

Review

Modification of Cellulose Nanocrystals Using Polydopamine for the Modulation of Biodegradable Packaging, Polymeric Films: A Mini Review

Amanda L. Souza ¹, Victor G. L. Souza ², Meirielly Jesus ³, Fernando Mata ³, Taila V. de Oliveira ¹
and Nilda de F. F. Soares ^{1,*}

¹ Laboratory of Polymeric Materials, Food Technology Department, Federal University of Viçosa, Viçosa 36570-000, MG, Brazil; amanda.souza1@ufv.br (A.L.S.); taila.oliveira@ufv.br (T.V.d.O.)

² International Iberian Nanotechnology Laboratory, 4715-330 Braga, Portugal; v.souza@fct.unl.pt

³ CISAS—Center for Research and Development in Agrifood Systems and Sustainability, Polytechnic Institute of Viana do Castelo, Rua da Escola Industrial e Comercial Nun'Alvares 34, 4900-347 Viana do Castelo, Portugal; meiriellyjesus@ipvc.pt (M.J.); fernandomata@ipvc.pt (F.M.)

* Correspondence: nfsoares10@gmail.com

Abstract: This review delves into environmentally conscious sustainable packaging materials, focusing on biodegradable polymers and innovative surface modification methodologies. Synthetic plastics have revolutionized various industries due to their physical attributes and affordability, particularly in packaging applications. Nonetheless, the substantial volume of plastic waste, especially from non-biodegradable sources, has provoked heightened environmental apprehensions. Notably, polymers derived from natural sources, such as cellulose, are classified as biopolymers and esteemed for their ecological benevolence. Among these, cellulose and its derivatives stand out as renewable and abundant substances, holding promise for sustainable packaging solutions. Nano-sized cellulose fibers' incorporation into biodegradable films garners interest due to their remarkable surface area, robust mechanical strength, and other commendable properties. Surface modification techniques, such as a polydopamine (PDA) coating, have been explored to improve the dispersion, interfacial compatibility, and mechanical performance of cellulose nanocrystals (CNC) when incorporated into biodegradable polymer films. In this sense, PDA, derived from mussel proteins' dopamine component, displays exceptional adhesion to diverse surfaces and has been extensively scrutinized for its distinctive attributes. Therefore, the core focus of this review was to approach ecologically friendly packaging materials, specifically investigating the synergy between CNC and PDA. The unparalleled adhesive characteristics of PDA serve as a catalyst for enhancing CNC, thereby elevating the performance of biodegradable polymers with potential implications across various domains.

Keywords: biopolymers; dopamine; surface modification; nanoparticle



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1. Introduction

According to ASTM standard D883-18 [1], biodegradable plastics are defined as materials capable of being decomposed by natural microorganisms into carbon dioxide, water, biomass, and/or methane under appropriate environmental conditions. This degradation process occurs through extracellular enzymes that break down polymer chains, enabling their assimilation [2].

Biodegradable polymers can originate from petrochemical or renewable sources. Among natural polymers—such as starch, cellulose, chitosan, and proteins of a plant or animal origin—biopolymers stand out due to their renewable origin and, in many cases, their biodegradability [3]. These materials have gained prominence as more environmentally sustainable alternatives to petroleum-derived plastics. Among them, cellulose is particularly attractive due to its abundance, low cost, biodegradability, and excellent film-forming capability [4].

Nanocellulose, particularly cellulose nanocrystals (CNCs), has been widely investigated as a reinforcing agent in biodegradable polymer matrices due to its high crystallinity, large surface area, and superior mechanical properties [5]. However, its high hydrophilicity and limited compatibility with hydrophobic polymers restrict its application in packaging films that require enhanced barrier properties and stability.

In this context, surface modification strategies for CNCs have been proposed to expand their functionality and compatibility with various matrices. Polydopamine (PDA), a bioinspired polymer formed by the self-oxidation of dopamine in an alkaline medium, has emerged as a promising alternative for surface functionalization. PDA exhibits strong adhesion, chemical stability, biocompatibility, and the ability to anchor various functional groups [6–8].

The modification of CNCs with polydopamine has the potential to enhance interfacial interactions with the polymer matrix, promoting the improved cohesion, mechanical strength, and overall performance of the resulting films [9]. These properties are especially desirable in the development of biodegradable packaging materials, which require mechanical resilience, flexibility, and potentially additional functionalities such as a gas barrier or antimicrobial activity.

Therefore, a focus on nanoscale cellulose fibers' modification and their potential in enhancing biodegradable films will be realized through information gathering in the literature, besides a new perspective in material science. The global packaging industry continues to grow, but with it, environmental concerns related to plastic waste also rise. In this context, the investigation of cellulose nanocrystals modified with polydopamine to improve the adhesion capacity of surfaces may broaden material applications, enabling the functionalization of biodegradable polymers.

