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Tailoring the Mechanical and Antimicrobial Properties of PLA Bioplastics Through Chitosan and Citronella Oil Additives

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ABSTRACT

This study investigates the effect of varying chitosan concentrations on the mechanical properties of polylactic acid (PLA)-based films modified with citronella essential oil as an antimicrobial agent. Characterization results indicate that the incorporation of chitosan at 3–5 wt% relative to PLA enhances the tensile strength of the material. This improvement is attributed to the formation of intermolecular hydrogen bonds between the hydroxyl groups of chitosan and the ester groups of PLA, which strengthen the film structure. However, the addition of citronella essential oil to the system significantly reduces the tensile strength. This reduction is associated with the poor compatibility between the essential oil and the PLA–chitosan matrix. Scanning electron microscopy (SEM) analysis revealed the presence of oil droplet agglomerations within the matrix, acting as stress concentration points that weaken the film's mechanical integrity. Such phenomena have been widely reported in PLA-essential oil composite systems. Regarding antimicrobial activity, all films containing both chitosan and citronella oil exhibited significant inhibitory effects against *Staphylococcus aureus* (Gram-positive) and *Escherichia coli* (Gram-negative). This antimicrobial efficacy is attributed to a synergistic mechanism, involving the electrostatic disruption of bacterial cell membranes by chitosan and the cytotoxic effects of citronella constituents such as citronellal and geraniol, which penetrate and damage microbial cell walls.

Key words: *PLA, chitosan, citronella essential oil, Escherichia coli, Staphylococcus aureus.*

PENDAHULUAN

The COVID-19 pandemic served as a pivotal moment that heightened global awareness of the significance of adopting a healthier lifestyle in daily routines. One key component of this shift has been the increased consumption of fruits and vegetables, which are widely acknowledged for their numerous health benefits. In the post-pandemic era, the demand for fresh-cut produce has grown substantially, driven by consumer preferences for fresh, natural, and convenient food products. However, this trend has posed significant challenges for the food industry, particularly in preserving the freshness and extending the shelf life of perishable items such as meat, vegetables, and fruits.

Fresh produce, being integral to a balanced diet, begins to degrade in quality after harvest due to the cessation of metabolic processes. This degradation is marked by changes in color, flavor, weight, and nutritional content. A critical factor contributing to this decline is water loss, which leads to nutrient depletion, textural softening, shrinkage, and eventual wilting.

In light of increasing environmental concerns and the limitations associated with petroleum-based packaging, the scientific community has shifted toward the use of bio-based materials. Among these, polylactic acid (PLA) has emerged as a leading candidate due to its favorable moisture and water vapor barrier properties, as well as its GRAS (Generally Recognized as Safe) status for food contact applications (Marano *et al.,* 2022). Despite its advantages, PLA presents certain limitations, including high sensitivity to UV radiation and inherent brittleness, which restrict its utility in some packaging scenarios.

PLA is among the most widely utilized biopolymers for sustainable plastic production. Yet, its practical application is constrained by inherent mechanical limitations: PLA exhibits brittleness, reduced toughness, and limited thermal resilience (Rajendran et al., 2024). To overcome these drawbacks, it is typically blended with more flexible biopolymers such as chitosan. Chitosan is a natural, non-toxic, biodegradable, and biocompatible polysaccharide derived from chitin, primarily sourced from crustacean shells

Studies investigating PLA-chitosan composites have revealed that a high chitosan content in PLA enhances the compatibility and bonding between CS and PLA, which leads to improved tensile strength of the composite. However, at higher chitosan loadings (3–5 wt %), agglomeration tends to occur, reducing both strength and modulus (Daramola *et al.,* 2021). These findings align with broader research emphasizing the benefits of biopolymer blending to address PLA's brittleness while retaining biodegradability and processability.

Meanwhile, chitosan has garnered significant attention in food packaging due to its exceptional film-forming capacity and broad-spectrum antimicrobial and antifungal activities. It has been repeatedly shown to inhibit post-harvest pathogens. Specifically, chitosan-based coatings with sea buckthorn oil films have been effective in suppressing fungal spoilage in strawberries, provided the lowest count of molds and yeasts until day 7 of the analysis (Popescu *et al.*, 2022). Another research shows that treatments with chitosan concentrations between 1–3 %, applied as sprays or dips, significantly reduce fungal infection rates, extending shelf life by delaying decay symptoms and preserving fruit quality during cold storage.

Furthermore, several focused studies integrating chitosan with essential oils, minerals, or specially formulated sprays have demonstrated significant enhancements in the shelf life of fruits and vegetables during storage. Therefore, the present study is designed to investigate the antibacterial performance of biodegradable films developed from a PLA/chitosan matrix incorporated with citronella oil, with the intended application as protective films for fresh produce.

