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POLYSACCHARIDE-BASED BIOPLASTICS FOR SUSTAINABLE PACKAGING: ROLE OF NONCOVALENT INTERACTIONS AND TRANSESTERIFICATION

Abstract: *The growing accumulation of plastic waste in ecosystems has catalyzed a global search for environmentally responsible packaging materials. Among biodegradable polymers, bioplastics derived from polysaccharides – especially from plant-based cellulose and its microbial analogue, bacterial cellulose (BC) – have attracted significant interest due to their renewability, biodegradability, and desirable mechanical attributes. Nevertheless, their practical application is frequently constrained by challenges such as hydrophilicity and vulnerability to environmental stressors. To overcome these issues, recent studies have explored structural modifications involving both noncovalent interactions (e.g., hydrogen bonding, ionic crosslinking) and covalent strategies such as transesterification. These approaches have been shown to improve mechanical integrity, flexibility and water resistance. This review discusses recent progress in*

engineering polysaccharide-based bioplastics, with a particular emphasis on how combined physical and chemical modifications can enhance performance. Special attention is given to hybrid systems incorporating BC, laponite, chitosan, and fatty acid esters, which demonstrate promising synergistic effects. Overall, the integration of noncovalent and covalent modifications offers a compelling strategy for developing next-generation sustainable packaging materials.

Key words: *bacterial cellulose; hydrogen bonding; ionic crosslinking; transesterification; laponite.*

Introduction

Rising environmental concerns related to plastic pollution, resource depletion, and restrictions on single-use plastics have intensified global efforts to identify sustainable alternatives to conventional packaging. Bioplastics based on natural polysaccharides have emerged as promising candidates due to their biodegradability, renewability and the potential for functional tailoring [1-3].

Among these, cellulose and its derivatives hold particular promise, owing to their widespread availability, favorable mechanical properties and processability. Bacterial cellulose (BC) highly purified form of cellulose produced by microbial fermentation, has gained increasing attention for its exceptional physical characteristics and compatibility with advanced packaging applications [4, 5]. Other polysaccharides such as chitosan, alginate, pectin, and starch have also been widely investigated for food-contact applications due to their natural origin and biocompatibility [6-8].

Despite these advantages, the practical use of polysaccharide-based materials remains limited by factors such as hydrophilicity, poor mechanical durability and sensitivity to humidity and temperature [9]. In response, researchers have focused on modifying these materials through noncovalent interactions – particularly hydrogen bonding and ionic crosslinking – to tune their molecular organization and improve bulk properties [10]. In parallel, transesterification reactions have emerged as a powerful covalent modification method, enabling the introduction of hydrophobic groups that enhance thermal stability and moisture resistance through targeted ester formation between polysaccharide hydroxyl groups and hydrophobic moieties [11].

This review provides a critical overview of current strategies in designing and modifying polysaccharide-based bioplastics using a combination of physical and chemical approaches. Emphasis is placed on the synergistic effects of noncovalent and covalent modifications, as well as the practical considerations for scaling up these materials for commercial use.

Structural Features and Functional Modifications of Cellulose and Bacterial Cellulose

Cellulose, the most abundant biopolymer on Earth, is a linear polysaccharide composed of β-(1→4)-linked D-glucose units. It is primarily obtained from plant cell walls and agricultural residues, offering a renewable and low-cost raw material for the development of bioplastics. Owing to its excellent mechanical strength, biodegradability and chemical modifiability, cellulose has become a central focus in the development of sustainable packaging materials [12]. However, the native form of cellulose exhibits limited solubility in water and most common solvents, primarily due to its dense network of intra- and intermolecular hydrogen bonds that create a highly crystalline structure. To overcome these limitations, various chemical modification strategies – such as esterification, etherification, and oxidation – are employed to improve its solubility and processability [13-15].