2. The Importance of Biodegradable Packaging

Biodegradable polymers are materials that can easily break down into simpler compounds when discarded in the environment. Microorganisms metabolize them, producing CO₂, water, and biomass as the byproducts [10]. These remnants are not visually distinguishable and pose no risk of hazardous environmental emissions [11]. The speed of this biodegradation process hinges on the temperature, humidity, and the amount and type of microorganisms present [12]. For rapid degradation, all three prerequisites must be fulfilled. Consequently, the significant advantage of using such polymers over conventional ones lies in their degradation time. Unlike conventional plastics, which are generally incinerated or left in landfills for centuries [13], biodegradable plastics can break down within weeks to months [14].

The biodegradability of polymeric materials is also affected by their chemical structure. This structure determines the stability of the functional group, reactivity, and hydrophilicity. Other important factors include physical and physico-mechanical properties, such as the molecular weight, porosity, and morphology (crystalline and amorphous) [15].

Biodegradable polymers can originate from both renewable natural resources and non-renewable sources [14]. Materials derived from renewable sources like plants, animals, and microorganisms are also referred to as biopolymers or biomaterials [16]. Biopolymers

fall into four main categories: those obtained from biomass, those resulting from microbial metabolism, those derived from biotechnological processes, and those originating from petrochemical products [14]. Developing polymeric products from biomass holds substantial promise for replacing synthetic polymers and diminishing global reliance on fossil fuels. This promise is due to their affordability, abundance, and renewable nature. Among the various options, proteins and polysaccharides are particularly notable [17].

Of the polysaccharides, cellulose stands out as a potential substitute for petroleum-based polymers due to its eco-friendly characteristics, including renewability, biocompatibility, biodegradability, and accessible cost [18,19].

3. Cellulose: Structure and Functions

Cellulose stands out as the most prevalent organic compound within the Earth's outer layer, constituting the primary building block of biomass. Brazil is the second-largest pulp producer in the world, with an annual production of 21 million metric tons, including 18.2 million metric tons of short-fiber pulp, primarily sourced from *Eucalyptus* spp. [20]. It is a high-molecular-weight polymer, renewable in nature, prominently found in the cellular walls of plants (comprising 40–60% of the structural components), as well as in certain algae cell walls, and through synthesis carried out by bacteria [21]. Typically, cellulose is obtained through the physical, chemical, and enzymatic breakdown of plant material. Within plant structures, cellulose is encompassed by two key components: hemicellulose and lignin, both of which form amorphous structures [22]. Noteworthy examples of cellulose sources are wood (constituting 40–50% of its weight), cotton (87–90% by weight), jute (60–65% by weight), flax (70–80% by weight), and ramie (70–75% by weight) [23].

Cellulose is a linear polysaccharide constructed from repeating β -(1 $\rightarrow$ 4)-D-glucopyranose units, interconnected through covalent acetal linkages. In this arrangement, the C1 carbon of one glucose ring forms a glycosidic bond with the C4 carbon of an adjacent glucose ring [23]. This chemical arrangement prompts a twisting pattern along the cellulose chain, alternating by 180°. Each glucose unit encompasses three hydroxyl groups at positions C2, C3, and C6, giving rise to intra- and intermolecular hydrogen bonding. The active –OH groups located at positions C2, C3, and C6 in cellulose govern its chemical modifications and properties [24]. When intramolecular and intermolecular hydrogen bonds arrange the cellulose molecule, this leads to the classification as either native cellulose or cellulose I (refer to Figure 1).

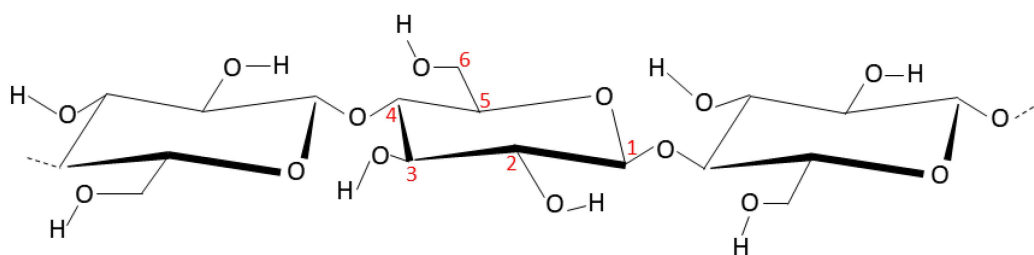


Figure 1. Molecular structure depicting intermolecular hydrogen bonding in the cellulose I system. Source: the authors. The numbering of carbon atoms in the molecule, following IUPAC rules, identifies the position of substituents and functional groups in the main chain.

Intramolecular hydrogen bonds link the hydroxyl group of the C3 position to the nearest ether oxygen located in the neighboring anhydroglucose units belonging to the same chain. In addition, it links the oxygen atoms in the hydroxyl group (–OH) of the C6 position to the adjacent –OH bond of the C2 position (Figure 1). These bonds are responsible for the stiffness of the cellulose polymer [24].