MATERIALS AND METHODS

Materials

The materials utilized in this study include PLA, chitosan, citronella essential oil, as well as both gram-negative and gram-positive bacterial strains. The experimental setup involves the use of a distillation apparatus, an extruder, and a hot press machine. Table 1. Provide the materials composition for each sample.

Table 1. composition of each sample

No	**Sample**	**Fillers**	
		chitosan	**Essential Oil**
1	Pure PLA	0	0
2	A1	3%	0
3	A2	5%	0
4	A3	7%	0
5	A4	3%	3 mL
6	A5	5%	3 mL
7	A6	7%	3 mL

Distillation

The lemongrass leaves were chopped and weighed to a total of 250 grams, then placed into a three-neck flask for distillation at 100 °C over a period of 5 hours. The resulting distillate collected during the process was subsequently analyzed using GC-MS for further characterization.

Films production

To fabricate the films, a total of 200 g of PLA was blended with chitosan at varying weight percentages (3%, 5%, and 7%) using a single-screw extruder. The resulting mixture was then transferred to a hot press, followed by the addition of 3 mL of lemongrass essential oil. Afterward, it was poured into petri dishes and dried in room temperature at 33 °C.

RESULTS

Tensile Strength

Polylactic acid (PLA) is recognized for its relatively high tensile strength and Young's modulus, indicative of its inherent stiffness. However, due to its semi-crystalline structure and lack of molecular flexibility, PLA exhibits poor ductility, with a notably low elongation at break. To overcome this brittleness, chitosan is frequently incorporated as a reinforcing biopolymer. The incorporation of chitosan, can enhance the stiffness of PLA composites by restricting the mobility of PLA chains through strong intermolecular interactions, including hydrogen bonding (Kamaludin *et al*., 2021).

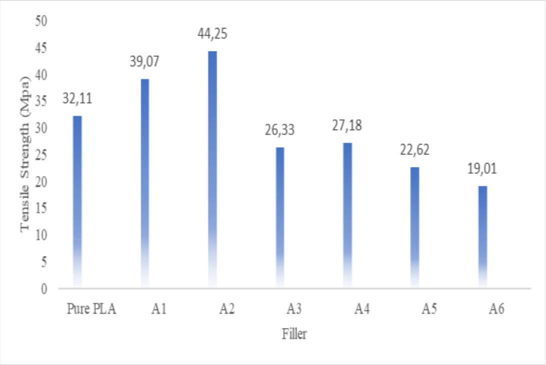

Figure 1. Tensile properties of films

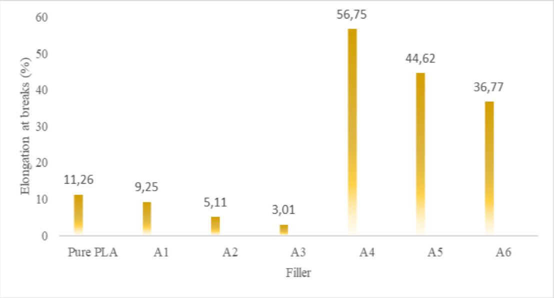

Figure 2. Elongation at breaks

Nonetheless, when chitosan content exceeds approximately 5 wt% (Figure 1), a decline in mechanical performance is often observed. This behavior is attributed to the limited thermodynamic compatibility between PLA and chitosan. Due to this immiscibility, phase separation occurs, and chitosan tends to form discrete microdomains dispersed within the PLA matrix, which can act as stress concentrators and compromise the structural integrity of the composite. Literature supports that the elastic modulus of PLA-based composites generally decreases upon chitosan incorporation, particularly at higher loadings, although this reduction is less pronounced when compared to plasticized PLA, which already exhibits reduced stiffness due to plasticizer-induced chain mobility.

The addition of citronella essential oil, commonly used as a natural plasticizer and antimicrobial agent, further alters the mechanical profile of the material (Abdou *et al.*, 2024). As expected from plasticization theory, the tensile strength decreased significantly due to enhanced chain mobility. Conversely, the elongation at break increases, indicating improved ductility and flexibility, which is consistent with the behavior of other essential oil-plasticized PLA systems.

Scanning Electron Microscope (SEM)

Based on the previously evaluated mechanical performance, the A4 formulation demonstrated superior properties, making it the most suitable candidate for food packaging applications. To further understand its structural integrity, the surface and cross-sectional morphology of the A4 film were examined using scanning electron microscopy (SEM), as illustrated in Figure 3. The SEM micrographs revealed a continuous and uniform matrix with a smooth surface, free from visible defects such as cracks, pores, or phase separations, suggesting efficient film formation and matrix cohesion.