Several cellulose derivatives have found widespread use in bioplastic films, including cellulose acetate, carboxymethyl cellulose (CMC), and hydroxypropyl methylcellulose (HPMC). These materials are valued for their excellent film-forming capabilities and compatibility with bioactive additives. When combined with plasticizers or blended with other biopolymers, they offer improved flexibility, optical clarity, and barrier properties [16,17]. A notable example is the study by Oliveira et al., who developed transparent nanocomposite membranes by incorporating synthetic laponite into a CMC matrix. The addition of clay nanoparticles resulted in enhanced tensile strength, thermal stability, and moisture barrier performance, all while maintaining high optical transparency. Such findings underscore the promise of CMC-laponite composites as eco-friendly materials for advanced packaging applications [18].

Bacterial cellulose (BC), on the other hand, represents a highly pure and structurally unique form of cellulose produced extracellularly by bacteria such as *Komagataeibacter xylinus* [19]. Unlike plant-derived cellulose, BC is free from lignin and hemicellulose and features an intricate nanofibrillar network characterized by high crystallinity, porosity, and exceptional water-holding capacity. These properties, combined with its mechanical robustness and structural uniformity, make BC particularly well-suited for applications in food packaging, biomedical devices, and cosmetics [20, 21].

Moreover, BC exhibits exceptional compatibility with various functionalization strategies, including ionic crosslinking and surface esterification, enabling fine-tuning of its water resistance, mechanical performance, and biological activity [22]. Through composite formulation, BC can be combined with other biopolymers such as starch, chitosan, or poly(lactic acid) (PLA), yielding hybrid materials with enhanced and complementary properties [23]. For example, Xu et al. developed a biodegradable film incorporating BC, chitosan of varying molecular weights, and curcumin to improve both the functional and structural attributes of active packaging materials. The incorporation of BC contributed to improved film uniformity and mechanical strength, while chitosan enhanced barrier performance and helped modulate moisture sensitivity. Curcumin, acting as a bioactive additive, imparted antioxidant functionality to the composite. Structural analysis confirmed uniform dispersion of BC within the polymeric matrix, and the resulting films exhibited good thermal stability along with reduced water vapor permeability – features desirable for food packaging applications [24]. In another study, Shi et al. engineered a BC-based film modified with a guanidine-functionalized polymer and gallic acid to improve its performance in sustainable packaging. The resulting nanocomposite demonstrated a dense and coherent nanofibrous architecture, significantly enhancing its tensile strength and flexibility. Additionally, the film exhibited broad-spectrum antibacterial activity, effectively suppressing the growth of *Escherichia coli* and *Staphylococcus aureus*. Beyond antimicrobial functionality, the material also provided UV protection, antioxidant properties and antifungal effects. Importantly, it showed the ability to biodegrade into non-toxic sugars, underscoring its potential as a safe and environmentally responsible alternative to conventional plastics [25].

Recent work by Liao et al. further illustrates the promise of BC-based composites. Their study focused on the development of multifunctional packaging films incorporating bacterial cellulose, chitosan, and ε-polylysine (ε-PL). The addition of ε-PL at controlled concentrations resulted in reinforced structural properties, as evidenced by higher tensile strength and improved thermal behavior [26]. These enhancements were attributed to increased intermolecular interactions within the composite network. The films also exhibited notable antimicrobial activity against common spoilage microorganisms. When applied to fish preservation, the films effectively prolonged shelf life by mitigating microbial proliferation and moisture loss. The composite's biodegradability and non-toxic profile make it a promising candidate for sustainable, bioactive packaging systems A schematic overview of the preparation steps is presented in Figure 1, which illustrates the integration of ε-polylysine into the polymeric network, contributing to enhanced film structure and bioactivity [26].

