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Review

Processing and Formulation Strategies for PFAS-Free Food Packaging: Nanocellulose–Natural Wax Hybrid Coatings for Barrier Applications

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Abstract

The environmental impacts of fossil-based plastic food packaging and the use of grease-resistant coatings containing per- and polyfluoroalkyl substances (PFAS) are driving the development of bio-based and fluorine-free alternatives. Nanocellulose and natural waxes are promising components for PFAS-free barrier coatings, but their performance depends strongly on how they are produced, formulated, deposited, and dried. Nanocellulose provides excellent oxygen-barrier performance, particularly under dry conditions, whereas natural waxes contribute primarily to moisture and grease resistance. Their combination can therefore provide complementary barrier functions, while also introducing challenges related to dispersion, interfacial compatibility, crystallization, and coating uniformity. This review critically examines the replacement of PFAS-based coatings on food paper packaging with formulations derived from nanocellulose and natural wax. The material selection and processing strategies are addressed in relation to component structure, formulation, and performance. Key contributions, compared to previous studies, include an explanation of how the combination of nanocellulose, natural waxes, and complementary barrier materials meets the diverse functional requirements of food packaging. Likewise, the study examines how formulation parameters and processing conditions can determine interfacial compatibility, coating morphology, and barrier performance. Particular attention is given to natural wax processing, including dispersion and emulsion formation, and to formulation parameters that control interfacial interactions, coating structure, surface morphology, and barrier performance. Coating and deposition methods, drying and film formation, and the effects of processing conditions on oxygen, moisture, and grease resistance are evaluated. The review also addresses mechanical performance, sustainability and end-of-life considerations, process scale-up, economic aspects, and compatibility with industrial coating technologies. Overall, nanocellulose–wax hybrid systems offer a promising processing platform for application-specific PFAS-free barrier coatings, although further optimization and validation under realistic food-packaging conditions are required for industrial implementation.

Keywords: nanocellulose; natural waxes; food packaging; PFAS-free coating; coating processes; formulation; barrier properties

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1. Introduction

Plastic packaging represents a major environmental challenge because of its high production volume, short service life, and persistence after disposal. Packaging accounts for a substantial fraction of global plastic use, with food packaging representing approximately 50% of plastics derived from fossil fuels [1,2]. Conventional food-packaging materials include polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), and polyethylene terephthalate (PET), which are widely used because of their low cost, processability, chemical resistance, and effective barrier properties [3]. However, their environmental impact is associated with fossil-resource consumption, waste generation, and recycling limitations, particularly for multilayer structures and food-contaminated materials.

Paper and paperboard offer renewable and recyclable alternatives to conventional plastic packaging. However, their hydrophilic and porous structures provide limited resistance to water, water vapor, oils, and greases. Consequently, paper-based food packaging is frequently combined with polymeric laminates or functional surface treatments to provide the required barrier properties. Among these treatments, per- and polyfluoroalkyl substances (PFAS) have historically been used to impart strong water and grease resistance because of their low surface energy and high chemical stability [4–7]. However, the environmental persistence and potential health concerns associated with some PFAS have led to increasing regulatory restrictions and accelerated the search for fluorine-free alternatives [8–12].

Nanocellulose has emerged as a promising bio-based material for developing such alternatives because it is renewable, biodegradable, lightweight, and capable of forming dense networks with excellent oxygen-barrier properties under dry conditions. Nanocellulose comprises several cellulose-based fibrillar and crystalline materials, including cellulose nanofibrils (CNF), cellulose microfibrils (CMF), and cellulose nanocrystals (CNC). CNF and CMF are particularly relevant to paper-based coatings because they can form continuous fibrillar networks and can be incorporated into aqueous coating formulations [13–16]. However, the hydrophilic nature of nanocellulose limits its performance in applications requiring simultaneous oxygen, moisture, and grease resistance. This has motivated the development of hybrid coatings that combine nanocellulose with hydrophobic and complementary barrier materials.

A promising strategy is therefore to combine nanocellulose with hydrophobic and complementary bio-based components. Natural waxes, including carnauba, candelilla, and beeswax, can provide water and grease resistance, whereas biopolymers such as chitosan, starch, alginate, proteins, and polylactic acid (PLA) contribute to film formation, adhesion, mechanical integrity, and other functional properties [17–19]. Combining these components with nanocellulose can produce multilayer or hybrid coatings in which different materials provide complementary barrier functions. These systems could enhance moisture and grease resistance while retaining the oxygen-barrier and reinforcing properties of nanocellulose.

However, achieving a multifunctional PFAS-free coating involves more than combining hydrophilic and hydrophobic materials. Differences in polarity and structure can affect interfacial compatibility, dispersion, coating uniformity, and mechanical integrity, while wax crystallization, migration, and formulation stability can influence long-term performance. In addition, the environmental and regulatory advantages of bio-based coatings cannot be assumed solely from the origin of their components; recyclability, compostability, food-contact safety, migration, and life-cycle impacts depend on the complete formulation and its intended application. These considerations become particularly important when moving from laboratory-scale formulations to industrial coating processes.

Replacing PFAS coatings in food packaging does not necessarily imply a direct, one-to-one substitution with a single alternative material. The feasibility of PFAS-free solutions

depends on the functional requirements of the specific application, including water, water-vapor, oil, and grease resistance, as well as mechanical integrity and processing requirements. Nanocellulose–natural wax coatings should therefore be considered application-specific alternatives that combine different barrier mechanisms, rather than universal replacements for PFAS-based treatments. Depending on the required performance, these systems may enable complete PFAS replacement in selected applications, partial reduction in PFAS use, or functional substitution for specific barrier properties.

Table 1 presents the main material categories and their applications in food packaging.

Table 1. Main material categories used for barrier and functional food-packaging applications.

Material Category	Representative Materials *	Main Function	Sustainability Considerations
Non-polymeric PFAS	PFOA, PFOS, FTOHs	Water and grease resistance	Highly persistent; some compounds are bioaccumulative and environmentally mobile
Fluoropolymers	PTFE, FEP, PVDF	High-performance barrier and protective coatings	Highly resistant to degradation; end-of-life management can be challenging
Conventional fossil-based polymers	PE, PET, EVOH	Moisture and oxygen barrier layers; structural and sealing functions	Generally non-biodegradable; recyclability depends on multilayer composition
Bio-based polymers and cellulosic materials	CNF/CMF, CNC, PLA, PHAs	Oxygen and moisture barrier; structural reinforcement; coating matrix	Renewable feedstocks; biodegradability and compostability depend on material and processing conditions
Natural waxes	Carnauba wax, candelilla wax, beeswax	Water and grease resistance	Renewable or bio-derived; biodegradability is generally favorable, depending on the formulation
Hybrid bio-based coatings	Nanocellulose + natural waxes and/or biopolymers	Multifunctional moisture, grease, and oxygen barrier functions	Recyclability and compostability depend on coating composition, loading, and substrate

* PFOA: Perfluorooctanoic acid, PFOS: perfluoro octane sulfonate, FTOHs: fluorotelomer alcohols, PTFE: polytetrafluoroethylene, FEP: fluorinated ethylene propylene, PVDF: polyvinylidene fluoride, EVOH: ethylene vinyl alcohol, PLA: polylactic acid, PHAs: polyhydroxyalkanoates.

This review focuses on processing and formulation strategies for nanocellulose–natural wax hybrid coatings as PFAS-free barrier solutions for paper-based food packaging. Its main contribution is to integrate PFAS replacement strategies with the design of multifunctional coatings based on nanocellulose, natural waxes, and complementary barrier materials, emphasizing the relationships between material selection, formulation, processing, interfacial structure, and final performance. The preparation of cellulose nanofibrils (CNF), microfibrillated cellulose (CMF), and cellulose nanocrystals (CNC), wax processing and dispersion, formulation parameters, coating methods, and drying conditions are examined in relation to coating morphology, interfacial compatibility, and oxygen, moisture, and grease barrier properties. Attention is given to how the distribution of hydrophilic and hydrophobic phases, wax crystallization, coating continuity, and substrate interactions influence barrier performance and mechanical integrity. By integrating these aspects within a processing–structure–performance framework, this review identifies the main opportuni-

ties, limitations, and scale-up challenges associated with developing application-specific PFAS-free coatings compatible with paper-based food-packaging technologies.

2. Methods

A structured literature search was conducted to identify and critically analyze studies relevant to PFAS replacement strategies and the processing and formulation of nanocellulose–natural wax hybrid coatings for paper-based food packaging. The search strategy and article selection criteria were designed to support a critical synthesis of the available literature.

2.1. Research Questions and Data Collection

The preparation of this critical review was guided by most of the applicable PRISMA 2020 [20] reporting items, adapted to the scope and methodological approach of the review. Items that did not apply to the nature of this critical review were not included [20]. The PRISMA checklist was added to the Supplementary Information. The objectives of the review were first refined and formulated as research questions (RQs):

RQ1. What are PFAS, where are they used in food packaging, and what are the main concerns associated with their use?

RQ2. What regulations and regulatory restrictions currently apply to PFAS in food-contact packaging?

RQ3. What processing and formulation strategies have been investigated for natural wax-based PFAS-free barrier coatings?

RQ4. How do nanocellulose preparation, wax processing, formulation, coating, and drying affect the structure and barrier performance of PFAS-free hybrid coatings?

RQ5. What are the main technological, economic, and scale-up challenges associated with implementing nanocellulose–natural wax coatings in paper-based food packaging?

Figure 1 shows the criteria for article inclusion or exclusion.

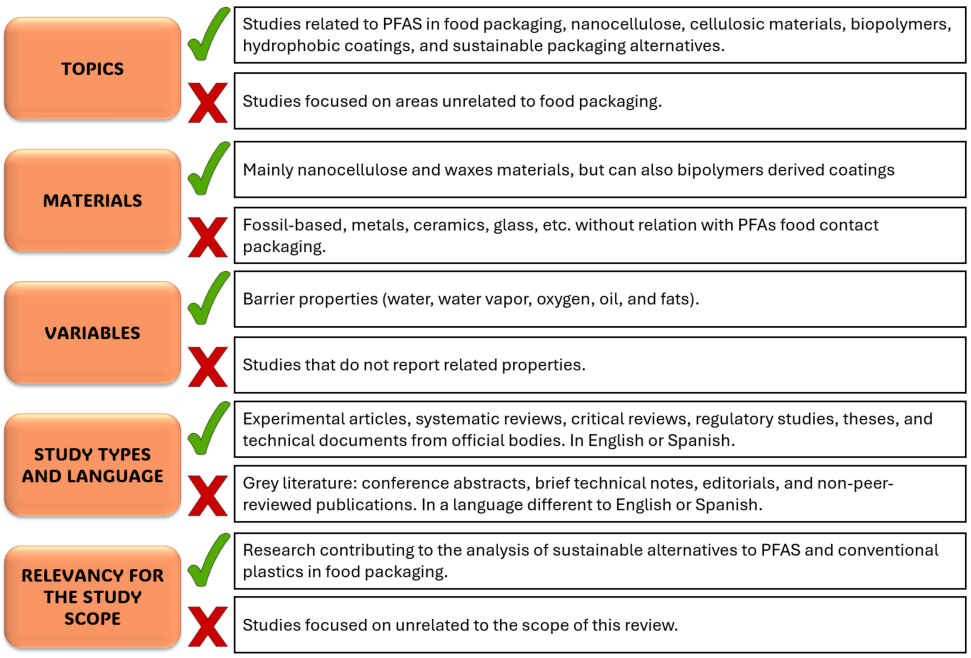


Figure 1. Criteria for inclusion or exclusion of articles considered for this review.