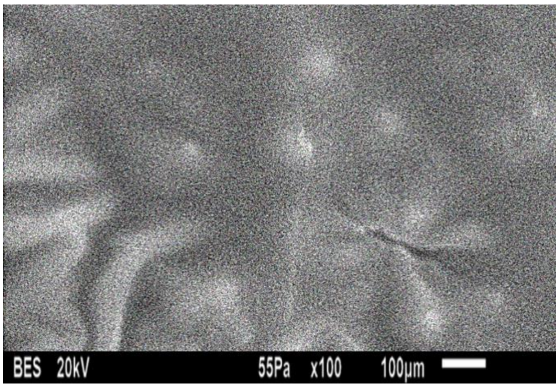

Figure 3. SEM of A4 Films

The homogeneity observed indicates a favorable dispersion of components and minimal interfacial stress, which are essential for maintaining mechanical strength and barrier function in packaging materials. The solubility and structural integrity of bioplastic films are known to be influenced by the compatibility between the polymer matrix and any added active compounds. In this context, the presence of chitosan was depending on its interaction with the PLA matrix which may result in variations in interfacial adhesion. Such differences were inferred across all samples, potentially due to chitosan's polar nature and its partial immiscibility with hydrophobic polymers like PLA.

Furthermore, the SEM images provide insights into the compactness and internal organization of the filmogenic network. A more compact structure suggests strong intermolecular interactions and reduced void spaces, which are advantageous for mechanical robustness and reduced water vapor permeability both critical parameters for food packaging materials.

Antibacterial Properties

The antibacterial activity of PLA-based films incorporated with chitosan and citronella essential oil was evaluated against *Staphylococcus aureus*, and the results are presented in Figure 4. All film formulations (A1–A6) demonstrated a reduction in viable bacterial cells after 24 hours of incubation, indicating the potential of these films to inhibit bacterial growth. Among the tested samples, film A6 showed the highest antibacterial activity, as evidenced by the most significant decrease in colony-forming units (CFU/mL), suggesting the effectiveness of the combined use of chitosan and essential oil as antimicrobial agents.

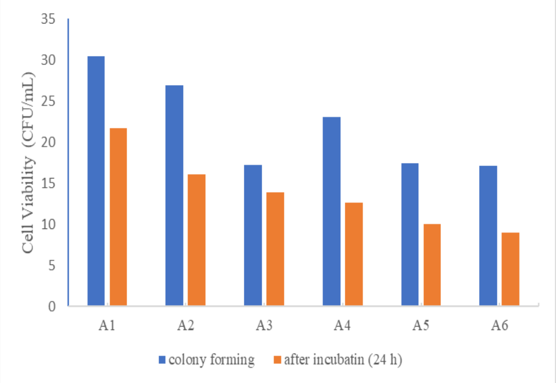

Figure 4. Antibacterial properties for *S. Aureus*

The observed reduction in CFU/mL is consistent with the known antibacterial mechanism of chitosan, which interacts electrostatically with the negatively charged bacterial cell membranes. This interaction can lead to leakage of intracellular components and eventual cell death. Films A3 to A6, which are presumed to contain increasing concentrations of chitosan, displayed progressively stronger antibacterial effects, with A6 achieving the most substantial bacterial suppression. This trend supports findings from previous studies, such as those by Ardean et al., (2021) which confirm chitosan's effectiveness as a natural antimicrobial polymer.

In addition to chitosan, the incorporation of citronella essential oil further enhanced the antibacterial properties of the films. Citronella oil contains active compounds such as citronellal, geraniol, and limonene, which are known for their ability to penetrate microbial cell membranes and disrupt essential biological functions (Kacaniova *et al.*, 2024). As demonstrated by Mielczarek et al., (2025) such essential oils can provide broad-spectrum antimicrobial activity when effectively dispersed within a biopolymer matrix. The synergistic interaction between chitosan and citronella oil likely accounts for the pronounced bacterial inhibition observed in the A5 and A6 samples.

The SEM analysis from earlier characterization likely revealed a uniform and compact film structure, especially in the more active formulations, supporting effective encapsulation and gradual release of the antimicrobial agents. These structural features, combined with the chemical activity of the additives, contributed to the observed antibacterial performance. The homogeneous distribution of chitosan and oil droplets in the matrix enhances contact with bacterial cells, while excessive phase separation or agglomeration (as potentially seen in A1–A2) may limit bioactive surface exposure and reduce efficacy.

Overall, the A6 film formulation emerged as the most promising for food packaging applications where antimicrobial activity is critical. Its strong inhibition against *S. aureus* indicates its potential to prolong shelf life and reduce foodborne contamination. The results align well with previous literature on bioactive bioplastics and underscore the importance of optimizing both the concentration and compatibility of active agents within the polymer matrix. Future work may further explore the release kinetics and shelf stability of such packaging films under real food storage conditions.