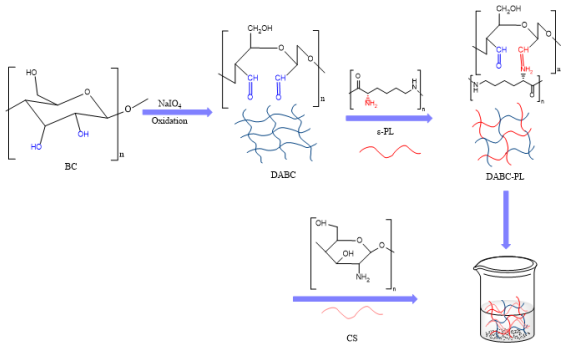

Figure 1 – Schematic illustration of the preparation of chitosan – bacterial cellulose composite films containing ε-polylysine. Adapted from [26]

Collectively, these findings highlight the adaptability and multifunctionality of bacterial cellulose when integrated into advanced material systems. Whether used alone or in combination with plant-derived cellulose, BC-based composites align with principles of circular bioeconomy through their renewable origin, low toxicity, and capacity for customization. The ability to tailor physical, chemical, and biological properties makes these materials particularly relevant for next-generation packaging solutions that balance performance with environmental responsibility [27].

Noncovalent Interactions in Polysaccharide-Based Bioplastics

Noncovalent interactions play a critical role in defining the structural integrity, mechanical strength, and barrier performance of polysaccharide-based bioplastics. These interactions – including hydrogen bonding, ionic interactions, van der Waals forces, and hydrophobic effects –

govern the self-assembly of polymer chains, the compatibility between polymer components, and the integration of functional additives within the matrix [28].

Hydrogen bonding is particularly prevalent in polysaccharide systems due to the high density of hydroxyl, carboxyl, and amine groups. In native cellulose and its derivatives, strong intra- and intermolecular hydrogen bonds form semi-crystalline structures that contribute to the material's tensile strength and stiffness [29]. However, these interactions can also limit solubility and reduce flexibility. To address this, targeted disruption or modulation of hydrogen bonding – through the use of plasticizers such as glycerol or sorbitol, or via blending with other biopolymers like starch or proteins – has been employed to optimize mechanical and moisture barrier properties [30,31]. The significance of hydrogen bonding in biopolymer interactions has been explored through computational methods. For instance, a Density Functional Theory (DFT) study by Pontoh et al. demonstrated that intermolecular hydrogen bonding between amylum (starch) and cellulose results in stable complexes with binding energies ranging from -5 to -15 kcal/mol. The formation of two to three hydrogen bonds per molecular pair was found to enhance the composite's flexibility and resistance to moisture. Reduced Density Gradient (RDG) analysis further confirmed the presence of strong and well-distributed hydrogen bonding interactions between these two polysaccharides (Figure 2) [32].

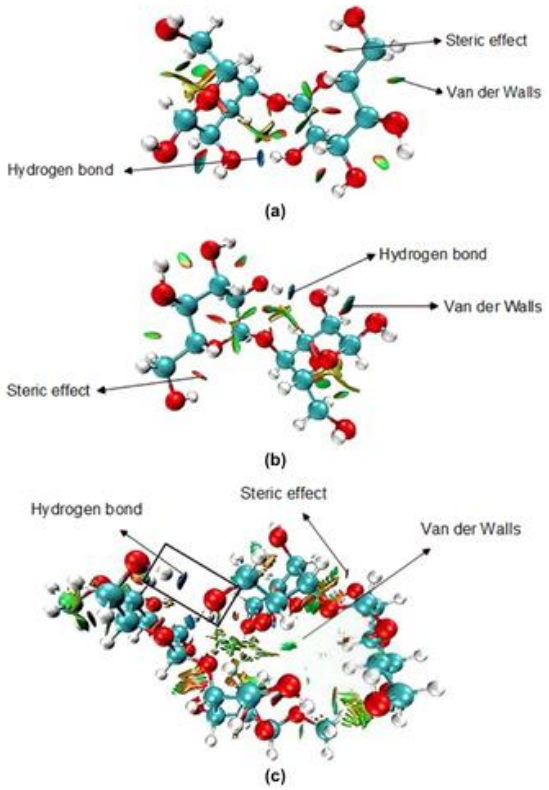

Figure 2 – Reduced Density Gradient (RDG) isosurface (a) amylum, (b) cellulose, (c) cellulose-amylum. Reproduced without modification from [32], licensed under CC BY-NC-ND 4.0