The literature search was conducted using ScienceDirect and Google Scholar. The main search terms included PFAS, fluorinated coatings, PFAS regulations, natural wax coatings,

biopolymer coatings, nanocellulose hybrid coatings, PFAS-free market, and grease barrier coatings. Boolean operators AND and OR were used to combine search terms and refine the results.

2.2. Methodological Boundaries

According to PRISMA 2020 reporting guidelines (Supplementary Information item 23c), several methodological boundaries were applied to the search strategy and screening process. The database and search scope were restricted to Google Scholar and ScienceDirect. Consequently, some relevant studies indexed exclusively in these omitted databases may not have been used in the initial search. The selection process was restricted to peer-reviewed original research articles and scientific reviews published in English. As a result, technical reports, regional conference proceedings, dissertations, or relevant research published in other languages were excluded from the formal screening pipeline, which may introduce a minor language bias. Gray literature was excluded from the quantitative evaluation. All searches were conducted by the first author without the use of automated tools. Including the additional literature suggested by the reviewers during the peer-review process, the total number of articles used was n = 103.

Titles and abstracts were screened to identify potentially relevant studies, followed by an assessment of the full texts of the selected articles against the predefined eligibility criteria. The selection process was conducted by one reviewer, who determined whether each study met the inclusion criteria. No automated tools were used in the selection process.

The main outcomes sought from the studies focused on process performance and product characteristics. Depending on the technology investigated, these included extraction and conversion yields, product recovery efficiencies, operating conditions, and the physicochemical and functional properties of the resulting products or materials. When reported, energy consumption, techno-economic indicators, and environmental performance metrics were also collected. All reported results relevant to the predefined outcome categories were considered, including results obtained under different experimental conditions. When multiple measurements or operating conditions were reported, the data were extracted and analyzed according to their relevance to the objectives of the review.

In addition to the main outcomes, the following study characteristics were extracted when available: publication year, biomass type and origin, feedstock composition, technology or process investigated, experimental conditions, operating scale, equipment configuration, and characteristics of the main products and coproducts. Techno-economic and environmental information was also recorded when relevant to the scope of the review. Missing or unclear information was treated as unreported unless it could be unambiguously established from other information provided in the article. No values were estimated when the available information was insufficient to support a reliable determination.

2.3. Use of Generative AI Tools

ChatGPT (OpenAI, GPT-6) was used to assist in the visual refinement and improvement of Figure 3. Grammarly (Grammarly Inc., San Francisco, CA, USA.) was used to support English language editing, including grammar, spelling, and sentence structure corrections. Neither tool was used to generate scientific data, perform data analysis, interpret results, or develop the scientific content of the manuscript. All suggested modifications were reviewed and approved by the authors, who retain full responsibility for the accuracy and integrity of the manuscript.

3. PFA Substances

3.1. Definition, Classification, and Structure

Per- and polyfluoroalkyl substances (PFAS) comprise a large and diverse family of fluorinated organic compounds characterized by carbon chains containing one or more highly fluorinated groups. The original structural definition proposed by Buck et al. (2011) [21] included perfluoroalkyl and polyfluoroalkyl substances containing at least one fully fluorinated carbon atom [21]. Subsequently, the Organization for Economic Co-operation and Development (OECD) broadened the definition to include compounds containing at least one fully fluorinated methyl (CF_3) or methylene (CF_2) [22]. In the United States, the Environmental Protection Agency (EPA) has used definitions based on specific fluorinated structural features, with the scope evolving according to regulatory objectives and scientific understanding [23]. These differing definitions reflect the scientific, regulatory, and operational contexts in which PFAS are addressed [21–23].

Figure 2 presents PFAS classification into structural and functional groups. This classification provides a useful framework for discussing differences in mobility, environmental behavior, and degradation pathways [21,22].

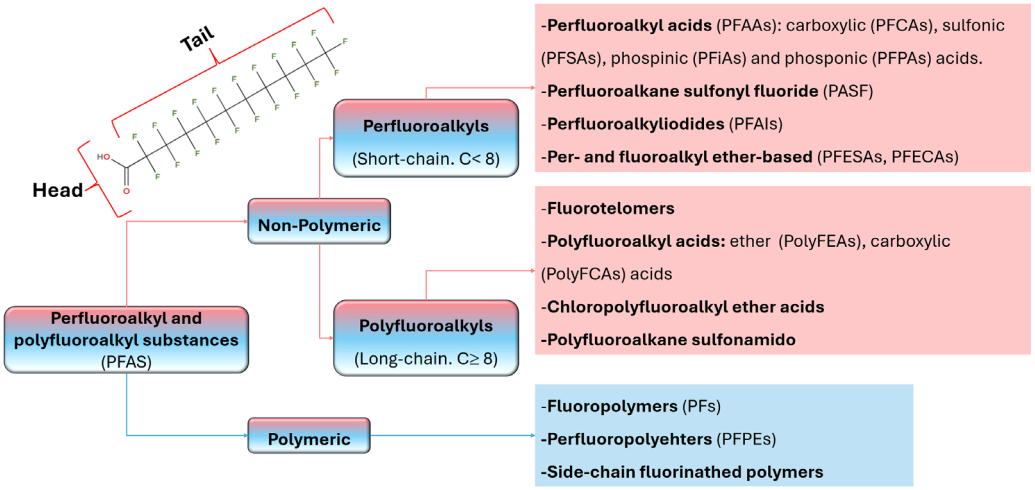


Figure 2. PFAS classification into structural and functional groups.

PFAS are broadly classified into non-polymeric and polymeric compounds, a distinction useful for understanding their mobility, environmental behavior, and degradation pathways [21–23]. Non-polymeric PFAS include perfluoroalkyl acids (PFAAs), such as perfluorocarboxylic acids (PFCAs) and perfluoroalkane sulfonic acids (PFSAs), as well as precursor compounds such as fluorotelomer alcohols (FTOHs) and sulfonamides. Some of these precursors can transform into persistent terminal PFAAs [23]. Polymeric PFAS include fluoropolymers, such as polytetrafluoroethylene (PTFE), which are widely used because of their high chemical and thermal stability and low surface energy [21]. Within non-polymeric PFAS, perfluoroalkyl and polyfluoroalkyl substances can also be distinguished according to the degree of fluorination. Perfluoroalkyl compounds contain fully fluorinated carbon chains, whereas polyfluoroalkyl compounds contain both fluorinated and non-fluorinated carbon atoms.

3.2. Uses and Migration of PFAS

PFAS have been widely used as surface treatments and coatings for paper- and paperboard-based food packaging because they provide effective resistance to water, grease, and heat by lowering the surface energy of cellulosic substrates [6,7,10,23–25]. Typical

applications include fast-food wrappers, grease-proof papers, microwave popcorn bags, beverage cups, and other packaging intended for fatty or high-temperature foods [7,23].

Despite their effective water- and grease-repellent properties, PFAS and their precursors can migrate from food-contact materials into food through processes involving diffusion, desorption, and partitioning. Migration levels depend on several factors, including molecular structure, molecular weight, contact time, temperature, and food composition [10,21,23,26]. Contact with hot or fatty foods can promote greater transfer of certain PFAS from packaging materials [7,10,23]. Some fluorinated precursors can transform into persistent perfluoroalkyl acids (PFAAs), complicating exposure assessment and potentially leading to an underestimation of total PFAS exposure when precursor compounds are not considered [10]. The combination of migration, environmental persistence, and potential health concerns has contributed to increasingly stringent regulatory measures [12,27,28].

3.3. Health, Environmental Concerns, and Regulatory Trends

Exposure to certain PFAS has raised concerns because of their persistence, bioaccumulation potential, and toxicity. Dietary exposure can occur through the migration of PFAS from food-contact materials into food, with epidemiological and toxicological studies linking exposure to certain PFAS to adverse effects on the liver, endocrine, and immune systems, as well as to an increased risk of certain cancers [24,29,30].

Owing to the high chemical stability of many PFAS, these substances have been detected in water, soil, sediments, wildlife, and remote environments, contributing to their widespread distribution and, for some compounds, accumulation in biota and food webs [31,32]. Their presence in waste streams and potential release during production, use, recycling, and disposal further complicate waste management and pose challenges to the development of circular packaging systems [10].

PFAS regulations have progressively tightened since the early 2000s, initially focusing on specific compounds such as (perfluoro octane sulfonates) PFOS and (perfluorooctanoic acid) PFOA and increasingly moving toward broader controls on the PFAS class. In Europe, PFOS was restricted to essential uses in 2006 and listed under the Stockholm Convention in 2009. In the United States, regulatory measures have included the US EPA's 2016 lifetime health advisory levels for PFOS and PFOA, followed by increasingly stringent measures addressing PFAS in food-contact applications. Recently, restrictions specifically targeting PFAS in food-contact packaging have been adopted in both the USA and the EU, reinforcing the need for effective fluorine-free barrier solutions for paper and paperboard packaging [33–35].

4. Processing Requirements for PFAS-Free Barrier Coatings

4.1. Barrier Requirements and Transport Mechanisms

Barrier performance is critical in food packaging, particularly for products containing water or fats. Depending on the application, coatings must limit the transport of water vapor and gases and resist the penetration and spreading of oils and greases while maintaining food quality and safety. Barrier performance therefore depends on the interplay among coating composition, microstructure, surface properties, and environmental conditions.

Coating permeability is governed by its chemical composition and microstructure, as well as by temperature and relative humidity. In nanocellulose-based coatings, the dense fibrillar network formed through extensive interfibril hydrogen bonding creates tortuous pathways that restrict oxygen transport, resulting in very low oxygen transmission rates (OTR) under dry conditions [36–38]. However, moisture can substantially reduce this barrier performance by disrupting hydrogen bonding, promoting swelling and increasing molecular mobility, thereby facilitating gas transport [39–41]. This moisture sensitivity has motivated the development of hybrid systems in which nanocellulose is combined with

hydrophobic components, such as natural waxes or hydrophobic biopolymers, to improve moisture resistance under conditions relevant to food packaging [42–45].

The hydrophilic nature of cellulose also limits resistance to water-vapor transport [46,47]. Incorporating natural waxes can reduce water sorption and vapor transmission by introducing a hydrophobic phase within or over the cellulosic coating [48,49]. The effectiveness of this approach depends on wax content, dispersion, and coating continuity, which determines whether the hydrophobic phase provides sufficiently uniform coverage without compromising the coating integrity [43].

Surface chemistry and coating microstructure influence grease resistance. Low surface energy can reduce wetting and spreading of oils, while a continuous and sufficiently dense coating can hinder grease penetration into the porous paper substrate. Several bio-based systems, including CNF–wax formulations [17], CNF–lignin coatings [50], and PLA–CNF multilayer structures [51], have demonstrated high grease resistance in application-specific tests.

