhan J, Srimarut Y, Woraprayote W, Malila Y. Mechanical and antibacterial properties of the chitosan coated cellulose paper for packaging applications: Effects of molecular weight types and concentrations of chitosan. Int J Biol Macromol [Internet]. 2020;155:1510-19. Available from: https://doi.org/10.1016/j.ijbiomac.2019.11.128.

22. El-araby A, Janati W, Ullah R, Uddin N, Bari A. Antifungal efficacy of chitosan extracted from shrimp shell on strawberry (*Fragaria × ananassa*) postharvest spoilage fungi. Heliyon [Internet]. 2024;10(7):e29286. Available from: https://doi.org/10.1016/j.heliyon.2024.e29286.

23. Bergel BF, da Luz LM, Santana RMC. Comparative study of the influence of chitosan as coating of thermoplastic starch foam from potato, cassava and corn starch. Prog Org Coat [Internet]. 2017;106:27-32. Available from: https://doi.org/10.1016/j.porgcoat.2017.02.010.

24. Zhang X, Teng Z, Huang R, Catchmark JM. Biodegradable starch/chitosan foam via microwave assisted preparation: Morphology and performance properties. Polymers [Internet]. 2020;12(11):2612. Available from: https://doi.org/10.3390/polym12112612.

25. Akuzawa S, Okada N, Tamaki Y, IkegamiK A, Fujita N, Vilpoux OF, et al. Physicochemical properties of starches isolated from five cassava (Manihot esculenta Crantz) landraces of Brazil. J

Appl Glycosci [Internet]. 2012;59(3):131-38. Available from: https://doi.org/10.5458/jag.jag.JAG-2011_030.

26. Prado KS, Spinacé MAS. Characterization of fibers from pineapple's crown, rice husks and cotton textile residues. Mater Res [Internet]. 2015;18(3):530-37. Available from: https://doi.org/10.1590/1516-1439.311514.
27. Abdellaoui H, Bensalah H, Echaabi J, Bouhfid R, Qaiss A. Fabrication, characterization and modelling of laminated composites based on woven jute fibres reinforced epoxy resin. Mater Des [Internet]. 2015;68:104-13. Available from: Available from: https://doi.org/10.1016/j.matdes.2014.11.059.
28. Arruda Filho AB, Lima PRL, Carvalho RF, Gomes OdFM, Filho RDT. Effect of number of layers on tensile and flexural behavior of cementitious composites reinforced with a new sisal fabric. Textiles [Internet]. 2024;4(1):40-56. Available from: https://doi.org/10.3390/textiles4010004.
29. Faber KT, Evans AG. Crack deflection processes-I. Theory. Acta Metall [Internet]. 1983;31(4):565-76. Available from: https://doi.org/10.1016/0001-6160(83)90046-9.
30. Tamlicha A, Rizal S, Hasanuddin I, Noor MM, Ikramullah I, Nazaruddin N. The effect of number of laminate layers on the ramie E-glass fiber hybrid composite for Jaloe kayoh material. Materials Science Forum [Internet]. 2025;1149:47-54. Available from: https://doi.org/10.4028/p-d718kw.
31. Hiremath VS, Reddy DM, Mutra RR, Sanjeev A, Dhilipkumar T, et al. Thermal degradation and fire retardant behaviour of natural fibre reinforced polymeric composites- A comprehensive review. J Mater Res Technol [Internet]. 2024;30:4053-63. Available from: https://doi.org/10.1016/j.jmrt.2024.04.085.
32. Kumari S, Rath PK. Extraction and characterization of chitin and chitosan from (Labeo rohit) fish scales. Procedia Mater Sci [Internet]. 2014;6:482-89. Available from: https://doi.org/10.1016/j.mspro.2014.07.062.