Recent developments in nanocomposite design have shown that incorporating laponite nanoclay into polysaccharide matrices significantly improves structural performance through noncovalent interactions. Yuan et al. reported that the inclusion of laponite in aqueous cellulose dispersions leads to the formation of a physically cross-linked gel network, predominantly stabilized by electrostatic interactions and hydrogen bonds. This internal architecture markedly enhanced the tensile strength and morphological uniformity of the resulting films [33].

Similar findings were observed in starch-based systems. Da Silva et al. demonstrated that adding laponite to cassava starch matrices yields nanocomposite films with lower water content, increased opacity, and superior barrier efficiency. The presence of laponite was found to promote tighter molecular packing and facilitate intermolecular interactions, resulting in films with improved mechanical durability and reduced moisture permeability [34].

Together, these findings highlight the critical role of noncovalent cross-linking mechanisms – particularly hydrogen bonding and electrostatic interactions – in reinforcing the structural

architecture of polysaccharide-based bioplastics [35]. The incorporation of laponite, in this context, emerges as a versatile strategy to tailor the mechanical and barrier properties of films for applications such as sustainable food packaging .

In BC, the inherent nanofibrillar structure stabilized by extensive hydrogen bonding provides exceptional mechanical strength and water retention capabilities. Functional enhancement of BC-based films can be achieved by introducing additional hydrogen-bonding moieties through the incorporation of polymers such as poly(vinyl alcohol), chitosan, or tannic acid, which strengthen intermolecular cohesion and improve overall film performance [36]. Recent advances have also explored the surface functionalization of BC to introduce antimicrobial activity while preserving its biodegradability. For example, lauric acid-modified BC films have shown selective antibacterial efficacy – particularly against *Bacillus subtilis* – without impairing their natural degradation behavior. In biodegradability assays, untreated BC films degraded by over 50% within three days and nearly completely within a week when placed in soil, underscoring their environmental compatibility. This combination of selective bioactivity and rapid degradation positions lauric acid-treated BC as a promising material for active and eco-conscious packaging applications [37]. Additional work by Gea et al. demonstrated the development of bioplastic films from cassava starch and bacterial cellulose, enhanced with *Zanthoxylum acanthopodium* extract. The resulting materials displayed improved tensile strength (2.34 MPa) and measurable antibacterial activity against *Bacillus cereus* (inhibition zone: 10.8 mm), confirming their potential for use in sustainable and functional food packaging systems [38].

Polysaccharides containing ionizable functional groups – such as carboxyls in carboxymethyl cellulose (CMC), amines in chitosan, and sulfate groups in carrageenan – can engage in ionic crosslinking with oppositely charged polymers or multivalent ions, forming polyelectrolyte complexes (PECs). These complexes often exhibit superior cohesion, mechanical strength, and moisture resistance compared to unmodified polysaccharide matrices [39].

For instance, CMC has been shown to form stable electrostatic complexes with biopolymers such as chitosan and alginate, or with metal ions like Ca^{2+} and Zn^{2+}. These interactions contribute to enhanced dimensional stability and in some cases, impart antimicrobial functionality – features particularly desirable in food packaging where material performance and bioactivity are essential [40, 41].

In composite systems, ionic interactions often serve as physical cross-linking points that stabilize the polymer network without requiring chemical crosslinkers or initiators, thus supporting the development of environmentally benign materials aligned with green chemistry principles [42]. A notable example is the work by Blilid et al., who fabricated chitosan-based bioplastics reinforced with phosphorylated micro- and nanocellulose. The introduced phosphate groups facilitated strong polyelectrolyte interactions with the chitosan matrix, leading to significant improvements in mechanical properties, including a tensile modulus increase to 1.649 MPa. Moreover, the composites demonstrated biological functionalities such as antibacterial activity and catalase-like behavior. Scanning electron microscopy (SEM) images (Figure 3) revealed a homogeneous dispersion of cellulose fillers and strong interfacial adhesion, attributed to both ionic and hydrogen bonding interactions. These morphological features directly correlate with the enhanced mechanical and functional performance of the resulting bioplastic films [43].

