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Functional multilayer films with nanocellulose core and plastic outer layers for high-performance oxygen and water vapor barrier food packaging

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ABSTRACT

Background: The increasing demand for sustainable, high-performance packaging has driven interest in multilayer films combining a cellulose nanomaterial (CNM) core layer with plastic outer layers. In these designs, the polar CNM core layer provides effective oxygen barrier properties, while non-polar plastics serve as protective layers against water vapor. This complementary structure enables improved recyclability and supports the development of circular packaging solutions.

Scope and approach: This review overview recent advances in preparation strategies, material interactions, and performance optimization of CNM-plastic multilayer films for packaging. Fabrication techniques, particularly lamination methods such as hot-roll, hot-pressing/thermo-compression, and extrusion, along with various coating methods, are reviewed regarding barrier performance, industrial scalability and recycling implications. Although such technologies are well established in traditional multi-plastic multilayer films, their transferability and limitations in CNM–plastic multilayer architectures have not been systematically analyzed. Moreover, the influence of CNM types, plastic substrates, and tie-layer adhesives on oxygen and water vapor permeability are discussed, particularly under high relative humidity, a known limitation of CNM single films.

Key findings and conclusions: This multilayer film provides excellent oxygen barrier and water vapor even at high humidity. Importantly, CNM core and plastic outer layers can be separated at lab scale by soaking in water, supporting recyclability and material reuse. Finally, challenges related to moisture sensitivity, scalability and standardization are identified, offering insights and recommendations for future research and commercial development. A deeper understanding of moisture equilibrium in interior CNM layers between water vapor barriers and interlayer adhesion is needed.

1. Introduction

Food packaging serves to protect food against environmental contamination to prolong its shelf life and reduce food waste (Jahangiri et al., 2024). To achieve this, packaging materials must exhibit excellent gas barrier properties, particularly against oxygen and water vapor (Adak et al., 2025; Karkhanis et al., 2021; Natarajan et al., 2022; Wongthanaroj et al., 2022; Yue et al., 2024). Even small amounts of oxygen can trigger oxidation reactions that degrade the quality and impact nutritional value, color, flavor, and texture (Xiao et al., 2025). Similarly, effective humidity control is essential, as moisture loss can cause dryness and texture deterioration, and moisture gain can promote microbial growth and spoilage (Zardetto et al., 2025). As a result, numerous businesses and academic researchers place a high value on research into packaging materials with excellent oxygen and water vapor barrier properties (Chen, Wang, et al., 2024; Chen, Xiao, et al., 2024; Qiao et al., 2025).

1.1. Challenges of conventional multilayer packaging

Multilayer packaging is a widely adopted strategy to enhance the overall functionality of flexible films by assigning specific roles to individual layers, thereby combining properties that cannot be achieved using a single material (Alias et al., 2022; Chen, Wang, et al., 2024). For example, polar polymers provide effective oxygen barriers due to oxygen transport is mainly governed by the solution–diffusion mechanism,

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where permeability equals the product of solubility and diffusivity. While polymer polarity can reduce the solubility of non-polar oxygen molecules, factors such as crystallinity, chain packing density, and restricted molecular mobility typically play a dominant role in limiting oxygen diffusion and overall permeability (Elfaleh et al., 2023). However, they typically perform poorly as water vapor barriers. Pairing polar layers with non-polar polymers in a multilayer configuration results in well-balanced, high-performance packaging. These multilayer films offer excellent gas and moisture barriers, improved mechanical strength, and sealing performance, driving increasing interest in food packaging applications (Alias et al., 2022; Barone et al., 2025).

The utilization of fully biopolymer-based food packaging supports sustainability goals and enhances food safety by offering a renewable, biodegradable alternative to traditional synthetic polymers (Elfaleh et al., 2023; Mathew et al., 2025; Stark & Matuana, 2021; Tyagi et al., 2021). Currently, there are several commercial offerings for fully biodegradable food packaging systems with barrier properties. Some examples of these include the compostable products lines from TIPA® (https://tipa-corp.com/), the NatureFlex™ cellulose based product line offered by Futamura (https://www.natureflex.com/), and the NATIVIA® range of polyhydroxyalkanoates (PHA) films offered by Taghleef Industries (https://www.ti-films.com/brands/nativia/). However, bio-derived natural polymers such as polylactic acid (PLA), PHA, polybutylene Succinate (PBS), polycaprolactone (PCL), etc. in their current form, tend to exhibit less effective barrier and mechanical properties compared to synthetics (Shah et al., 2024). Therefore, synthetic polymers such as polypropylene (PP), polyethylene (PE), polyamide (PA), poly(vinylidene chloride) (PVDC), polyethylene terephthalate (PET), ethylene vinyl alcohol copolymer (EVOH), and others remain dominant in packaging (Yue et al., 2024). Coincidentally, these plastics degrade exceedingly slowly due to their stable chemical structure, contributing to over 400 million metric tonnes of plastic debris every year.