However, direct comparison with fluorinated treatments is complicated by differences in coating composition, coat weight, substrate characteristics, and test methods. Consequently, grease resistance should be evaluated together with moisture and oxygen barrier performance under standardized and application-relevant conditions. Although nanocellulose–wax coatings offer potential PFAS-free solutions for selected applications, their suitability must be established according to the specific barrier requirements, because improvements in one property do not necessarily translate into equivalent performance in others.

4.2. Surface Chemistry, Roughness, and Wetting

Wettability is governed by physical and chemical processes involving intermolecular forces, surface energies, and fluid–solid interactions. Surface chemistry and topography together govern the wettability of a coated surface. Polar surface groups, such as hydroxyl groups, generally promote hydrophilic behavior, whereas non-polar groups favor hydrophobicity. Surface roughness can modify the apparent wettability by changing the actual solid–liquid contact area and, in some cases, the fraction of liquid–air interface. In the Wenzel regime, where the liquid completely follows the surface topography, roughness amplifies the intrinsic surface wettability. Thus, it can increase spreading on hydrophilic surfaces or enhance repellency on hydrophobic ones [52]. In the Cassie–Baxter regime, air is trapped within the surface irregularities, reducing the effective solid–liquid contact area and potentially increasing water repellency [53]. Transitions between these regimes may occur depending on surface chemistry, roughness, liquid properties, and coating morphology. However, these models describe liquid–surface interactions and do not, by themselves, establish coating barrier performance.

Untreated paper is intrinsically hydrophilic and highly porous, and its irregular surface facilitates liquid spreading and penetration into the substrate. Consequently, reducing surface roughness and sealing surface pores can improve the effectiveness of barrier treatments. Nanocellulose coatings can contribute to this effect because their small dimensions allow them to fill surface irregularities and form relatively continuous films. The resulting smoother and more homogeneous surface can reduce initial water penetration and increase the apparent water contact angle, while water absorption is commonly evaluated by methods such as the Cobb test [25,40]. Combining CNF with lignin can further improve coating uniformity and barrier performance, including under moderately humid conditions [50]. Lignin can also modify surface energy, although its effect depends on its type and chemical structure. Its incorporation into CNF has been associated with reduced surface free energy, increased water contact angle, and improved grease resistance of coated paper [54]. Similarly, PLA/CNF-based multilayer systems have exhibited surface properties comparable to

those of PE and PET, while maintaining low oxygen transmission rates [51,55]. Nevertheless, barrier performance also depends on coating continuity, thickness, porosity, roughness, interfacial adhesion, and environmental conditions.

Natural waxes can substantially modify surface topography during drying because they crystallize or recrystallize as the coating solidifies [56]. The resulting morphology depends on the wax type, dispersion characteristics, application method, and drying conditions. Carnauba wax, for example, can form lamellar or platelet-like structures, whereas beeswax may generate more particulate or nodular morphologies [57–59]. Wax distribution and surface morphology can influence water repellency; however, their effects on barrier performance depend on the continuity of the wax-containing layer and the presence of defects, rather than on surface roughness alone. Coarse or heterogeneous crystallization may generate discontinuities or preferential pathways for liquid penetration if coating integrity is compromised [59].

The distribution of wax within the coating is therefore important for obtaining a continuous and reproducible barrier layer. Aqueous wax dispersions stabilized with anionic CNF have been reported to improve wax distribution and modify coating morphology, potentially reducing heterogeneous aggregation and promoting more uniform coatings [58]. These effects highlight the role of nanocellulose not only in determining surface characteristics but also in controlling the distribution of wax within the hybrid formulation.

4.3. Formulation and Interfacial Compatibility

The surface chemistry of nanocellulose influences its interactions with water and other formulation components. The abundant surface hydroxyl groups contribute to its hydrophilic character and facilitate interactions with polar liquids [59,60]. Surface properties can be modified through chemical functionalization, such as acetylation or esterification, or by incorporating hydrophobic components [61–63]. The resulting changes in wettability depend on the nature and degree of surface modification, the distribution of the different components, and the morphology of the coating.

In nanocellulose–wax hybrid formulations, interfacial compatibility depends on the surface chemistry of the components, the stability of the dispersed wax phase, and the structural organization developed during coating formation. The hydrophilic character of cellulose nanofibrils (CNF) contrasts with the predominantly hydrophobic nature of natural waxes, making wax dispersion and stabilization important formulation challenges. Depending on the composition and preparation method, inadequate dispersion may lead to aggregation or phase separation, potentially compromising coating uniformity and continuity.

The formation of nanocellulose–wax hybrid coatings involves interconnected mechanisms, from the stabilization of wax droplets in the aqueous formulation to the structural changes occurring during drying. These mechanisms influence component distribution, coating microstructure, and the resulting surface and barrier properties. Figure 3 schematically summarizes the main stages and mechanisms involved in hybrid coating formation.

Studies on nanocellulose-stabilized Pickering emulsions have shown that nanocellulose can adsorb at oil–water interfaces and contribute to emulsion stability, although its effectiveness depends on the characteristics of the nanocellulose and the formulation conditions [64]. More specifically, recent work on beeswax-in-water nanoemulgels stabilized with TEMPO-oxidized cellulose nanofibers (TOCNFs) demonstrated the formation of a dense interfacial nanocellulose shell and a continuous viscoelastic network that immobilized the wax particles and improved resistance to phase separation. The authors also showed that confinement within the nanocellulose shell modified the crystallization behavior and crystalline organization of the wax [56]. These findings provide mechanistic

evidence that nanocellulose can influence both the stability of a dispersed wax phase and its structural organization.

However, these results were obtained in nanoemulgels and should not be directly extrapolated to dried coatings. During coating formation, the redistribution of components, the continuity of the deposited layer, and adhesion to the substrate may also affect the final structure and barrier performance.

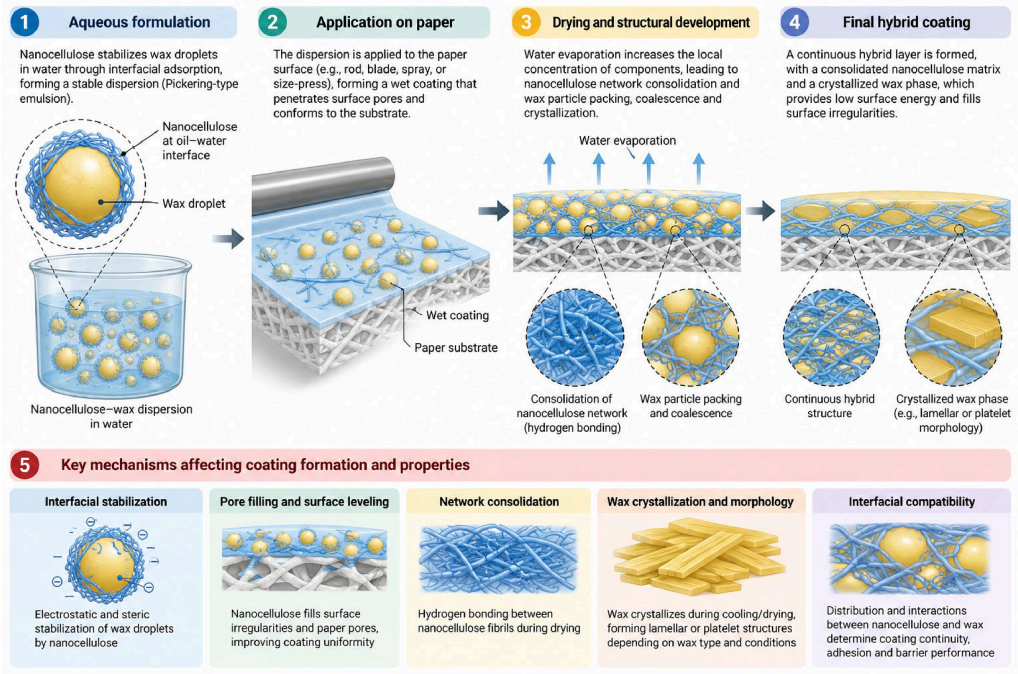


Figure 3. Schematic illustration of the main mechanisms involved in the formation of nanocellulose–natural wax hybrid coatings on paper (prepared by the authors and visually refined with the assistance of ChatGPT).

The extent to which aggregation, phase separation, or changes in wax crystallization occur therefore needs to be considered in relation to the specific formulation and processing conditions.

4.4. Coating Processes and Structure Development

Coating performance on paper depends on formulation composition, coat weight, drying conditions, interfacial compatibility, and application technique. These variables influence coating thickness, surface coverage, penetration into the porous substrate, and the formation of a continuous layer. In nanocellulose-based coatings, suspension rheology and water retention are particularly important because they affect flow, leveling, and the ability of the coating to cover surface irregularities. The uniformity and integrity of the resulting layer therefore depend on the combined effects of formulation and processing conditions, which also influence coating formation and adhesion to the substrate [65].

Table 2 summarizes the main coating methods reported for bio-based barrier coatings, with emphasis on their applicability to paper-based packaging.

Rod coating provides good control over coat weight and thickness and is therefore widely used for laboratory and pilot-scale evaluation of nanocellulose- and wax-based formulations [58,66,67]. Blade coating is more relevant to industrial processing and can provide relatively high coat weights and continuous coverage. However, coating rheology, blade pressure, and substrate characteristics must be controlled to avoid excessive penetration, streaking, or surface defects [68,69]. Spray coating enables non-contact deposition and

is useful for thin coatings and irregular substrates, but achieving uniform coverage over large areas requires careful control of droplet formation, formulation rheology, substrate wettability, spray parameters, and drying conditions [67,70–72].

Table 2. Comparison of application methods for bio-based barrier coatings.

Method	Typical Scale/Applicability	Main Applications	Ref.
Rod coating	Laboratory and pilot scale	Nanocellulose- and wax-based coatings; biopolymers	[58,66,67]
Blade coating	Laboratory, pilot, and industrial scale	Surface and barrier coatings	[68,69]
Spray coating	Laboratory, pilot, and selected industrial applications	Thin surface and barrier coating	[67,70–72]
Size/film-press	Pilot and industrial papermaking	Surface sizing and barrier treatments	[73]
Curtain/slot die coating	Pilot and industrial scale	Continuous high-coverage barrier coatings	[74,75]
Extrusion/co-extrusion	Industrial and commercial scale	Thermoplastic coatings and multilayer structures	[43,76,77]
Dip coating	Mainly laboratory scale	Formulation development and substrate studies	[72]

Size-press and film-press technologies are particularly relevant to scale-up because they can be integrated into existing papermaking operations. They are routinely used for aqueous surface treatments, although the achievable coat weight and penetration depend strongly on formulation solids, viscosity, substrate properties, and nip conditions [73]. Continuous methods such as curtain and slot-die coating can provide more uniform and relatively high coat weights and are therefore attractive for barrier applications requiring complete surface coverage [74,75]. These methods are increasingly relevant when translating laboratory formulations into continuous coating processes.

