

Development of A Bioactive Composite Coating Based on Cotton Linter Nanocellulose, Chitosan, And Sodium Alginate and Optimization of The Printability of Packaging Materials

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Abstract

In this study, a bioactive composite coating was developed based on nanocellulose ("C" _6 "H" _10 "O" _5)_n derived from cotton linters, chitosan ("C" _6 "H" _11 "NO" _4)_n, and sodium alginate ("C" _6 "H" _7 "NaO" _6)_n. Glycerol ("C" _3 "H" _8 "O" _3) was employed as a plasticizer, and "CaCl" _2 served as an ionic crosslinking agent. The resulting three-dimensional polymer network enhanced surface energy, ink adhesion, optical color density, dot gain reduction, and moisture resistance. FTIR, SEM, XRD, and contact angle analyses confirmed molecular-level interactions and structural modifications on the surface. Based on domestic cotton raw materials, an innovative bioactive coating technology was proposed for eco-friendly, biodegradable packaging substrates with superior printability.

Keywords: Cotton linters, nanocellulose, chitosan, sodium alginate, bioactive composite coating, biodegradable packaging, printability, ink adhesion, surface energy, barrier properties.

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1. Introduction

The rapid development of sustainable practices and green chemistry principles has established biodegradable polymer systems as a top research priority within polymer chemistry, materials science, and graphic arts engineering. Due to surging production volumes of

conventional synthetic packaging polymers—such as polyethylene ($-(C_2H_4)-_n$), polypropylene ($-(C_3H_6)-_n$), and polyethylene terephthalate (PET)—persistent polymer waste accumulation poses acute environmental threats. Their resistance to biodegradation, generation of microplastics, and high lifecycle carbon footprint make the fabrication of

functional, environmentally safe materials derived from renewable lignocellulosic biomass an urgent scientific task.

Cellulose ($C_6H_{10}O_5$)_n derived from lignocellulosic biomass represents a viable natural polymer for nanocellulose production owing to its high degree of crystallinity, abundance of hydroxyl ($-OH$) groups, and excellent mechanical strength. Uzbekistan ranks among

the leading cotton-producing countries, and cotton linters exhibit immense scientific and economic potential as a domestic raw material with high α -cellulose content. Nanocellulose produced through sequential bleaching and acid hydrolysis markedly upgrades the physicochemical characteristics of polymer composites through its high specific surface area, dense hydrogen-bonding network, and reactive hydroxyl groups.

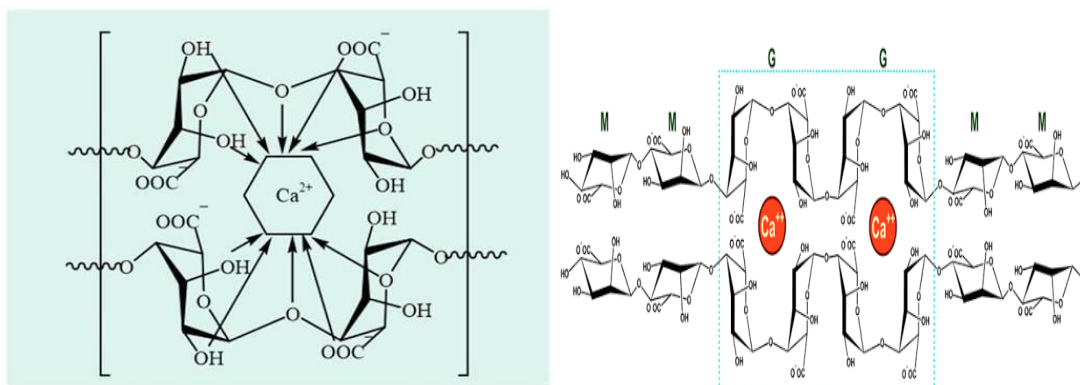


Figure 1. Schematic illustration of the three-dimensional macromolecular network formed via the "egg-box" coordination model of sodium alginate in the presence of Ca^{2+} ions.