Regarding the cellulose II type, hydrogen is linked in many ways. Intramolecular bonds within the structure are unfavorable due to the preferential pre-existing hydrogen

bonds between the hydroxyl groups of C6 and C2 from the neighboring chain. Additionally, intermolecular hydrogen bonds can form between the –OH group from C2 of one chain and the –OH group from C2 of the nearest adjoining chain (Figure 2).

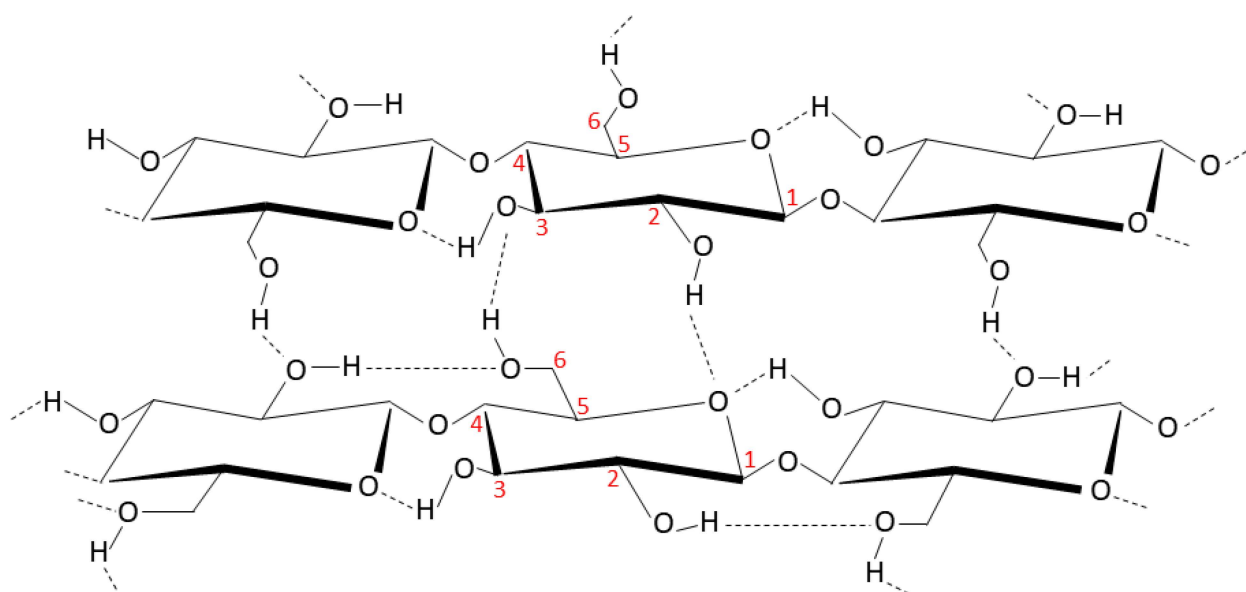


Figure 2. Chemical structure depicting the intermolecular hydrogen bonding interaction within the cellulose II system. Source: the authors. The numbering of carbon atoms in the molecule, following IUPAC rules, identifies the position of substituents and functional groups in the main chain.

Interactions within and between molecules play a significant role in the characteristics of cellulose. These characteristics encompass crystallinity, chirality, hydrophilicity, and solubility, besides various physical, chemical, and thermal properties [25,26]. All features also depend on the length of the polymer chain, which is contingent on the source of the cellulose. Typically, the number of glucose units or the extent of polymerization in natural lignocellulose can be remarkably high, reaching up to 10,000 units [27]. In essence, cellulose cannot be found as a pristine and isolated molecule in nature; instead, it takes the form of a geometric fiber composed of multiple molecular chains of cellulose [28]. Depending on the plant species, a range from 18 to 36 individual cellulose chains are attached, held by hydrogen bonds, to create larger entities known as elementary fibrils. These elementary fibrils are then compacted into larger microfibrils and bundles of microfibrils, also referred to as macrofibrils. Furthermore, these microfibrils and macrofibrils, along with hemicellulose and lignin, constitute the essential structure of plants' cell walls, which can be attached to other components, like proteins and inorganic compounds, forming the entire plant structures [28,29]. Within each microfibril, the cellulose molecular chain is composed of two regions, one with a highly ordered structure (crystalline) and the other being disordered (amorphous). The crystalline regions are responsible for a dense structure, while the disordered regions are randomly entangled [28,29].

In this context, due to the substantial molecular weight and tight packing of linear cellulose chains, besides the robust intra- and intermolecular hydrogen bonds, cellulose does not dissolve in water, has a limited water absorption capacity (roughly 8–14% of water at 60% relative humidity and a temperature of 20 °C), and modestly dissolves in organic solvents [23,28,29].

Cellulose can be isolated using several techniques to characterize the morphology and size, in which the features can be modulated based on the natural source and extraction process employed. In this context, owing to their unique biodegradability and impressive

physical and mechanical attributes, cellulose and nanocellulose, as well as their derivatives, can be engaged in the food packaging industry.