For thermoplastic systems, particularly PLA-based multilayers, extrusion and co-extrusion enable continuous coating or lamination and are already established at industrial scale [43,76,77]. Their solvent-free operation and ability to produce continuous layers are advantageous, but processing temperatures must be controlled to prevent thermal degradation of cellulose and other temperature-sensitive components. Dip coating remains primarily a laboratory technique because coating thickness and uniformity are more difficult to control, although it is useful for rapidly evaluating formulation–substrate interactions and coating formation [72].

The choice of coating method must be considered together with formulation rheology, solids content, target coat weight, drying requirements, substrate characteristics, and line speed. Therefore, for nanocellulose–wax hybrid systems, the most relevant scale-up challenge is not simply transferring a laboratory formulation to a larger coater but adapting its rheological and drying behavior to a continuous process while maintaining uniform coverage and barrier performance. This formulation–process interaction is a key consideration for industrial implementation of PFAS-free coatings.

4.5. Drying and Coating Formation

Drying is a critical processing stage in the production of nanocellulose-based barrier coatings because it determines the development of the coating structure, surface morphol-

ogy, adhesion, and final barrier performance. Drying temperature, relative humidity, air circulation, and coating thickness jointly influence water evaporation and the rate at which the coating consolidates. Rapid evaporation can generate steep moisture and concentration gradients through the coating, potentially producing nonuniform shrinkage, internal stresses, surface defects, or cracks [78]. Conversely, slower drying may facilitate more uniform consolidation but increase processing time and the risk of manufacturing bottlenecks. The appropriate drying conditions therefore depend on formulation composition, coat weight, substrate characteristics, and the requirements of the intended coating process.

The effects of drying conditions are particularly relevant to nanocellulose coatings, whose interconnected fibrillar networks undergo consolidation as water is removed. Andrade et al. [78] demonstrated that drying method, temperature, and applied pressure significantly affect the surface roughness, density, porosity, and wettability of cellulose nanofibril films. These results highlight the importance of controlling drying conditions to obtain uniform surfaces and reproducible material properties. However, the behavior of coatings deposited on porous paper substrates may differ from that of free-standing nanocellulose films because water can penetrate the substrate and the coating can interact with the underlying fiber network.

In nanocellulose–natural wax hybrid coatings, drying also involves the evolution of the dispersed hydrophobic phase. For aqueous wax emulsions, water evaporation increases the local concentration of wax particles and other formulation components, potentially affecting particle packing, coalescence, distribution, and the continuity of the hydrophobic layer. Where wax crystallization occurs during cooling or solvent removal, its kinetics and resulting crystal morphology may influence surface topography, coating cohesion, and barrier performance [79]. However, the extent of coalescence, redistribution, or crystallization during drying depends on wax composition, emulsion stability, particle characteristics, and thermal history; these mechanisms should therefore be established for the specific formulation rather than assumed to occur universally.

Coating adhesion and integrity are additional considerations. The substrate's porosity, surface chemistry, and roughness affect coating penetration and interfacial contact, while drying-induced shrinkage can generate stresses that compromise adhesion or promote cracking. Saremi et al. [80] demonstrated that nanocellulose coating adhesion and stability depend on substrate characteristics and on strategies used to strengthen the coating structure. Although their study focused on polymer films and textiles, it illustrates the importance of interfacial compatibility and coating morphology. For paper-based packaging, drying conditions must therefore be optimized to balance production rate with uniform film formation, adequate adhesion, and the preservation of the desired barrier properties.

The thermal behavior of nanocellulose–wax hybrid coatings depends on the properties of the individual components, their relative proportions, and the structure developed during coating formation. The melting range of a natural wax reflects its transition from a solid to a softened or liquid state and should be distinguished from its thermal stability, which refers to its resistance to chemical decomposition upon heating. Depending on its composition, wax may soften or melt well below the temperature at which significant thermal degradation occurs. Studies on wax-coated paper have shown that wax type and its thermal properties, crystallinity, and crystal morphology are relevant characteristics for understanding the performance of the resulting coatings [79]. The nanocellulose matrix also undergoes thermally induced changes, including moisture loss and, at higher temperatures, chemical degradation, while studies of cellulosic materials and nanocellulose-containing polymer coatings demonstrate that material morphology, surface modification, and matrix interactions can influence thermal and thermomechanical behavior [81]. In hybrid coatings, increasing the wax fraction may modify the thermal transitions and the relative contributions

of the wax and cellulose-based phases, but the resulting behavior cannot be predicted from the properties of either component alone. Therefore, the incorporation of natural wax should not be assumed to improve or reduce the overall thermal stability of the coating without experimental evidence. Differential scanning calorimetry (DSC) can be used to characterize melting and crystallization transitions, whereas thermogravimetric analysis (TGA) provides information on mass loss and thermal degradation. These complementary measurements can help distinguish changes in phase transitions from changes in the resistance of the hybrid coating to thermal decomposition [82].

5. Nanocellulose: Processing, Structure, and Barrier Mechanisms

Nanocellulose comprises several cellulose-based fibrillar and crystalline materials, including cellulose nanofibrils (CNF), cellulose microfibrils (CMF), and cellulose nanocrystals (CNC), which differ in morphology, crystallinity, and preparation route. CNF and CMF are generally produced by mechanically fibrillating purified or pretreated cellulose fibers using processes such as refining, grinding, or homogenization. Chemical or enzymatic pretreatments, including TEMPO-mediated oxidation, can facilitate fibrillation by weakening interfibrillar interactions and reducing the energy required for mechanical processing. In contrast, CNC are typically produced by acid hydrolysis, which preferentially removes the less ordered regions of cellulose and yields shorter, rigid, highly crystalline particles [25,40].

Various factors contribute to the barrier performance of nanocellulose. Under dry conditions, CNF can form dense films through extensive hydrogen bonding, reducing oxygen transmission by restricting molecular diffusion [36–38,67]. In addition, the entangled fibrillar network increases diffusion-path tortuosity, making transport pathways longer and more complex and thereby reducing effective permeability. This effect depends on fibril aspect ratio, network density, crystallinity, coating thickness, and surface uniformity [36–38,67]. The abundance of surface hydroxyl groups also provides opportunities for chemical functionalization and incorporation of functional agents, enabling the surface chemistry and barrier properties of nanocellulose coatings to be tailored for specific applications [83,84].

The processing route therefore strongly influences the dimensions, surface chemistry, crystallinity, rheological behavior, and subsequent coating performance of the resulting cellulose material. CNCs are rigid, highly crystalline particles with high specific surface area and reinforcing potential. CNFs are flexible, high-aspect-ratio fibrils that form highly entangled networks with strong film-forming ability. CMFs are generally more heterogeneous and less fibrillated than CNFs, but their lower degree of fibrillation can facilitate processing at higher solids contents and integration into conventional coating operations. For paper-based food packaging, CNFs and CMFs are particularly relevant because of their film-forming and coating properties, with CNFs generally providing more effective oxygen-barrier structures and CMFs offering potential processing advantages [13–16].

The barrier performance of nanocellulose therefore depends closely on its morphology and the structure of the resulting coating. High fibrillation, sufficient network density, and uniform substrate coverage favor continuous pathways that restrict gas diffusion. However, these characteristics may also increase suspension viscosity and complicate coating at high solids content. Consequently, optimizing nanocellulose morphology and coating structure requires balancing barrier performance with processability, coat weight, and drying requirements.

The processing conditions used to produce nanocellulose directly influence the resulting material characteristics and, consequently, its behavior during coating. The degree of fibrillation, fibril dimensions, and their distribution depend on the intensity and sequence of mechanical and chemical treatments, which also determine energy demand and surface characteristics. These structural features strongly affect suspension rheology,

particularly viscosity, shear-thinning behavior, and network formation, thereby influencing solids handling, mixing, pumping, and deposition.

The rheological behavior of nanocellulose suspensions depends on particle morphology, concentration, surface chemistry, and interactions between fibrils or crystals. These factors influence apparent viscosity, shear-thinning behavior, viscoelasticity, and the development of structured networks. Hubbe et al. [85] emphasized that rheology is also affected by flow history and changes in particle interactions, with direct implications for suspension preparation, pumping, mixing, storage, metering, and application. More recently, Xu et al. [86] further demonstrated that rheological behavior depends on both concentration and time, with cellulose nanofibril (CNF) suspensions exhibiting stronger thixotropy and higher yield stress than cellulose nanocrystal (CNC) suspensions. These differences were attributed to the higher aspect ratio of CNF and the formation of a more developed structural network. The importance of evaluating rheology under processing-relevant conditions was also demonstrated by Kumar et al. [87], who investigated the coatability of different CNF suspensions and showed that their properties depend on the production method and can substantially affect processability. Together, these studies indicate that no single rheological parameter is sufficient to predict coating performance; rather, the interplay between flow behavior, network structure, and water retention must be considered.

The implications of these properties depend on the coating method. Apparent viscosity over a wide range of shear rates is particularly relevant to pumping and metering, as well as to the formation of a uniform wet layer. Shear-thinning behavior can facilitate flow through coating equipment by reducing the apparent viscosity at high shear rates, whereas yield stress may hinder pumping and deposition when it is too high. For continuous slot-die coating, Kumar et al. [87] successfully applied different CNF suspensions in a roll-to-roll process and showed that modifying the formulations with carboxymethyl cellulose (CMC) improved coatability by affecting both low- and high-shear rheology and water retention. Koppolu and Toivakka [65] likewise reported that CMC reduced yield stress and slowed structural recovery after high shear, contributing to improved coating quality in continuous slot-die processing. Nechyporchuk et al. [88] also distinguish between mechanically fibrillated suspensions with highly flocculated structures and chemically pretreated suspensions with greater colloidal stability. This is useful for explaining why processing history and nanocellulose morphology influence rheological behavior.

These findings illustrate that formulation design must account not only for the viscosity during deposition but also for the suspension's structural recovery and dewatering behavior after application. For blade and rod coating, these properties are relevant to metering, leveling, and the formation of a continuous layer; in spray coating, the flow behavior must be compatible with atomization and droplet deposition. Oscillatory measurements, including storage and loss moduli and their frequency dependence, can provide complementary information on the strength and dynamics of the suspension network, although their relevance should be assessed in relation to the specific formulation and coating process. Temperature may also influence flow behavior and should be controlled or evaluated when processing conditions involve appreciable temperature changes. Because the cited studies primarily investigate aqueous nanocellulose suspensions or nanocellulose-based coating systems rather than the specific nanocellulose–natural wax hybrid formulations discussed here, the rheological behavior and processability of these hybrids should be determined experimentally rather than inferred directly from the properties of the individual components Hubbe [85], Xu [86], Kumar [87], Koppolu and Toivakka [65]. Thus, control of the fibrillation process is essential not only for tailoring nanocellulose morphology but also for achieving the rheological and processing characteristics required for uniform coating for-

mation and, ultimately, the desired barrier performance. Temperature should be controlled or evaluated when relevant to the formulation and operating conditions.