Transesterification as a Strategy for Hydrophobic and Thermal Modification

Transesterification has gained prominence as a flexible and sustainable chemical approach for modifying polysaccharides to improve their functional performance in bioplastic applications. This reaction involves the exchange of ester groups between a hydroxyl-rich polymer – such as cellulose or its derivatives – and ester-containing compounds, typically in the presence of a catalyst or base [44]. Compared to conventional acylation processes, transesterification offers a greener alternative, particularly when performed in solvent-free media or using environmentally benign solvents [11].

In the context of polysaccharide chemistry, transesterification primarily targets the hydroxyl groups along the sugar backbone. Reaction with ester donors such as vinyl esters, fatty acid methyl esters (FAMEs), or triglycerides leads to the formation of hydrophobic ester linkages. This structural modification reduces polymer polarity, enhances resistance to water and increases thermal stability. It also contributes to slower biodegradation under humid conditions – an advantage for materials intended for controlled degradation in packaging contexts [45].

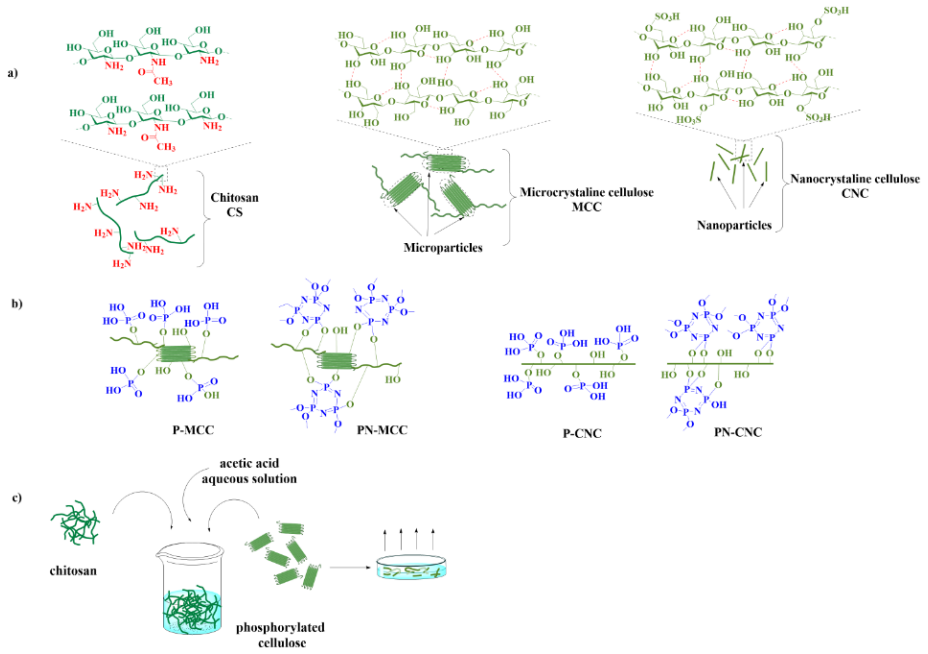

Figure 3 – Schematic representation of the preparation of chitosan films reinforced with phosphorylated cellulose:
(a) Chemical structures of chitosan and cellulose, highlighting structural differences between amorphous microcrystalline cellulose (MCC) and crystalline cellulose nanocrystals (CNC). (b) Structure of phosphorylated cellulose containing ionic phosphate groups. (c) Stepwise fabrication process of composite films and corresponding digital images of chitosan matrices embedded with phosphorylated cellulose. Adapted from [43]