Cellulose-derivate polymers can be utilized to manufacture various polymeric composites since they are non-corrosive, non-toxic, and come from globally abundant sources, making them cost-effective [30]. Nevertheless, cellulose and its derivatives have limitations, including a lack of plasticity, reduced or absent antibacterial properties, low dimensional stability, insolubility in water, and limited solubility in organic solvents [31]. For that, research is focused on modifying cellulose to mitigate these challenges.

4. Nanocellulose

Nanotechnology is currently acknowledged as a driving force that is boosting a new industrial revolution spanning multiple interdisciplinary sectors. Nanomaterials are characterized as materials with at least one external dimension ranging from 1 to 100 nm and they have been explored as reinforcement materials. Interest in nanomaterials has surged due to their impressive specific surface area relative to their mass ($>100 \text{ m}^2\text{g}^{-1}$), enabling a more effective interaction with the continuous phase of composites compared to their micrometer-sized counterparts [32,33]. Among nanocompounds, nanocellulose, a natural fiber derived from cellulose or produced by microorganisms, stands out as a biodegradable material with a low weight and density (around 1.6 g/cm^3). It boasts remarkable mechanical and thermal resistance [28]. Thanks to the removal of its amorphous regions during the downsizing process, leaving only the crystalline structure, it possesses unique traits like high rigidity (reaching up to 220 GPa in the elastic modulus), surpassing even Kevlar fiber, and outstanding tensile strength (up to 10 GPa), exceeding cast iron. Notably, nano-cellulose's strength-to-weight ratio is eight times greater than stainless steel [33]. Additionally, nanocellulose exhibits transparency and boasts a high density of reactive hydroxyl groups on its surface, allowing for functionalization to fine-tune various surface properties [34].

Nanocellulose can be classified into three primary categories: cellulose nanocrystals (CNC), cellulose nanofibers (CNF), and bacterial nanocellulose (BCN). While these types share similar chemical compositions, they differ in terms of their morphology, particle size, crystallinity, and others, owing to variations in the sources and extraction methods [35].

Bacterial nanocellulose is an exopolysaccharide which has been biotechnologically manufactured. Bacteria can synthesize ultrafine cellulose fibrils ranging from 50 to 80 nm in diameter and 3 to 8 nm in thickness, creating a three-dimensional network structure on a micro- to nanoscale [36]. Bacterial nanocellulose is produced by various aerobic bacteria, including *Achromobacter*, *Alcaligenes*, *Aerobacter*, *Agrobacterium*, *Azotobacter*, *Komagataeibacter* (formerly *Gluconacetobacter*), *Pseudomonas*, *Rhizobium*, *Sarcina*, *Dickeya*, and *Rhodobacter* [37]. The extensively studied *Komagataeibacter* genus can assimilate a wide range of carbon and nitrogen sources [38]. BCN is characterized by its remarkable purity (in contrast to plant cellulose closely associated with hemicellulose and lignin) and excellent properties. These include a distinctive nanostructure, substantial water retention capacity (bacterial nanocellulose has the potential to absorb water more than 100 times its dry weight) [39], exceptional mechanical strength (165 MPa a 208 MPa) [40], a high degree of polymerization (2000–8000), pronounced crystallinity (84–89%), as well as impressive biocompatibility and biodegradability [34,36,41].

BCN fibers, being a hundred times finer than plant-derived cellulose, exhibit a unique morphology of a porous and three-dimensional network. The ease of sterilization and good compatibility with living tissues make BCN a suitable substance primarily for biomedical purposes [42]. This attribute leads to increased and enhanced interactions with adjacent components and molecular segments [43]. Although the purification of BCN is more cost-

effective and environmentally friendly, its primary drawback remains its high production cost and limited productivity, serving as a constraining factor [34,37,44]. Consequently, nanofibers extracted from plants continue to be extensively researched and sought-after in the materials sector. Furthermore, given that the agricultural and food sectors generate a significant amount of waste, which can be used as a raw material for obtaining nanofibers, this utilization of agro-industrial waste would, in turn, contribute to the circular economy, waste reduction, and the creation of a high-value-added product.

Cellulose nanofiber is also known as nanofibrillar cellulose [45], cellulose nanofibrils [46], and fibrillated cellulose [47]. They are structures considered long, entangled, and flexible [28]. Typically, the dimensions span from 1 to 100 nm in diameter and 500 to 2000 nm in length, as reported by reference [33]. These fibers also encompass two regions, amorphous and crystalline, at the individual fiber level. The amorphous region contributes to the flexibility and plasticity of materials, while the crystalline region contributes to their rigidity and elasticity [48]. CNF exhibits some interesting properties, such as being non-toxic and non-abrasive. In the work reported by [49], they obtained CNF from wood with high mechanical strength (13 MPa) due to the crystalline ordering of the glucan chains in the cellulose fibrils, which is approximately 50 times higher than cellulose foam and more than 30 times greater than cellulose foam (stronger thermal insulation materials used commercially) [28].