6. Processing of Natural Waxes and Hybrid Formulations

6.1. Natural Waxes: Composition and Processing

Natural waxes are promising hydrophobic components for cellulose- and nanocellulose-based food-packaging coatings because of their low polarity, low surface energy, and ability to form continuous hydrophobic phases. Their composition and properties vary substantially according to botanical or animal origin, geographic source, extraction and purification processes, and subsequent formulation and processing conditions. Consequently, the performance of a wax coating cannot be predicted solely from its generic classification as a "natural wax", which is a complex mixture containing different proportions of long-chain hydrocarbons, fatty acids, esters, fatty alcohols, and other minor components. The relative abundance of these constituents affects melting behavior, crystallinity, hardness, flexibility, surface energy, adhesion, and moisture-barrier performance. Waxes with higher proportions of long-chain hydrocarbons and esters generally exhibit high hydrophobicity and crystallinity, whereas the presence of fatty alcohols, free fatty acids, and other polar components can influence flexibility and interactions with the cellulosic substrate [43,57,72]. These compositional differences are particularly important when comparing waxes from different sources or suppliers.

Candelilla, carnauba, and beeswax are among the most frequently investigated natural waxes for food-packaging applications [18,43,49,58,60,89–91]. Candelilla wax is characterized by a relatively high content of long-chain hydrocarbons and esters and a comparatively high melting range, contributing to its hardness, crystallinity, and moisture-barrier performance [89,90]. Carnauba wax is also highly crystalline and has a high melting temperature [18,43,91], whereas beeswax contains a more complex mixture of esters, hydrocarbons, fatty acids, and alcohols and generally exhibits a lower melting range [49,58,89]. These differences influence coating formation, crystallization morphology, mechanical behavior, and resistance to water and oils.

The origin and composition of waxes should therefore be considered together with their processing history. Extraction, refining, fractionation, emulsification, particle size, solid content, and drying conditions can modify wax dispersion, crystallization, surface morphology, and adhesion to the substrate [92]. Recent studies comparing different bio-based waxes have demonstrated that the source of the wax can significantly affect the water and oil resistance of treated packaging papers, highlighting the importance of characterizing individual waxes rather than treating natural waxes as a homogeneous material class [92,93].

For food-packaging coatings development, relevant characterization should include chemical composition, melting and crystallization behavior, thermal stability, crystallinity, particle size or dispersion characteristics, surface energy or water contact angle, and rheological behavior of the coating formulation. Depending on the intended application, coating thickness or coat weight, surface morphology, adhesion, water absorption, water-vapor transmission, grease resistance, and mechanical durability should also be evaluated. Such characterization is essential for relating wax composition and processing conditions to coating performance and for comparing results obtained with different wax sources [57,69,89,91,92].

The barrier performance of wax coatings on cellulosic substrates is attributed to the formation of a low surface energy layer, physical blocking, and surface crystallization. While water is primarily repelled by wax's low surface energy, grease resistance is achieved through a mechanical blocking effect and a compact crystalline matrix that prevents oil diffusion. The effectiveness of these mechanisms depends on both wax characteristics and

substrate properties, including porosity, surface roughness, and surface chemistry. These factors influence coating coverage, adhesion, and barrier performance, making substrate–wax interactions important for the development of effective food-packaging coatings.

6.2. Wax–Cellulose Interactions and Coating Formation

Cellulose-based substrates are suitable for natural wax coatings because their porous structure allows wax particles or droplets to penetrate and become physically retained within surface irregularities. However, the low affinity between a hydrophobic wax phase and the hydrophilic cellulosic surface can limit interfacial adhesion and promote coating defects. The hydroxyl-rich surface of cellulose can provide sites for interfacial interactions, but these are generally insufficient to ensure strong adhesion of non-polar waxes. Consequently, substrate porosity, surface roughness, surface chemistry, and coating formulation all influence wax distribution, adhesion, and barrier performance. Paper and paperboard are nevertheless compatible with a wide range of coating technologies.

When combined with nanocellulose (CNC, CMF, or CNF; Figure 4), waxes can be distributed around fibrillar structures and within surface cavities, generating heterogeneous or hierarchical surface morphologies that can contribute to water and grease resistance [58,60,90]. Aggregation or incomplete coverage can generate defects that act as preferential pathways for liquid penetration and compromise barrier performance [94].

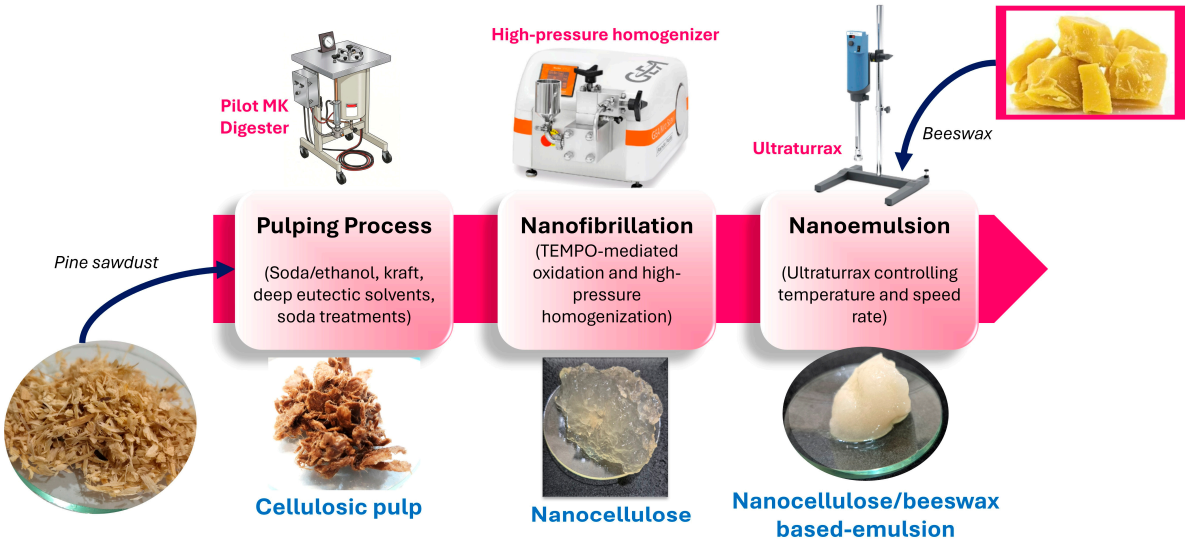


Figure 4. CNF/beeswax-based emulsions prepared at the Programa de Celulosa y Papel (PROCYP; Pulp and Paper Program) research group (elaborated by the authors).

6.3. Effect of Wax Processing on Surface and Barrier Properties

The incorporation of natural waxes into cellulose- and nanocellulose-based coatings can improve water repellency, water-vapor barrier performance, and resistance to oils and greases through their low polarity and ability to form hydrophobic domains. Their effect on oxygen permeability is generally more limited because oxygen transport remains strongly influenced by the continuity and density of the underlying cellulosic or nanocellulose network [58,60,90]. Table 3 summarizes the main effects of natural waxes on surface and barrier properties.

Applied studies have also reported stable hydrophobic properties on regenerated paper, cotton fabrics, and kraft paper, supporting the potential of natural waxes for food-packaging applications [18,90]. However, performance is strongly application-specific and

depends on wax composition, formulation, coating weight, substrate characteristics, and processing conditions.

Grease and oil resistance are particularly relevant to PFAS-free food packaging because they are key performance requirements traditionally addressed by fluorinated treatments. Although natural waxes generally do not provide the extreme oleophobicity associated with fluorinated surfaces, well-designed CMF/CNF–wax hybrid systems could demonstrate promising resistance to grease and oil in application-specific tests. Direct comparison with PFAS-based treatments remains difficult because reported values depend on substrate, coating weight and morphology, and test method.

Table 3. Effect of natural waxes on surface and barrier properties of cellulose-based coatings.

Property	Effect of Natural Waxes	Main Mechanism	Ref.
Water wettability	Increases water contact angle and reduces surface wetting	Low surface energy and formation of hydrophobic surface domains; wax crystallization can additionally modify surface topography	[17,43,90]
Water absorption	Reduces water uptake by the coated substrate	Reduced interaction between water and cellulose and partial filling or sealing of surface pores	
Water-vapor transmission	Reduces water-vapor transmission rate (WVTR)	Lower water sorption and increased resistance to vapor diffusion through the hydrophobic phase	[58,60]
Grease and oil resistance	Improves resistance to lipid penetration	Hydrophobic surface chemistry, pore blocking, and formation of relatively compact crystalline wax domains	[60,79]
Oxygen transmission	Generally, provides a limited or moderate improvement	Partial sealing of pores and reduction in transport pathways; the overall oxygen barrier remains strongly dependent on the underlying cellulose or nanocellulose matrix	[38]
Surface morphology	Modifies surface roughness and can generate hierarchical structures	Wax deposition, particle coalescence, and crystallization during drying	[79,90]
Mechanical properties	Effects depend on wax type and concentration; excessive wax can reduce coating cohesion	Wax crystallinity, phase distribution, and coating cohesion	[76,79]

7. Processing Limitations and Technological Challenges

Despite their potential, natural wax-based coatings still face several challenges before they can be widely adopted as alternatives to PFAS-based treatments. Mechanical durability is an important concern, particularly under bending, folding, and abrasion. Thermal stability can also limit their use in applications involving heat processing, sealing, or sterilization, as some natural waxes soften or become brittle within relatively narrow temperature ranges. In addition, the hydrophobic nature of waxes can hinder their adhesion to hydrophilic cellulose, making homogeneous dispersion, coating uniformity, and interfacial compatibility particularly important. These effects are especially relevant for highly porous cellulosic substrates, where wax penetration, aggregation, or incomplete surface coverage can lead to variations in barrier performance [79]. Increasing wax content generally improves moisture resistance, but excessive amounts may compromise adhesion, coating integrity,

or mechanical performance. Consequently, the optimum wax concentration represents a balance between hydrophobicity, barrier performance, coating cohesion, and processability rather than a simple maximization of wax content. Similar trade-offs have been reported for beeswax and other lipid-containing coatings, highlighting the importance of formulation optimization [76].

Complete replacement of PFAS remains challenging in applications requiring exceptionally high and durable grease and moisture resistance. Although cellulose–wax hybrid systems can provide effective barriers for several food-packaging applications, their performance depends strongly on wax composition, formulation, coating structure, substrate characteristics, coat weight, and processing conditions. In addition, migration, long-term durability, recyclability, and performance under realistic food-contact conditions require validation at the level of the complete coated packaging system rather than the coating alone.

Candelilla, carnauba, and beeswax have all shown potential as hydrophobic components, but their performance is application-specific and cannot be considered equivalent to that of fluorinated treatments. Candelilla wax, for example, combines relatively high crystallinity and hydrophobicity and has demonstrated promising moisture and grease-barrier properties [90]. However, the choice of wax should consider not only barrier performance but also melting behavior, crystallization, adhesion, processability, and compatibility with the intended recycling or end-of-life pathway [79,89,90].

Future research should therefore focus on controlling wax morphology and distribution, improving wax–cellulose interfacial adhesion, optimizing coating weight and formulation stability, and developing processes compatible with continuous industrial coating. Attention should be given to standardized application-relevant testing of grease, water-vapor, oxygen, mechanical, and thermal performance, with migration, repulpability, and end-of-life assessment. These aspects are essential for determining whether a given natural wax formulation can provide a technically and environmentally viable alternative to PFAS-based treatments.