The printing quality of packaging materials depends directly on surface free energy, wettability, surface roughness, and interfacial ink-substrate adhesion. Chitosan ($C_6H_{11}NO_4$)_n and sodium alginate ($C_6H_7NaO_6$)_n bioactive composites have attracted significant interest for optimizing these properties. While amine ($-NH_2$) groups in chitosan form strong hydrogen bonds with nanocellulose hydroxyl ($-OH$) groups, carboxylate ($-COO^-$) groups in sodium alginate crosslink ionically with Ca^{2+} ions into an "egg-box" three-dimensional network (Figure 1). These structural transitions improve coating mechanical durability, surface energy, barrier performance against oxygen and water vapor, and ink adhesion.

Glycerol ($C_3H_8O_3$) functions as a plasticizer, adjusting intermolecular spacing to increase coating flexibility and crack resistance. As a result, the synergistic nanocellulose–chitosan–sodium alginate matrix forms a bioactive coating that provides high print definition, optical density, reduced dot gain, rapid drying kinetics, and elevated moisture resistance.

The scientific relevance of this study lies in formulating a novel bioactive composite coating using the molecular synergy of cotton linter nanocellulose, chitosan, and sodium alginate, determining the relationships between

its physicochemical architecture and printability, and establishing an eco-friendly coating technology for the packaging and printing industry.

2. Literature Review

Recent work in biodegradable packaging chemistry focuses on developing high-performance composites from natural polysaccharides. Polymer formulations combining cellulose ($C_6H_{10}O_5$)_n, chitosan ($C_6H_{11}NO_4$)_n, and sodium alginate ($C_6H_7NaO_6$)_n stand out as particularly promising.

Wang Feijie, Li Zhihua, Zhao Zhicheng, and Ma Shufen demonstrated that nanocellulose builds stable hydrogen bonds within polymer matrices due to its high crystallinity, large specific surface area, and abundant hydroxyl ($-OH$) groups. Their findings confirmed that incorporating nanocellulose reinforces composite mechanical durability, restricts oxygen and water vapor transmission, and improves surface stability. Extracting nanocellulose from agricultural processing by-products was also highlighted as both environmentally and economically viable.

Xu Yanting, Wu Zhengzheng, Li Ao, and Chen Naijun reported that the one-dimensional fibrillar morphology of nanocellulose forms an effective reinforcing phase

between polymer chains. The authors observed substantial increases in tensile strength, thermal stability, and UV resistance in composite films, with surface hydroxyl groups playing a central role in tuning surface energy.

Jin Xin, Luo Bodan, Xuan Simin, Shen Peng, and Wu Zhenghua demonstrated that amine ($-\text{NH}_2$) groups in chitosan $(\text{C}_6\text{H}_{11}\text{NO}_4)_n$ bio-coatings impart antibacterial activity through electrostatic interactions with microbial cell membranes. Their results showed that chitosan exhibits film-forming capabilities that promote ink adhesion, reduce oxygen permeation, and extend packaged shelf life.

Studies on sodium alginate confirmed that coordination between carboxylate ($-\text{COO}^-$) groups and Ca^{2+} cations produces a stable 3D macromolecular network. This ionic crosslinking improves moisture resistance, mechanical strength, and barrier efficiency. In addition, hydrophilic segments within alginate modulate surface wettability, yielding uniform coatings.

Russian researchers Aleksandr Volkov, Dmitriy Komarov, and co-workers analyzed the extraction of high-purity cellulose from lignocellulosic biomass and its subsequent conversion into nanostructured materials. They established that combined oxidative and hydrolytic treatments raise cellulose crystallinity, while functional activation of hydroxyl groups strengthens intermolecular bonding with adjacent polysaccharides.