Cellulose nanofibers are acquired using various techniques: chemical, mechanical, and enzymatic treatments, either employed individually, sequentially, or in combination [50,51]. Some of the methods employed encompass enzymatic hydrolysis [52], dissolution in N, N-Dimethylacetamide (DMAc)/LiCl—commonly known as catalytic oxidation [53]—the employment of ionic liquids [54], chemical hydrolysis [55,56], high-pressure homogenization [13,57], steam explosion within a high-pressure autoclave [58], milling [59,60], electrospinning [61,62], high-intensity ultrasonication [61,62], and cryocrushing [63,64].

To be more specific, mechanical methods, even though they are effective in producing CNF, come with several drawbacks. These include low efficiency, a high cost for the process, and a substantial energy requirement to break down the highly organized hydrogen bonds and the densely interconnected cellulose structure [65]. As a result, in order to reduce energy consumption, various pre-treatment technologies are employed prior to the mechanical procedures. These techniques modify the material's properties for commercial use. Some notable examples of pre-treatment methods include enzymatic hydrolysis, mild acid hydrolysis, phosphorylation, periodate oxidation, carboxymethylation, and the oxidation of 2,2,6,6-tetramethylpiperidine 1-oxyl (TEMPO) [66].

From cellulose nanofibrils, it is feasible to extract a substance called nanocrystalline cellulose, commonly referred to as cellulose nanocrystals [5], cellulose nanowhiskers [67], and crystalline nanocellulose [68]. These nanocrystals can take on shapes reminiscent of rods or elongated structures, with lengths ranging from 100 to 500 nanometers and diameters spanning 2 to 20 nanometers. This material is notable for its highly organized crystalline composition [28]. The extraction of nanocrystals from cellulose or materials containing cellulose primarily entails acid hydrolysis, involving the selective removal of the less structured part of the cellulose [69,70].

CNC exhibits intriguing characteristics within the realm of Materials Science. These include a substantial surface area ($\sim 150 \text{ m}^2 \text{ g}^{-1}$), impressive tensile strength (reaching up to 9500 MPa), flexural resistance (approximately 10 GPa), a Young's modulus of around 150 GPa, and a significant aspect ratio (~ 70) [5,65,71]. The crystallinity indices of CNC vary depending on the source of the lignocellulosic material, as well as the time of the purification and fiber hydrolysis processes. It is worth pointing out the interconnection of

crystallinity and the rigidity of cellulose. In simpler terms, the increase in the percentage of crystalline regions enhances the stiffness and tensile strength in the fibers [5].

Besides the nanocellulose advantages, unlike CNF, the drawbacks of CNC include limited flexibility due to the removal of amorphous regions and the heterogeneity of its diameters [34]. Nanocellulose's reactive hydroxyl groups on the surface allow for the adjustment of its properties through chemical modifications. Consequently, this modification reduces the interaction between particles, prevents clumping, and promotes the even distribution of these compounds within a polymer matrix or solvent. As a result, chemical modification becomes essential to enhance the CNC's ability to disperse better and integrate smoothly into the production of polymeric nanocomposites [34].

5. Modification of Nanocellulose

The modification of the nanocellulose surface forwards the enhancement of the properties to extend its potential use [5], including the packaging sector.

There are several techniques available to alter the surface properties of nanocellulose. These methods include the physical adsorption, chemical modifications, and covalent attachment of polymers. The primary aim of surface modification is to introduce new functional groups or essential biological components into the nanostructure without causing harm to the nanoparticle itself [72].

There are several techniques to modify the nanocellulose (NC) surface. To modify nanocellulose, more polarity ester or acetyl groups can be introduced onto its surface using the esterification/acetylation method, which improves its hydrophobic characteristics and makes it more dispersible in nonpolar organic solvents and polymeric matrices [72,73]. One of the methods is the silylation process that incorporates silyl groups on the surface of the NC, increasing its compatibility with non-polar polymers [73]. On the other hand, to improve the compatibility of NC with polar polymers, there is the method known as TEMPO-mediated oxidation, which involves the oxidation of the nanocellulose surface using TEMPO and sodium hypochlorite, introducing carboxyl groups on the surface of the NC [74]. In addition, the phosphorylation method introduces phosphate groups [75] and the amidation method introduces amide groups, improving the polarity of the NC [76]. Non-covalent cross-linking involves the use of interactions (mainly hydrogen bonds) to link nanocellulose to other compounds/materials. The grafting method involves attaching functional groups to the nanocellulose surface through chemical reactions or physical adsorption [77].

The selection of a suitable surface modification method depends on the desired properties of the nanocellulose and its intended application. Notably, researchers are increasingly interested in the grafting technique utilizing PDA for surface modification [78,79]. This approach holds promise in developing various biomaterials due to its ability to interact with different surfaces through distinct mechanisms [80].