Table 4 summarizes representative studies of natural wax coatings applied to cellulosic materials. Comparing the different waxes with the paper itself makes it possible to observe changes in barrier properties.

Jahangiri et al. [79] found that the water and oil contact angles of the uncoated paper increased after the addition of wax coatings. The water contact angle increased from 98.5° to 105° in bio-based wax, 107° in beeswax, 109° in soywax, and 113° in paraffin wax-coated papers. In oil contact angle, the trend was different, following values from 43° (paper) to 58° after bio-based wax, 52° beeswax, 57° soywax, and 54° paraffin wax addition. Regarding WVP, the trend showed a decrease from 4.5×10^{-6} g/(m^2·s·Pa) for the base paper to 1.54×10^{-10} g/(m^2·s·Pa) following the application of a beeswax coating. When evaluated at 37.8 °C and 90% RH, the uncoated paper exhibited a WVP of 874.2 g·mil/(d·m^2·kPa), whereas the beeswax-coated paper reduced this value to 196.0 g·mil/(d·m^2·kPa), a decrease of nearly 80%.

Meanwhile, the values obtained for papers coated with biowax, soywax, and a paraffin-based emulsion were approximately 360 g·mil/(d·m^2·kPa), 470 g·mil/(d·m^2·kPa), and 670 g·mil/(d·m^2·kPa), respectively.

Another study on kraft paper demonstrated a 67.5% reduction when carnauba wax was applied as a coating (WVP value from 3.1×10^{-6} g/m·Pa·h to 0.9×10^{-6} g/m·Pa·h) [95]. The same author observed that the wettability rate of these papers decreased from 16.1%/min to 2.36%/min with the carnauba wax coating.

Table 4. Selected studies on natural wax coatings for cellulosic materials.

Wax Type	Cellulosic Material	Application Methods	Main Findings	Ref.
Beeswax, candelilla wax, paraffin	Paper and cellulose-based films	Melt coating	Candelilla wax showed high crystallinity and provided favorable water-vapor barrier and stiffness compared with the other waxes.	[89]
Beeswax	Regenerated cellulose	Immersion/melting	Beeswax treatment produced persistent hydrophobicity after repeated use and washing cycles.	[96]
Refined natural waxes	Kraft paper	Bar coating	Wax coating increased surface hydrophobicity and reduced water-vapor transmission.	[79]
Natural waxes	Nanocellulose and paper	Spray coating, size press	Wax incorporation improved surface water repellency and barrier performance; the resulting properties depended on coating structure and processing conditions.	[97]
Beeswax	Cellulosic fabrics	Hot-emulsion coating	Beeswax treatment provided moisture repellency with adequate adhesion and moderate durability.	[95]
Natural hydrophobic waxes	CNF and CMF	Spray coating, self-assembly	Wax–nanocellulose systems produced hydrophobic surfaces and demonstrated the potential of combining fibrillar cellulose with hydrophobic wax phases.	[98]
Candelilla wax	Paper and food substrates	Fusion and ironing	Candelilla wax provided effective grease and moisture resistance, although performance was dependent on coating conditions and substrate characteristics.	[90]

CNF, cellulose nanofibrils; CMF, cellulose microfibrils.

8. Processing–Structure–Performance Relationships

8.1. Comparison of Barrier Mechanisms

PFAS-based treatments have historically been used in paper and paperboard food packaging primarily because they provide strong water and grease repellency through the low surface energy of fluorinated structures. Their high chemical stability contributes to the persistence of these materials in the environment, which is one of the main concerns associated with their use. The overall barrier performance of PFAS-treated packaging, however, also depends on the substrate, coating formulation, coat weight, and application method.

In contrast, bio-based barrier systems exhibit a more heterogeneous performance profile that depends strongly on formulation, microstructure, coating thickness, substrate characteristics, and processing conditions. Nanocellulose is particularly effective as an oxygen barrier because dense fibrillar networks formed through extensive intermolecular interactions create tortuous pathways that restrict gas diffusion and reduce effective porosity. High oxygen-barrier performance has been demonstrated for nanocellulose coatings on paper and paperboard, particularly when continuous and homogeneous layers are obtained [38]. Multilayer configurations can further improve overall barrier performance by combining the oxygen-barrier properties of nanocellulose with hydrophobic layers that protect it from moisture [99].

The main limitation of hydrophilic bio-based coatings is their comparatively limited resistance to moisture. Natural waxes can reduce water uptake and water-vapor transmission,

while lignin or nanolignin may modify surface properties and interfacial interactions. The effectiveness of these approaches depends on material chemistry, dispersion, and coating morphology [100]. Thus, hybrid systems offer complementary barrier functions, although their performance depends on the formulation and the quality of the resulting coating.

For grease and oil resistance, nanocellulose coatings can substantially improve the resistance of paper to lipid penetration by reducing surface porosity and forming continuous, dense layers [38]. However, this mechanism differs from the strong surface repellency associated with fluorinated treatments. Consequently, nanocellulose alone may be insufficient for applications requiring high and persistent grease resistance, particularly under demanding contact conditions. Combining nanocellulose with hydrophobic components such as natural waxes provides an opportunity to introduce complementary moisture and grease resistance while retaining the oxygen-barrier contribution of the cellulose network.

The comparison is therefore not simply between PFAS and bio-based materials, but between different combinations of barrier mechanisms. PFAS-based treatments offer highly effective surface repellency, whereas nanocellulose provides a particularly strong oxygen barrier under dry conditions. Hybrid bio-based systems can combine these functions through complementary components, but their performance remains more dependent on formulation, structure, processing, and environmental conditions. The central technical challenge is consequently to achieve sufficient moisture and grease resistance without compromising the oxygen barrier, mechanical integrity, processability, or end-of-life performance of the packaging system. Recent reviews similarly identify moisture sensitivity, scalability, and validation under realistic conditions as major barriers to the industrial implementation of nanocellulose-based packaging [36,43,60].

8.2. Mechanical Performance of Bio-Based Barrier Systems

Mechanical performance is an important consideration for food-packaging coatings, particularly when coated substrates must be converted, folded, bent, handled, and abraded. PFAS-based treatments are generally applied as thin functional layers and are not intended to reinforce the substrate. Their contribution to the mechanical properties of the final packaging is therefore limited, making direct mechanical comparisons with nanocellulose-based systems inappropriate.

Nanocellulose can contribute to the mechanical performance of bio-based barrier systems through its high aspect ratio, large specific surface area, and extensive inter-molecular interactions. CNF and CMF can form interconnected networks that increase tensile strength and elastic modulus, particularly when the fibrils are well dispersed and strongly associated with the surrounding matrix [13]. When applied as a coating, however, the extent of mechanical reinforcement depends on coating weight, adhesion to the substrate, drying conditions, and the integrity of the resulting layer. Excessive coating solids or nanocellulose content can also increase formulation viscosity and complicate coating and processability [73]. The increase in strength and stiffness may be accompanied by reduced elongation at break and increased brittleness. This trade-off is particularly relevant for nanocellulose-rich films and coatings, which may crack during folding or deformation if sufficient flexibility and adhesion are not achieved. In starch-based and other bio-based matrices, the balance between strength and ductility can be modified through plasticization and formulation design [46]. Hybrid systems incorporating polymers or other flexible components, such as PLA–CNF or PVA–CNF, have therefore been investigated to improve the balance between mechanical strength and flexibility [19,55].

For coated paper and paperboard, mechanical performance should consequently be considered at the level of the complete packaging structure rather than the coating alone. A coating that provides excellent barrier properties but cracks, delaminates, or loses adhesion

during converting or use may not provide effective protection in practice. The main challenge is therefore to achieve sufficient coating cohesion and substrate adhesion while maintaining flexibility, barrier performance, and processability.

Nanocellulose offers a potential mechanical advantage over conventional functional surface treatments because it can contribute both barrier functionality and reinforcement. However, this advantage is highly dependent on coating architecture and application conditions and should not be interpreted as an intrinsic property of all nanocellulose-based coatings.

9. Scale-Up and Industrial Processing

Table 5 compares the main bio-based coating systems discussed in this review in terms of barrier and mechanical performance, processability, advantages, and limitations.

Beyond laboratory-scale performance, successful industrial implementation requires stable formulations, reproducible coating quality, compatibility with existing coating and papermaking equipment, adequate mechanical and thermal durability, and compliance with food-contact requirements. In addition, the economic viability of the complete coating process must be considered, including raw-material availability, formulation cost, energy consumption, drying requirements, and achievable production rates.

9.1. Material and Formulation Limitations

At the industrial level, a key challenge is ensuring that coating performance remains consistent under variable processing, storage, and end-use conditions. Formulations must therefore be designed and validated to maintain adequate barriers and mechanical performance across the humidity ranges relevant to the intended application. Moisture sensitivity remains an important consideration in the industrial implementation of nanocellulose-based packaging [77,78].

The studies analyzed show changes in OTR following the addition of nanocellulose-based coatings. Aulin et al. [38] showed that, starting from a free-flowing state on kraft-type base paper, the OP was reduced to 0.006 $cm^3 \cdot \mu m/m^2 \cdot d \cdot kPa$ (0% RH) and 0.85 $cm^3 \cdot \mu m/m^2 \cdot d \cdot kPa$ (at 50% RH). Similarly, after the addition of CNC to PLA/PHB mixtures, the OTR decreased from 13.3 $cm^3 \cdot mm/m^2 \cdot day$ to 13.0 $cm^3 \cdot mm/m^2$ [38]. Tayeb et al. [50] achieved a similar trend in regular printing grade (basis weight of 78 g/m^2) paper. The uncoated paper showed high permeability and the OTR test failed, while CNF and LCNF showed lower OTR values. OTR values at 50%RH were 19.9 $cm^3/m^2 \cdot day$ and 33.7 $cm^3/m^2.day$ with CNF and LCNF addition respectively. The presence of lignin increased the OTR by around 50% compared to the addition of CNF.

WVP/WVTR showed decreases after the addition of CNF or CNF/waxes emulsions. In paper printing (WVP: 36.2 $g \cdot mm/m^2 \cdot kPa \cdot day$), the addition of CNF and LCNF (16 g/m^2 coating) reduced the WVP values to 5.0 and 5.3 $g.mm/m^2 \cdot kPa \cdot day$ respectively [17]. When a CNC coating, collagen, and lignin are added to a base printing paper, the WVP decreased from 65.6 $g/m^2 \cdot day$ to 6.1 $g/m^2 \cdot day$ [101]. The trend is similar in food-grade paper sheets, where the addition of CNF/wax emulsion reduced the WVTR from 700 $g/m^2.day$ to 100 $g/m^2.day$ [58]. The decrease in WVP values with CNF addition was also observed in PVA films (from $1.7.10^{-8}$ to 6.10^{-9} $g/Pa^2 \cdot s^2 \cdot m^2$) [55]. The air permeability values were also variable according to the coating and base paper. Bayes et al. [58] registered an increase in the time of the Gurley test when CNF/wax was applied (90 s vs. 600 s).

Table 5. Comparison of bio-based barrier coating systems for paper-based food packaging.