A representative example is the work by Yan et al., who successfully grafted cellulose with poly(L-lactide) (PLLA) chains through ring-opening polymerization of L-lactide in the ionic liquid AmimCl, using 4-dimethylaminopyridine (DMAP) as a catalyst. The resulting cellulose-g-PLLA copolymers exhibited thermoplastic behavior due to the disruption of native hydrogen bonding and the incorporation of flexible ester linkages. These modifications not only improved melt processability and thermal flexibility but also enabled the formation of films and fibers suitable for biodegradable packaging [46]. Another noteworthy example is the study by Onwukamike et al., who developed a direct transesterification protocol using high oleic sunflower oil and cellulose in a DBU-CO_2 switchable solvent system. This reaction proceeds under mild conditions without the need for prior activation or the use of toxic reagents. The resulting cellulose fatty acid esters displayed reduced hydrophilicity, enhanced thermal stability, and favorable mechanical characteristics. These attributes collectively increase the material's suitability for application in sustainable packaging solutions. The reaction mechanism, presented in Figure 4, illustrates the formation of covalent ester linkages between cellulose hydroxyl groups and fatty acid esters, which are central to the enhanced performance of the modified polymer [47].

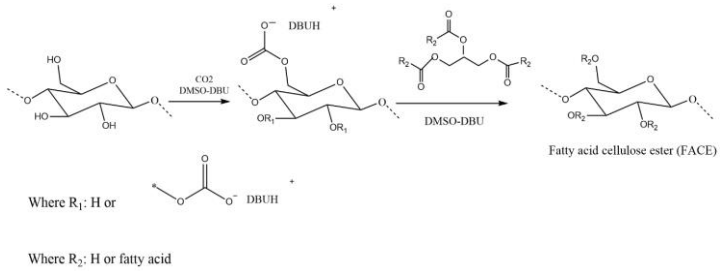

Figure 4 – Proposed mechanism of cellulose transesterification with high oleic sunflower oil in the presence of DBU-CO_2 switchable solvent. Adapted from [47]

Conclusion

Polysaccharide-based bioplastics offer a viable and environmentally sustainable alternative to petroleum-derived polymers, especially for packaging applications where biodegradability, biocompatibility and renewability are critical. Nonetheless, their broader adoption continues to be limited by challenges such as high water sensitivity, insufficient mechanical strength and vulnerability

to environmental conditions. A growing body of research has demonstrated that noncovalent interactions – particularly hydrogen bonding and ionic crosslinking – can significantly improve the structural integrity and functional properties of these materials. At the same time, covalent modification via transesterification has emerged as a practical strategy for reducing hydrophilicity and enhancing thermal resistance, without compromising the biodegradability of the polymer backbone.

The integration of components such as bacterial cellulose, laponite, and bioactive fillers illustrates the potential of combining physical and chemical approaches to develop materials with tailored performance. This synergistic design philosophy enables the creation of bioplastics with improved mechanical robustness, barrier properties, and functional versatility – key attributes for next-generation sustainable packaging. To move these materials closer to real-world application, future research should prioritize scalable and cost-effective synthesis methods, a deeper understanding of structure – property relationships, and compliance with regulatory and industrial standards. Equally important is the evaluation of end-of-life behavior, including biodegradation kinetics, environmental impact, and suitability for composting or recycling systems. Addressing these areas will be essential for translating laboratory innovations into durable, high-performance, and ecologically sound packaging solutions.