3. Methodology

The experimental work was carried out across five consecutive stages:

1.Extraction of Nanocellulose from Cotton Linters: Cotton linters underwent dewaxing, alkaline delignification, bleaching (using H_2O_2 or NaClO_2), and sulfuric acid hydrolysis (H_2SO_4) to isolate highly crystalline nanocellulose. Reagent concentrations, temperatures, reaction times, and solid-to-liquid ratios were systematically optimized. Crystallinity index, particle size, ζ -potential, surface hydroxyl reactivity, and thermal stability were characterized using standard physicochemical methods.

2. Molecular Synthesis of the Chitosan–Sodium Alginate–Nanocellulose Formulation: Isolated nanocellulose was blended with chitosan $(\text{C}_6\text{H}_{11}\text{NO}_4)_n$ and sodium alginate $(\text{C}_6\text{H}_7\text{NaO}_6)_n$

solutions at varying mass ratios. Glycerol ($\text{C}_3\text{H}_8\text{O}_3$) was added as a plasticizer, and CaCl_2 was introduced as an ionic crosslinking agent. Intermolecular hydrogen bonding ($(-\text{OH} \cdots \text{OH}-)$,

$(-\text{OH} \cdots \text{NH}_2-)$ and $(-\text{COO}^-)$ chelation with Ca^{2+} ions were analyzed to select the most stable formulation.

3. Structural and Physicochemical Characterization:

The synthesized formulation was applied to packaging paper substrates. Layer morphology, functional groups, crystallinity, and thermal degradation profiles were assessed using FTIR spectroscopy, X-ray diffraction (XRD), scanning electron microscopy (SEM), and thermogravimetric analysis (TGA). Coating thickness, surface free energy, contact angle, and roughness parameters were measured across formulation variations.

4.Printability and Functional Performance Testing:

Coated specimens were tested for ink adhesion strength, optical color density, dot gain, absorption kinetics, drying rate, gloss, and surface energy. Barrier and mechanical parameters—including water vapor transmission rate (WVTR), oxygen transmission rate (OTR), moisture resistance, tensile strength, and elastic modulus—were measured against uncoated control substrates.

5.Optimization and Technical Protocol Development:

Experimental data were evaluated statistically to identify how nanocellulose, chitosan, and alginate concentrations correlate with print quality, strength, and barrier performance. Based on these relationships, optimal composition ratios, an operating synthesis protocol, and practical implementation guidelines were established for industrial packaging production.

4. Results and Discussion

Developing multifunctional bioactive composites from natural polysaccharides integrates polymer chemistry, colloid science, and packaging technology. Cotton linters contain 90–95% α -cellulose, providing a uniform source for nanocellulose extraction. During bleaching and acid hydrolysis, amorphous regions are selectively degraded while crystalline domains remain intact. This produces cellulose nanocrystals tens of nanometers in diameter with accessible surface hydroxyl ($-\text{OH}$) groups that serve as reaction sites for downstream modification.

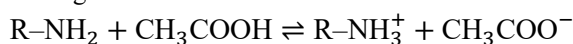
Acid hydrolysis of cellulose proceeds according to the general reaction:



where $m < n$; amorphous cellulose domains degrade into soluble sugars, leaving crystalline nanocellulose intact.

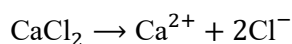
The high crystallinity, large surface area, and abundant hydroxyl groups of the resulting nanocellulose promote intermolecular bonding with adjacent biopolymers. Intermolecular hydrogen bonds densify the inner composite structure, raising elastic modulus, tensile integrity, and thermal stability. Consequently, nanocellulose acts simultaneously as a reinforcing nano-filler and a macromolecular coupling agent.

Chitosan protonates in acidic environments. In an acetic acid (CH_3COOH) solution, protonation of primary amines generates cationic sites:

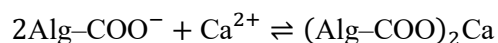


These protonated chitosan chains display strong electrostatic affinity toward carboxylate groups on sodium alginate, forming an interpolymer polyelectrolyte complex that improves composite rheology and mechanical strength.