Coatings	Barrier Performance	Mechanical Performance	Processability	Advantages	Limitations	Ref.
CMF: 0.85 wt%, using a rod coater. Six coats on one side and dried under ambient conditions. CNF: 0.2% wt%, multilayer films (40 g m^{-2}), pressing at 100 bar and heating at 100 C°. (a) CNF film; (b) sandwich: wax layer + CNF films; (c) multilayer: sandwich + lignin with chitin nanofibers	Excellent oxygen barrier under dry conditions; limited moisture resistance. CMF: 0.85 OP at 50% RH Multilayers: OP (50% RH): (a) 1.4, (b) 1.5, (c) 1.5 WVTR: (a) 197, (b) 5, (c) 6.	High stiffness and tensile strength. CMF: DMA modulus: 30 GPa (5% RH), 18.9 GPa (at 90% RH). Tan δ below 0.1 and independent of RH CNF: Mix configuration: similar mechanical properties (strength, strain, and Young's Modulus) as CNF film. Sandwich and top layer: similar maximum strain but lower strength and Young's modulus.	Spray, blade, size press	Renewable; strong oxygen barrier; film-forming	Humidity sensitivity; high suspension viscosity	[38,99]
CNC-coated recycled paper. CNCs (1%, 3%, 5%, 7%, 10%), 10% collagen hydrolysate, and (0.5–1) mL of glycerin. Rod coating, 35 g m^{-2}, thickness 0.03 mm, drying by hot air flow.	Improved water resistance. Coated papers: (a) passed the hot oil barrier test, (b) water-vapor permeability <15 g m^{-2} day^{-1}, (c) air permeability <50 mL min^{-1}	Improved compression resistance. 3% coated paper: (a) tensile index 74.43 N m g^{-1} (5.28 times higher than uncoated), (b) burst index 2.10 ± 0.09 kPa m^2 g^{-1} (1.79 times higher than uncoated), (c) burst index 2.10 ± 0.09 kPa m^2 g^{-1} (1.79 times higher than uncoated).	Spray coating	Compatible with recycled paper; relatively simple application	Increasing CNC content can increase viscosity and affect workability	[101]

Table 5. *Cont.*

Coatings	Barrier Performance	Mechanical Performance	Processability	Advantages	Limitations	Ref.
Lignin-containing coatings. The 1 wt% CNF and LCNF were applied to a round cut through vacuum-assisted filtration, mild pressure to be oven dried at 70 °C, substrate paper 78 g/m^2, coated papers 94 g/m^2 containing 16 g/m^2 of CNF or LCNF.	Variable; potential improvement in moisture resistance. Compared to CNF, the presence of lignin increased the OTR by around 50% for the low humidity case and by 100% for the high humidity case.	Formulation-dependent. Tensile strength increased from 36 to 41 and 45 MPa with the addition of LCNF and CNF layer respectively.	Formulation- and process-dependent	Renewable; reduced surface energy; potential hydrophobic contribution	Interfacial compatibility and dispersion limitations	[50]
Starch, PLA, PVA (a) 75 wt% of PLA + 25 wt% of PHB + 5 wt% CNC, (b) PVA films adding CNF (1, 3, 5, and 7% wt% of PVA), (c) 10 wt% CNF + TPS + glycerol.	Variable; strongly formulation dependent. (a) OTR 15.3 cm^3 326 mm m^{-2} day^{-1} for PLA-PHB-CNC, 13 cm^3 mm m^{-2} day^{-1} for PLA-PHB-CNCs, (b) 5% CNF-PVA: WVP decreasing from 0.1749 for pure PVA to 0.0622 (g·Pa^{-1}·s^{-1}·m^{-2}·10^{-7}).	Strongly formulation dependent. (a) PLA film deformation was reduced with PHB and CNC, increasing PLA modulus and reducing tensile strength; (b) tensile strength: PVA: 29.78 MPa, PVA-5%CNF: 41.28 MPa.	Casting; selected extrusion and coating processes	Bio-based; tunable properties	Moisture sensitivity; brittleness in some formulations; limited industrial maturity	[19,55,102]
CNF/CMF–wax hybrids (a) molten wax + CNF films (0.2 g) with a heated glass rod, (b) 0.7 wt% and 0.9 wt% TEMPO-CNF, wax and glycerol: 10 wt% and 1 wt%, smooth roll blowing air at 50 °C for 60 s, and dry at room temperature.	Improved moisture and grease resistance while retaining oxygen-barrier contribution from nanocellulose. (a) Wax coating significantly reduced the WVTR: from 219.2 (uncoated) to 77.8 (wax-coated) g.m^{-2}.day^{-1}; (b) barrier properties to air and water vapor were greatly improved over those of the original paper	Formulation- and adhesion-dependent.	Spray, size press, blade	Renewable; multifunctional; PFAS-free; complementary barrier functions	Wax migration; emulsion stability; interfacial compatibility; humidity sensitivity	[58]

CNF: cellulose nanofibrils. CNC: cellulose nanocrystals. CMF: cellulose microfibrils. PLA: polylactic acid. PVA: poly(vinyl alcohol).

Baral et al. [101] found the same trend when applying a CNC/collagen/glycerin, reaching a decrease in air permeability from 263 mL/min to 10 mL/min.

Water CA for the base papers studied was in the range of 40° to 60°. But, with the addition of CNF/beeswax emulsions, the values can reach up to 115° [58]. When CNC/collagen/glycerin was applied to office paper, the CA increased from 60° to 99° [101]. If we compare the addition of CNF with LCNF, it is observed that the residual lignin increased even more from 59° (CNF) to 82° (LCNF) [50].

Oil contact angle also increased from 20–30 to above 60 when CNF/waxes emulsions are added as a coating [58]. In most cases, when performing the kit test on base papers, a kit number of 0 or 1 is recorded. Upon the application of coatings, these values reach 5 when using a CNF/beeswax emulsion [58], up to a maximum of 12 with CNF, LCNF, or a mixture of CNC/collagen/glycerin [101].

The stability of waxes and other hydrophobic components is another important consideration. Crystallization, phase separation, migration, and redistribution during storage or use can alter coating morphology and compromise uniformity and long-term barrier performance, particularly in multicomponent formulations [79]. Emulsion stability, wax particle size, solids content, and rheological properties must therefore be controlled to obtain reproducible coatings. These parameters are particularly important for water-based formulations intended for high-speed coating processes, where changes in viscosity or dispersion stability can directly affect coat weight, coverage, and drying behavior.

Interfacial compatibility between waxes or bio-based polymers and nanocellulose or cellulose also remains challenging. Differences in polarity and chemical structure can promote phase separation, poor adhesion, and microstructural defects, which may adversely affect both mechanical integrity and barrier performance [103]. Improving interfacial interactions through formulation design, surface modification, compatibilization, or multilayer architectures can enhance coating uniformity, but these strategies may also increase formulation complexity and processing requirements.

Another consideration is the balance between barrier performance and processability. Increasing the concentration of nanocellulose can improve network formation and oxygen-barrier properties, but also increases suspension viscosity and may complicate pumping, metering, and coating at industrial solids contents. Similarly, increasing wax concentration can improve water and grease resistance but may compromise adhesion, coating cohesion, or mechanical durability. Consequently, formulation optimization should consider barrier performance and processing behavior simultaneously rather than maximizing the concentration of any individual component.

Finally, the thermal stability and potential migration of functional components, including nanomaterials, pigments, and active agents, must be considered. Long-term migration data under realistic conditions of use, storage, and recycling remain limited for many emerging bio-based formulations, representing an important barrier to regulatory acceptance and industrial adoption. For nanocellulose-containing systems, regulatory assessment should consider the specific form and surface characteristics of the nanomaterial, the complete coating formulation, its intended use, and the potential for migration into food.

The main challenge is not the lack of bio-based components with suitable individual properties, but the need to combine them into stable, homogeneous, and processable formulations that maintain their performance throughout manufacturing, storage, and use. This formulation–process interaction is particularly critical for nanocellulose–wax hybrid coatings, in which hydrophilic and hydrophobic components must be simultaneously controlled.

9.2. Cost and Scalability

Economic competitiveness is one of the main challenges for replacing established surface treatments with bio-based barrier systems. PFAS-based grease-proofing treatments historically benefited from mature industrial production, established supply chains, and optimized application processes. However, their use in food-contact paper and paperboard is being progressively eliminated. In the United States, PFAS-containing grease-proofing substances were no longer being sold for food-contact use by February 2024, and the FDA subsequently determined that 35 related food-contact notifications were no longer effective [6,7]. In the European Union, restrictions on PFAS in food-contact packaging entered into application on 12 August 2026, under the Packaging and Packaging Waste Regulation (PPWR) [34,35].

The economic comparison with bio-based alternatives is complex because the coating cost depends not only on the price of its individual components but also on coating weight, solids content, formulation composition, application method, drying requirements, and production scale. For nanocellulose, published techno-economic assessments report production costs ranging from approximately USD 4 to 11/kg for CNF and related materials, with lower values potentially achievable under favorable feedstock and large-scale process conditions [104–106]. These estimates, however, should not be directly compared with the price of conventional coating chemicals because the required dosage and processing conditions may differ substantially.

The relatively high cost of nanocellulose is associated with pretreatment, chemical consumption, mechanical fibrillation, energy demand, wastewater treatment, and capital investment. Consequently, its use as a coating component should be optimized rather than maximized. The required weight coating is particularly relevant because a relatively expensive functional material can remain economically attractive when applied at low add-on levels. For PFAS-based treatments, historical applications included relatively low additions for grease-resistant papers, although higher levels were used for some demanding food-contact boards [6,7]. This illustrates why cost per kg alone is insufficient for comparing alternative coating technologies.

Natural waxes offer a different economic profile. Their cost varies considerably with wax type, purity, origin, and supply chain. Commodity natural waxes may be less expensive than specialty nanocellulose grades, but their availability and composition can be more variable. Direct comparison on a USD/kg basis is therefore of limited value; the relevant parameter is the contribution of each component to the cost per unit area of coated substrate while meeting the required barrier performance.

Formulation composition is consequently critical to the economic feasibility of hybrid systems. Nanocellulose can provide oxygen-barrier performance, whereas natural waxes can contribute to hydrophobicity and grease resistance. Combining the two may reduce the amount of each component required to achieve the target performance. The economic potential of hybrid coating should therefore be evaluated based on formulation cost, coating weight, process yield, drying energy, and production rate rather than the cost of individual ingredients alone.

Feedstock selection and process integration provide additional opportunities to reduce costs. Producing nanocellulose and other functional components from agricultural and forestry residues can reduce dependence on dedicated feedstock while improving resource efficiency. Integration into biorefinery schemes can further improve process economics by sharing pretreatment, separation, energy, and wastewater-treatment infrastructure among multiple products [107]. However, heterogeneous feedstocks can introduce variability in composition and quality, requiring robust pretreatment and process-control strategies to ensure consistent product properties during scale-up.

Scalability also depends on the ability to produce stable formulations at industrially relevant solids contents and production rates. Increasing solids content can reduce drying requirements but may increase viscosity and complicate pumping, metering, and coating. Conversely, dilute formulations may be easier to process but require greater drying capacity and energy consumption. These trade-offs must be considered together with coating speed, coat weight, substrate characteristics, and the selected application technology.