6. Polydopamine-Modified Cellulose Nanocrystals and Application in Biodegradable Polymeric Films

Polydopamine is a biomimetic polymer originally reported in 2007 [81]. Bio-mimetics is a scientific field that studies natural phenomena and processes to design practical materials and systems that can replicate the structure and functions of native biological systems [82]. In this context, the adhesive properties of marine mussels served as inspiration for investigating a mechanism in which a protein called Mytilus edulis 5 (Mefp-5), responsible for this phenomenon, was found to be abundant in 3,4-dihydroxy-L-phenylalanine (dopamine) and lysine amino acid units [83]. The desire to comprehend and reproduce this mechanism led to the creation of polydopamine—a biocompatible polymer known for

its robust adhesive properties (capable of adhering to almost any kind of inorganic and organic surface).

The synthesis of polydopamine involves the oxidative polymerization of dopamine monomers containing both catechol and amine functional groups. This process occurs under alkaline conditions commonly found in marine environments (Figure 3).

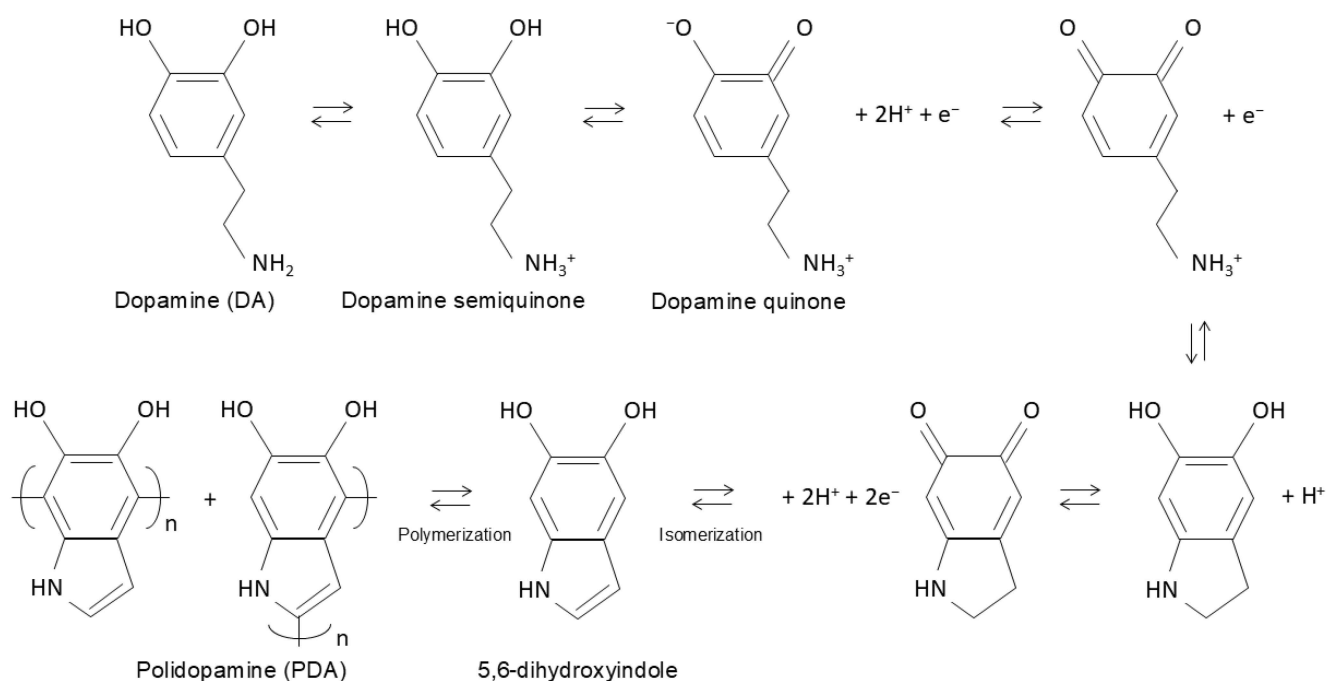


Figure 3. Scheme of the dopamine polymerization mechanism [28] (adapted from Liu et al., 2014).

However, the precise mechanism behind the formation of polydopamine is not yet fully comprehended [77]. In most cases, dopamine polymerization follows a chemical process documented by Lee et al. [81]. A potential mechanism of the polymerization process is depicted in Figure 3, in which the molecular structure of dopamine holds two phenolic groups, susceptible to deprotonation, alongside a primary amine group that can be protonated, enabling the occurrence of three dissociation constant values (pKa) [83].

When the pH remains below 8.5 (around 7.5), the primary amine group of dopamine becomes protonated, preventing the dissociation of hydroxyl groups. As the pH approaches 8.5, the hydroxyl group with higher acidity dissociates, resulting in an equilibrium state. The suggested oxidation mechanism at pH = 8.5 is through an electron transfer reaction, forming a semiquinone radical. Subsequently, this radical transforms into dopamine quinone. The dopamine quinone structure encompasses a ring deficient in electrons and an amine group that contributes to electron sharing. The addition of 1,4 Michael and the deprotonation of the amine group can induce an intramolecular cyclization reaction. This series of events eventually triggers oxidation, causing the conversion into 5,6-dihydroxyindole. This compound exhibits self-polymerization properties, resulting in polydopamine [80].