Sodium alginate consists of alternating guluronate and mannuronate segments bearing carboxylate ($-\text{COO}^-$) groups. Upon dissociation of calcium chloride:



The liberated Ca^{2+} cations coordinate with alginate carboxylate moieties to construct a three-dimensional chelate network:



This coordinated spatial framework lowers aqueous solubility, enhances moisture durability, and hinders gas diffusion. Hydrogen bonds between nanocellulose ($-\text{OH}$), chitosan ($-\text{NH}_2$), and alginate ($-\text{OH}$) groups generate a cohesive supramolecular matrix that enhances film continuity, flexibility, and fracture resistance.

Adding glycerol ($\text{C}_3\text{H}_8\text{O}_3$) relieves internal stress between polymer chains, preserves segmental mobility, and prevents embrittlement. Its three hydroxyl groups establish supplemental hydrogen bonds with nanocellulose and chitosan, improving film elasticity and inhibiting microcrack formation.

Applied to packaging substrates, this ternary system levels surface micro-irregularities, lowers roughness, and increases surface free energy. These adjustments decrease printing ink contact angles, encouraging uniform spreading. Controlled capillary absorption limits excessive pigment penetration into substrate pores, resulting in higher optical reflection density, minimal dot gain, and sharper edge definition. Concurrently, the crosslinked macromolecular network lengthens diffusion pathways for permeating oxygen and moisture molecules, improving barrier performance and extending the shelf life of packaged contents.

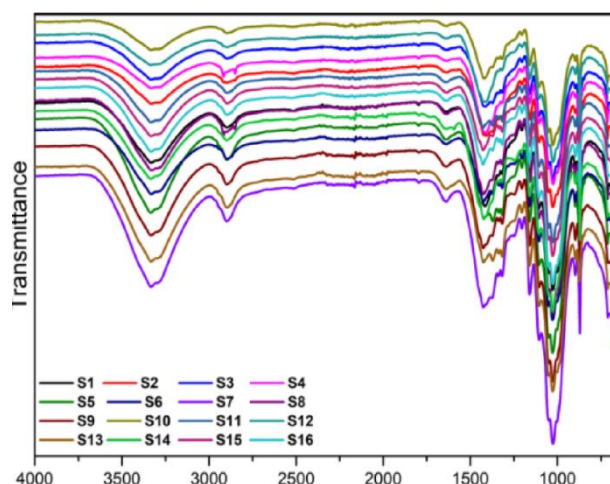


Figure 2. FTIR spectra of bioactive composite specimens based on cotton linter nanocellulose, chitosan, and sodium alginate.

FTIR spectra of all formulated specimens showed a broad absorption band at $3300\text{--}3400\text{ cm}^{-1}$, characteristic of hydrogen-bonded hydroxyl ($-\text{OH}$) stretching. Peaks around 2920 cm^{-1} correspond to aliphatic " C-H " stretching vibrations. The $1600\text{--}1420\text{ cm}^{-1}$ region reflects overlapping ($-\text{COO}^-$) asymmetric/symmetric

stretching and ($-\text{NH}_2$) bending vibrations, whereas the $1150\text{--}1020\text{ cm}^{-1}$ range is assigned to pyranose ring C-O-C skeletal vibrations along cellulose β -($1\rightarrow4$)-glycosidic linkages. Progressive peak shifts and intensity variations confirm multi-component interfacial

compatibility and successful synthesis of the ternary composite (Figure 2).

5. Conclusion

This study established and validated a bioactive composite coating system derived from domestic cotton linter nanocellulose, chitosan, and sodium alginate. The scientific contribution involves converting local agricultural cellulose into functional nanocellulose, integrating it into an ionically and hydrogen-bonded polysaccharide network, and improving packaging substrate printability and barrier performance. This bio-based approach provides a practical framework for manufacturing sustainable, biodegradable packaging materials that meet modern printing performance standards.

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