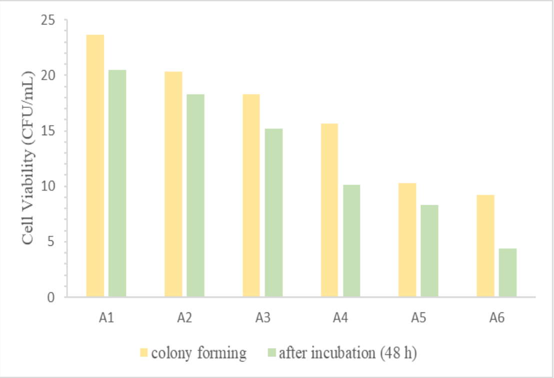

Figure 5. Antibacterial properties for *E.Coli*

The figure demonstrates a progressive reduction in *E. coli* viability across film formulations A1 through A6 following 48 hours of incubation. The initial CFU counts are represented by the yellow bars, while the green bars reflect bacterial viability after exposure to each film formulation. It is evident that films with higher content of bioactive agents, presumably chitosan and citronella essential oil exhibit greater antibacterial efficacy, with formulation A6 showing the most pronounced inhibition, reducing *E. coli* viability by more than half compared to the initial level.

This trend highlights the effectiveness of combining chitosan and essential oils in disrupting Gram-negative bacterial activity. While *E. coli* possesses an outer membrane rich in lipopolysaccharides that can hinder the diffusion of antimicrobial agents, chitosan has been shown to overcome this barrier by interacting with negatively charged microbial surfaces, leading to changes in membrane permeability and eventual cell lysis. Chitosan's antimicrobial action is both concentration-dependent and influenced by molecular weight and degree of deacetylation (Kovacs et al., 2022).

Citronella essential oil, rich in citronellal and geraniol, further enhances the antibacterial effect through its hydrophobic components, which penetrate lipid bilayers and disrupt bacterial cell membranes. These compounds interfere with ATP production and enzyme activity within bacterial cells. The incorporation of essential oils into polymeric matrices like PLA enables sustained antimicrobial release, extending protection over time especially relevant in the 48-hour used in this study.

In samples A1 and A2, the antibacterial effect is relatively modest, suggesting that these films may contain minimal amounts of chitosan or essential oil, or that their dispersion within the PLA matrix was suboptimal. As the formulation progresses toward A6, the increasing reduction in bacterial viability indicates either a higher active compound concentration or improved matrix compatibility. Particularly, A6 demonstrates an efficient synergy between chitosan and citronella oil, consistent with literature reports of combined systems outperforming single-agent films in antimicrobial tests against Gram-negative bacteria.

From a packaging perspective, the results affirm the potential of biopolymer-based films enhanced with natural antimicrobial agents for inhibiting *E. coli* as a key foodborne pathogen. The gradual increase in antimicrobial efficacy from A1 to A6 suggests a concentration-effect relationship, where both the amount and distribution of active agents play critical roles. The A6 film may be ideal for fresh produce, meat, or ready-to-eat packaging where extended microbial protection is necessary, while A4 or A5 could be suitable for moderate-barrier applications.

CONCLUSION

This study confirms that the incorporation of chitosan and citronella essential oil into polylactic acid (PLA) matrices significantly enhances the functional properties of bioplastic films for food packaging applications. Mechanically, the optimal chitosan content (≤5 wt%) improved the tensile strength of PLA films by reinforcing the polymer matrix through hydrogen bonding. However, beyond this concentration, phase separation due to thermodynamic immiscibility between chitosan and PLA adversely affected mechanical integrity. The addition of citronella oil, acting as a natural plasticizer, further modified the film's mechanical behavior by reducing tensile strength but improving elongation at break, thereby increasing the film's flexibility as a desirable feature for packaging.

Scanning electron microscopy (SEM) of the A4 film, which showed the most balanced mechanical properties, revealed a homogeneous and compact morphology. This uniformity indicates favorable interaction among the polymer components, crucial for ensuring consistent mechanical performance and barrier properties. A compact internal structure is particularly beneficial for reducing moisture permeability and enhancing shelf life in food packaging systems.

The antibacterial assays demonstrated a clear trend: films with higher chitosan and essential oil content exhibited superior antimicrobial activity. Against *Staphylococcus aureus* (Gram-positive) and *Escherichia coli* (Gram-negative), the A6 formulation was the most effective, reducing bacterial viability significantly after 24 and 48 hours, respectively. This confirms the synergistic effect of chitosan, which disrupts bacterial membranes via electrostatic interaction, and citronella oil, whose hydrophobic constituents penetrate and destabilize microbial cell structures.

These findings align with existing literature and reinforce the role of natural antimicrobials in developing active packaging materials. Importantly, the gradual improvement in antimicrobial efficacy from A1 to A6 illustrates that both the concentration and compatibility of bioactive agents are critical parameters in designing efficient functional films.

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