Biodegradable polymer films combining cellulose nanocrystals with polydopamine have shown potential for various sustainable applications. The addition of CNC improves the mechanical strength and thermal stability, while PDA facilitates surface adhesion and functionalization. Studies like that of Xu et al. [84] demonstrate that incorporating cellulose nanocrystals decorated with polydopamine significantly enhances the properties of polylactic acid (PLA). PDA acts as a bridge between CNC and the PLA matrix, increasing the tensile strength and elongation at the break. Moreover, PDA provides UV protection to the composite, making it promising for packaging food and light-sensitive products. This

approach offers an efficient strategy to overcome the low compatibility between CNC and PLA, extending the material's lifecycle and applications [84].

In another study, dopamine was self-polymerized to coat CNC surfaces, combining their functional and structural characteristics in polyvinyl alcohol (PVA) films. The resulting films exhibited mechanical and thermal improvements, such as an increased Young's modulus, higher tensile strength, and greater elongation at the break. The decomposition temperature rose to 328.2 °C with 15% nanocrystals of cellulose modified with polydopamine (PDA @ CNC) loading. Additionally, the presence of PDA provided UV protection, eliminated free radicals, and reduced oxygen and water vapor permeability, highlighting its potential for advanced packaging [85].

In another study, cellulose nanocrystals were functionalized with polyethylene glycol (PEG) of different lengths (1, 2, 5, and 10 kDa) via PDA in an aqueous solution. These nanocrystals of cellulose functionalized with polyethylene Glycol (CNC-PEGs) reinforced PVA films at various concentrations (0 to 7 wt%). Short PEG chains increased the stiffness and strength due to a more efficient stress transfer, while longer grafts formed rubbery layers, enhancing the toughness of the composites. The results demonstrated how the PEG length affects the mechanical properties and flexibility of PVA/CNC nanocomposites [86].

Modifying nanocellulose with polydopamine offers notable benefits: (i) PDA improves interfacial adhesion between nanocellulose and the polymer matrix, resulting in substantial increases in mechanical strength and elongation at the break; (ii) an enhanced mechanical performance makes the films more durable and suitable for practical applications; (iii) PDA provides UV protection, essential for preserving light-sensitive products; and (iv) it reduces oxygen and water vapor permeability, extending the shelf life of packaged products. However, this approach faces challenges, including production costs, environmental stability, and scalability. Despite the challenges, research in this area is promising, as it may lead to the development of sustainable materials that meet the growing demand for eco-friendly solutions, driving innovations in packaging and industrial applications. To advance in the material field, the literature suggests that future investigations should focus on optimizing modification processes and evaluating the environmental stability of the composites. New research will not only deepen the understanding of the interactions between components, but also expand opportunities for sustainable applications, which are essential to meet current demands.

Moreover, several studies highlight the importance of modifying CNC with PDA to create materials with enhanced properties for industrial use, particularly in biodegradable packaging [84–86]. The research suggests that these advanced materials can significantly reduce the environmental impact of packaging materials while providing a better performance. A notable example is the use of PDA-modified CNCs to stabilize the dispersion of MXenes (MXene is a new class of graphene like two-dimensional transition metal carbon (nitrogen) compounds obtained by selectively etching specific atomic layers from multiple layered carbon (nitrogen) compounds), forming PDA-coated CNC-MXene sheets that, when incorporated into polyacrylamide hydrogels, result in conductive and transparent materials with high conductivity which can be used in transparent electronic sensors for health monitoring [87]. Another relevant study demonstrated that modifying CNCs with polyethyleneimine and polydopamine (PPCs) resulted in highly effective, biocompatible antibacterial agents, which were incorporated into PLA films, improving both the antibacterial and mechanical properties of the material [88]. Additionally, the immobilization of enzymes, such as lipase, on PDA-coated CNCs demonstrated an efficient and sustainable method to increase the stability and reuse of biocatalysts, with applications in biocatalysis at the oil–water interface [89]. In this context, expanding the scope of future studies to focus on large-scale production processes and assessing the long-term performance

of PDA-modified CNC composites under real-world conditions will be crucial for their successful implementation in commercial applications. The continuous exploration of new functionalization techniques and material combinations will continue to drive the development of highly efficient and eco-friendly materials for a variety of applications, including packaging, textiles, and medical devices. A recent study [90] demonstrated that the modification of CNCs with polydopamine significantly improved their interaction with cellulose acetate matrices. This resulted in the enhanced dispersion, improved stress transfer, and overall reinforcement of the film. Theoretical analysis further confirmed the stability of dopamine polymerization at the CNC surface, offering valuable insights into the mechanisms of interfacial bonding. These findings support the growing potential of PDA-modified CNCs for developing sustainable and high-performance biopolymer packaging materials.

The incorporation of PDA-modified nanocellulose into polymer matrices has shown improvements in the tensile strength, barrier properties, and interfacial adhesion. These enhancements vary depending on the type of polymer, filler, and application target. A concise overview is presented in Table 1, summarizing key polymer–filler systems and their reported benefits.