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ТҰРАҚТЫ ОРАУЫШ МАТЕРИАЛДАРҒА АРНАЛҒАН ПОЛИСАХАРИД НЕГІЗІНДЕГІ БИОПЛАСТИКТЕР: КОВАЛЕНТТІ ЕМЕС ӘРЕКЕТТЕСУЛЕР МЕН ТРАНСЭТЕРИФИКАЦИЯНЫҢ РӨЛІ

Қоршаған ортада пластик қалдықтарының шектен тыс жиналуы экологиялық жағынан қауіпсіз және ыдырайтын орауыш материалдарын жасауға деген жаһандық қызығушылықты арттырды. Биологиялық ыдырайтын полимерлердің ішінде өсімдік тектес целлюлоза мен оның микробтық аналогы – бактериалды целлюлозадан (БЦ) алынатын биопластиктер жаңартылатындығы, ыдырайтындығы және қолайлы механикалық қасиеттері арқасында ерекше назар аудартып отыр. Алайда, олардың практикалық қолданылуы гидрофильділігі мен сыртқы орта әсерлеріне сезімталдығы сияқты мәселелермен шектеледі. Бұл кемшіліктерді жою үшін соңғы зерттеулерде полисахаридтер құрылымын ковалентті емес әрекеттесулер (мысалы, сутектік байланыстар, иондық айқас байланыстар) және ковалентті модификациялар (мысалы, трансэтерификация) арқылы түрлендіру жолдары қарастырылуда. Мұндай тәсілдер материалдардың механикалық беріктігін, икемділігін және суға төзімділігін едәуір арттыра алады. Осы шолуда полисахарид негізіндегі биопластиктерді жетілдіруге бағытталған соңғы ғылыми жетістіктер талданып, физикалық және химиялық модификацияларды біріктіру арқылы олардың жұмыс сипаттамаларын жақсарту мүмкіндіктері қарастырылады. Әсіресе бактериалды целлюлоза, лапонит, хитозан және май қышқылының эфирлерінен тұратын гибридті жүйелердің синергетикалық әсеріне ерекше көңіл бөлінеді. Жалпы алғанда, ковалентті емес және ковалентті модификацияларды біріктіру – келешегі зор тұрақты орауыш материалдарын жасау үшін тиімді стратегия ретінде қарастырылады.

Түйін сөздер: *бактериалды целлюлоза; сутектік байланыс; иондық айқас байланыс; трансэтерификация; лапонит.*

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БИОПЛАСТИКИ НА ОСНОВЕ ПОЛИСАХАРИДОВ ДЛЯ УСТОЙЧИВОЙ УПАКОВКИ: РОЛЬ НЕКОВАЛЕНТНЫХ ВЗАИМОДЕЙСТВИЙ И ТРАНСЭТЕРИФИКАЦИИ

Усиление проблемы загрязнения окружающей среды пластиковыми отходами стимулировало во всём мире активный поиск экологически безопасных и устойчивых упаковочных материалов. Среди биоразлагаемых полимеров особый интерес вызывают биопластики на основе полисахаридов – особенно из растительной целлюлозы и её микробного аналога, бактериальной целлюлозы (БЦ) – благодаря их возобновляемости, способности к биодеградации и благоприятным механическим свойствам. Тем не менее, их практическое применение часто ограничивается такими факторами, как гидрофильность и уязвимость к внешним воздействиям. Для преодоления этих ограничений в последние годы активно изучаются структурные модификации, включающие как нековалентные взаимодействия (например, водородные связи, ионная сшивка), так и ковалентные подходы, такие как трансэтерификация. Эти методы доказали свою эффективность в повышении механической прочности, гибкости и водостойкости материалов. В данном обзоре рассматриваются современные достижения в области разработки биопластиков на основе полисахаридов с особым акцентом на сочетание физических и химических модификаций для улучшения эксплуатационных характеристик. Особое внимание уделяется гибридным системам с участием бактериальной целлюлозы, лапонита, хитозана и эфиров жирных кислот, демонстрирующим перспективные синергетические эффекты. В целом, интеграция нековалентных и ковалентных модификаций представляет собой перспективную стратегию для создания упаковочных материалов нового поколения, ориентированных на устойчивое развитие.

Ключевые слова: *бактериальная целлюлоза; водородные связи; ионная сшивка; трансэтерификация; лапонит.*

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