The economic feasibility of PFAS-free bio-based coatings will depend on achieving an appropriate balance among material cost, coating add-on, barrier performance, processing requirements, drying energy, and production scale. Rather than seeking a one-to-one replacement of PFAS by a single bio-based component, hybrid formulations combining complementary functions may provide a more economically viable strategy. Further techno-economic assessments at pilot and industrial scales are needed to identify competitive combinations of nanocellulose, natural waxes, and other bio-based components.

Table 6 presents some selected economic and technological parameters relevant to PFAS-free barrier coating systems.

Table 6. Selected economic and technological parameters relevant to PFAS-free barrier coating systems.

Parameter	PFA-Based Treatment	Nanocellulose	Natural Waxes	CNF/CMF Hybrids	Ref.
Material cost	Historically competitive at low add-on	Approximately USD 4–11/kg in reported techno-economic scenarios, depending on material, process, and scale	Highly dependent on wax type, purity, origin, and grade	Formulation-dependent	[104–110]
Typical coating/add-on	From 2 to 3 g/m^2 for some grease-resistant papers; higher levels reported for demanding food-contact boards	Application-dependent; low coating weights are desirable	Application-dependent	Application-dependent	[111,112]
Main cost drivers	Material cost and established application infrastructure; regulatory transition	Pretreatment, chemicals, fibrillation, energy, wastewater treatment, and capital investment	Raw material, purity, availability, and formulation	Nanocellulose and wax content, dispersion/emulsion stability, and drying	[104–110]
Effect of scale-up	Mature industrial scale	Strong economies of scale reported	Dependent mainly on raw-material supply and process scale	Expected to benefit from integration with existing coating operations	[104–106]
Industrial maturity	High historically; restricted for food-contact applications	Emerging to increasing	Commercially established raw materials	Emerging	[104–106]
Main economic opportunity	—	Low-cost residues and biorefinery integration	Renewable feedstocks and regional availability	Complementary barrier functions may reduce the required amount of each component	[79]
Key scale-up issue	Regulatory transition	Viscosity, energy demand, drying, and production cost	Adhesion, crystallization, and morphology control	Formulation stability, interfacial compatibility, and process control	[104–106]

CNF: cellulose nanofibrils. CMF: cellulose microfibrils.

9.3. Process Integration and Industrial Implementation

Compatibility with existing manufacturing infrastructure is a relevant factor determining the industrial adoption of bio-based barrier coatings. For paper and paperboard,

nanocellulose-based formulations can be applied using established aqueous coating technologies, including size press, spray, blade, and other continuous coating processes, potentially reducing the need for major modifications to existing production lines [113]. The choice of application method affects coating weight, thickness, uniformity, drying requirements, and ultimately barrier performance. Size-press and spray coating are particularly attractive for integration with continuous paper-processing operations, whereas rod and blade coating provide greater control over coating thickness at laboratory and pilot scales [113–115]. The critical issue is therefore not the availability of suitable coating equipment, but whether the formulation can be processed reliably within the operating window of industrial equipment.

Scale-up requires simultaneous control of formulation rheology, solids content, coat weight, drying, and coating uniformity. The relatively high viscosity of concentrated nanocellulose suspensions can limit solids content and affect pumping, metering, and coating speed, whereas dilute formulations increase the amount of water that must be removed during drying. At industrial line speeds, these factors directly affect productivity, energy consumption, and coating quality.

For hybrid nanocellulose–wax systems, process integration introduces additional challenges because the formulation must remain stable during storage, pumping, and application while allowing controlled deposition and drying. Wax particle size, emulsion stability, rheology, substrate wettability, and drying conditions can influence phase distribution and surface morphology. Maintaining a homogeneous coating at high production rates is therefore essential for obtaining reproducible barrier performance.

Multilayer structures provide another integration strategy, particularly for flexible packaging. Bio-based barrier layers can be combined with other functional layers to provide complementary oxygen, moisture, grease, mechanical, or sealing properties without requiring complete replacement of existing converting technologies [99]. Such architectures may facilitate a gradual transition toward PFAS-free packaging while allowing individual functions to be optimized separately.

The most immediate route to industrial implementation is therefore optimized bio-based formulations integration into existing coating and converting operations rather than entirely new manufacturing process development. Pilot-scale studies should establish operating windows for solids content, rheology, coating speed, coat weight, drying, and formulation stability before industrial implementation [116,117].

9.4. Food-Contact Safety and Migration

The suitability of nanocellulose–natural wax coatings for food packaging depends not only on their functional performance but also on the safety of their constituents and their potential to migrate into food. Assessment should consider the complete formulation, including nanocellulose, wax components, emulsifiers and surfactants, other additives, and residual chemicals arising from raw-material preparation or coating manufacture. Natural origin alone does not establish food-contact safety; the purity, composition, intended use, potential exposure, and toxicological properties of the individual constituents must also be considered [118].

Migration may occur through direct contact between the coated surface and food, transfer through the gas phase, or set off when packaging materials are stacked or wound into rolls. Abrasion or release of particulate material may also be relevant for coatings containing nanomaterials. The extent of migration depends on factors such as the chemical nature and concentration of the migrant, coating structure and thickness, food composition, contact time, temperature, and the ratio of packaging surface area to food volume [118,119]. These variables should be considered when selecting test conditions representative of the

intended application, including the types of food to be packaged and foreseeable storage and use conditions.

Overall migration and specific migration provide complementary information. Overall migration measures the total amount of material transferred under defined test conditions, whereas specific migration quantifies individual substances of concern. For complex coatings, the assessment should also consider non-intentionally added substances (NIAS), including impurities, reaction by-products, residual processing chemicals, and degradation products that may form during manufacture, storage, or use [118]. Depending on the formulation, analytical screening may be required to identify potential migrants before their toxicological relevance can be assessed. Emulsifiers, surfactants, and other formulation aids deserve particular attention because their presence in an aqueous coating does not guarantee that they remain immobilized after drying or during contact with food.

Nanocellulose introduces additional questions related to material identity, particle size and morphology, surface modification, purity, and the potential release of nanoscale or microscale material from the coating. Hu et al. [119] reviewed recent advances in the safety assessment of nanocellulose in food packaging and highlighted the importance of appropriate characterization, migration assessment, and toxicological evaluation. Depending on the manufacturing route and raw materials, the assessment may also need to consider residual chemicals from pretreatment or purification, degradation products, and mineral or metal contaminants. The relevance of these hazards depends on the specific production process and should be established through appropriate analytical characterization rather than presumed.

Food-contact compliance must be evaluated within the applicable regulatory framework. In the European Union, Regulation (EC) No. 1935/2004 [120] establishes general requirements for materials intended to contact food, including that their constituents must not transfer to food in quantities that could endanger human health, cause unacceptable changes in food composition, or deteriorate its organoleptic properties. Regulation (EC) No. 2023/2006 [121] establishes requirements for good manufacturing practice. Additional requirements depend on the material category and the applicable national or regional provisions; the specific migration limits and overall migration limit established for plastics should not automatically be assumed to apply to paper and board or to every coating formulation.

For nanocellulose–natural wax coatings, a suitable safety assessment should therefore combine formulation and raw-material characterization with migration testing under relevant time and temperature conditions, targeted analysis of potential migrants, and toxicological assessment where necessary. Particular attention should be paid to the purity of natural waxes, the identity and concentration of emulsifiers and surfactants, residual processing chemicals, and possible particulate release. Demonstrating satisfactory barrier properties is not, by itself, sufficient to establish food-contact safety. Further validation under conditions representative of the intended application is needed before these hybrid coatings can be considered suitable for specific food-packaging uses. However, migration testing may need to address particulate release in addition to conventional chemical migration [122].

9.5. Remaining Barriers to Industrial Implementation

Although several bio-based coating technologies are compatible with established application methods, their industrial adoption remains constrained by the need to demonstrate consistent performance under realistic production and use conditions. The remaining barriers therefore extend beyond the intrinsic properties of the materials and include formulation robustness, coating reproducibility, durability, food-contact safety, regulatory compliance, and economic competitiveness.

A key requirement is demonstrating that the complete coated packaging system provides the required combination of oxygen, water-vapor, and grease resistance while maintaining mechanical integrity during converting, storage, and use. Performance should be evaluated at application-relevant coating weights and under realistic temperature, humidity, and food-contact conditions rather than solely under optimized laboratory conditions.

For nanocellulose–wax hybrid systems, particular attention should be given to the long-term stability of the coating. Changes in wax morphology, crystallization, migration, or interfacial adhesion may alter barrier performance during storage or use. These effects should be evaluated together with repulpability, recyclability, and, where relevant, compostability of the complete packaging structure.

Food-contact compliance represents an additional barrier to commercialization. Migration assessment should consider the complete formulation, including nanocellulose, waxes, additives, processing aids, and any other functional components, under conditions representative of the intended food application. Regulatory requirements will also vary among markets and may influence formulation selection and commercialization strategies.

The transition from laboratory-scale formulations to commercial products requires simultaneous optimization of material composition, coating process, cost, durability, and regulatory performance. For nanocellulose–wax systems, the most realistic pathway is likely to involve application-specific formulations integrated into existing paper and packaging production technologies rather than a universal replacement for PFAS. Demonstration at pilot and industrial scales, supported by standardized barrier testing, food-contact evaluation, life-cycle assessment, and techno-economic analysis, will be essential to establish their commercial viability.

10. Conclusions and Future Perspectives

The evidence reviewed highlights the importance of integrating PFAS replacement strategies with the design of multifunctional barrier coatings based on nanocellulose, natural waxes, and complementary materials. Nanocellulose provides strong oxygen-barrier performance, particularly under dry conditions, whereas natural waxes primarily improve moisture and grease resistance. However, achieving complementary and reliable barrier functions requires careful control of phase distribution, interfacial interactions, coating continuity, wax crystallization, and substrate coverage. These relationships demonstrate that the performance of hybrid coatings depends not only on the intrinsic properties of their components but also on how they are formulated, deposited, and dried.

Future research should focus on pilot- and industrial-scale validation under realistic food-packaging conditions. Considerable attention should be given to formulation stability, rheological behavior, coating uniformity, drying and crystallization, and the durability of coatings during converting, storage, and use. Food-contact safety, recyclability, end-of-life behavior, and the economic and environmental performance of complete coated structures should also be systematically evaluated.

Rather than pursuing a one-to-one replacement of PFAS, future development should focus on application-specific formulations and processing strategies that combine complementary materials to achieve the required barrier performance at viable coating weights. Linking material preparation and formulation with coating processes, structure development, and final performance will be essential for translating promising laboratory systems into practical and commercially viable PFAS-free food-packaging technologies.

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Abbreviations

The following abbreviations are used in this manuscript:

CA	Contact angle
CMF	Cellulose microfibrils
CNC	Cellulose nanocrystals
CNF	Cellulose nanofibrils
PHAs	Polyhydroxyalkanoates
PLA	Polylactic acid
PVA	Polyvinyl acid
PFAS	Per- and polyfluoroalkyl substances
OTR	Oxygen transmission rate
WVTR	Water-vapor transmission rate

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