Table 1. Polymers reinforced with PDA-modified cellulose nanocrystals for packaging applications.

Polymer Matrix	Filler Type	Key Improvements	Reference
PLA	CNC-PDA	↑ Tensile Strength, ↑ UV barrier, ↑ Interfacial adhesion	[84]
PVA	CNC-PDA	↑ Modulus, ↓ Permeability, ↑ UV protection	[85]
PVA	CNC-PEG-PDA	Tailored strength and toughness	[86]
Cellulose Acetate	CNC-PDA	↑ Reinforcement, ↑ Interfacial bonding	[90]

↑ The symbol refers to increasing and ↓ decreasing properties.

As summarized in Table 1, the incorporation of PDA-modified cellulose nanocrystals into various biodegradable polymer matrices has consistently led to improvements in mechanical strength, barrier properties, and interfacial adhesion—key attributes for packaging materials. These findings underscore the versatility and effectiveness of polydopamine as a surface modifier for nanocellulose, offering a promising route toward the development of sustainable, high-performance packaging systems. While the results are encouraging, continued research is needed to optimize formulations, understand structure–property relationships, and explore the scalability of these materials for real-world industrial applications.

7. Future Research Directions in Sustainable Polydopamine-Modified Nanocellulose for Biodegradable Packaging

The integration of PDA with CNC presents significant opportunities for advancing sustainable and biodegradable packaging materials. However, further research is required to optimize the performance and ensure scalability for widespread commercial adoption. Future studies should focus on the following key areas.

Enhancing mechanical and barrier properties sustainably: Optimizing CNC modification techniques to improve the mechanical performance while maintaining environmental compatibility. Enhancing thermal stability using bio-based cross-linkers to support high-temperature applications. Developing natural coatings to further reduce oxygen and moisture permeability, extending the food shelf life without synthetic additives.

Integration with other biodegradable and renewable materials: Exploring dual functionalization with PDA and other natural polymers to improve compatibility and biodegradability. Combining PDA-modified CNC with other plant-derived biopolymers, such as chitosan or starch, for fully renewable packaging solutions. Investigating natural antimicrobial agents to

enhance food preservation without the use of chemical preservatives. Exploring waste-derived bio-fillers to maximize material circularity and reduce raw material consumption.

Biodegradability and environmental impact: Conducting real-world degradation studies to assess the complete life cycle of PDA-modified CNC materials. Evaluating compostability and integration into existing waste management systems to minimize environmental footprint. Investigating the potential for reprocessing and upcycling PDA-CNC composites into second-life applications.

Scalability and industrial feasibility: Developing cost-effective and energy-efficient manufacturing methods to enable large-scale production. Collaborating with industries to assess real-world performance and regulatory compliance. Conducting comprehensive life-cycle assessments to compare the economic and environmental benefits of PDA-CNC composites versus traditional plastics.

Expanding sustainable applications beyond packaging: Investigating PDA-modified CNC for sustainable biomedical applications, such as eco-friendly wound dressings and biocompatible implants. Exploring its use in renewable electronic materials, such as flexible sensors and conductive bio-based films. Developing PDA-CNC hybrid composites for water filtration and environmental remediation to support cleaner ecosystems.

While polydopamine has shown promising results, future studies should compare its performance and environmental impact with other bio-based modifiers, such as tannic acid, lignin derivatives, or synthetic catecholamine analogues. These alternatives may offer distinct interactions with cellulose surfaces and potentially lower costs or toxicity.

The origin of polydopamine as a derivative of dopamine—a major catecholamine in human neurophysiology—invites philosophical and scientific reflection. Could residual dopamine or its analogs in packaging materials influence human neurochemical states? While speculative, such questions underline the importance of considering the biointerface between advanced materials and human biology.

8. Conclusions

Petrochemical plastics are widely used due to their advantageous properties and cost-effectiveness, especially in the field of packaging. However, the environmental concerns posed by non-biodegradable plastic waste, a significant portion of the total plastic waste, cannot be ignored. Biopolymers, such as natural polymers like cellulose and its derivatives, have immense potential for sustainable packaging due to their abundance and inherent biodegradability. A noteworthy candidate is nanoscale cellulose fibers due to their high surface area. When applied to biodegradable films, they promise to improve their characteristics. Innovative surface modification techniques, like the application of a polydopamine coating derived from mussel proteins, can modulate nanocellulose compounds' properties for their incorporation into polymeric matrices, improving the films' features. The nanocellulose coated with PDA primarily enhances the compatibility between biodegradable polymeric matrices, raising performance standards in their respective packaging sector applications.

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Abbreviations

The following abbreviations are used in this manuscript:

BCN	bacterial nanocellulose
CNC	cellulose nanocrystals
CNC-PEG	polyethylene glycol
CNF	cellulose nanofibers
NC	Nanocellulose
PDA	Polydopamine
PEG	polyethylene glycol
PET	polyethylene Terephthalate
PLA	polylactic acid
PVA	polyvinyl alcohol
TEMPO	2,2,6,6-tetramethylpiperidine 1-oxyl

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