Multilayer films constitute a significant portion of plastic packaging waste and present recycling challenges because their composite structures hinder material separation and mechanical recovery (Abdelwahab et al., 2025). Most synthetic polymer-based multilayer films, which often incorporate materials like EVOH or aluminum with those synthetic polymers, are not environmentally sustainable and difficult to recycle (hard to separate layers). While aluminum foil and metallized coatings provide excellent barrier properties attributable to their crystalline structures and low porosity (Wang et al., 2018), they make laminated films non-recyclable, and there is large, embodied carbon in aluminum production (Yang et al., 2025). Ceramic (SiOx and AlOx)-coated polymers have been widely explored as high-barrier alternatives due to their excellent oxygen barrier performance. However, conventional inorganic oxide coatings can suffer from defects such as pinholes and microcracks, as well as brittleness under mechanical stress. Recent advances in crack-resistant AlOx coatings and hybrid inorganic–organic multilayer structures have significantly improved flexibility and durability, although challenges related to large-area defect control and long-term reliability remain (Sharbafian et al., 2024; Struller, 2013; Wang et al., 2016).

Polyvinyl alcohol (PVOH or PVA) and its ethylene copolymer version, EVOH, remain widely used for their cost-effectiveness, availability, film-forming ability, and excellent oxygen barrier properties, especially in dry or protected multilayer systems (Macnamara Jr, 2024). There is a growing interest in bio-based alternatives due to the environmental concerns surrounding PVOH, such as the questions on the impacts of having large amounts of water-soluble PVOH in our waterways (Alonso-López et al., 2021; Rolsky & Kelkar, 2021). In addition, as fossil-derived polymers, their life cycle greenhouse gas emissions depend on feedstock origin, processing energy, and end-of-life pathways, which may limit their alignment with circular material strategies. Without considerable reduction in usage and increased recycling, the world faces severe challenges, including increased greenhouse gas (GHG) emissions and growing microplastic pollution. These limitations highlight the urgent need for biodegradable, bio-based materials with excellent oxygen barrier properties to enable truly sustainable multilayer packaging solutions.

1.2. Nanocellulose as an oxygen barrier layer

Cellulose nanomaterials (CNMs) are gaining significant attention as oxygen barrier layers in multilayer film systems because they exhibit oxygen barrier properties equal to or better than those of synthetic materials like EVOH and PVOH at low relative humidity. Meanwhile, they are fully biodegradable (in both natural or landfill environmental conditions) and exhibit a lower carbon footprint relative to non-biodegradable synthetic materials. CNMs can be isolated from natural plant fibers through degradation or synthesized by bacterial fermentation. Plant-derived nanocellulose can be classified into three categories: cellulose nanocrystals (CNCs) and cellulose nanofibers (CNFs) and cellulose microfibrils (CMF). CNCs are short, rod-shaped nano-sized fibers with a diameter ranging from 3 to 35 nm, a length between 200 and 500 nm and aspect ratio 11 to 67, produced via acid hydrolysis (Jonoobi et al., 2015). On the other hand, CNF and CMF are flexible, long, intertwined fibers with nanoscale diameters, often used interchangeably in literature. However, according to a recent standard terminology for cellulose nanomaterials based on morphology (TAPPI Standard WI 3021), CNFs are defined by widths of 5–30 nm and aspect ratio exceeding 50, while CMFs possess widths between 10 and 100 nm and lengths ranging from 0.5 to 50 μm, both produced through mechanical treatment of lignocellulosic biomass (Qua et al., 2011). CNFs produced via TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl) mediated oxidation or TEMPO CNFs have also drawn attention due to rich carboxyl groups introduced by TEMPO mediated oxidization, which enable further functionalization. Another type of nanocellulose, known as bacterial nanocellulose (BNC) with a diameter of 2–4 nm, can be produced by fermentation processes using specific bacterial strains (Shan et al., 2026).

CNM-based individual films and coated papers have shown excellent oxygen barrier performance, particularly under low-humidity conditions owing to their high aspect ratio, dense hydrogen-bonded network, and intrinsic crystallinity. To date, CNCs and CNFs have been the primary focus on packaging research, with numerous studies comparing their oxygen and moisture barrier properties. In contrast, BNC has received comparatively limited attention in multilayer packaging applications, and systematic comparisons with plant-derived CNMs remain scarce. This is largely attributed to challenges related to large-scale production, higher manufacturing costs, and processing limitations, which restrict its practical implementation in cost-sensitive packaging systems despite its high purity and crystallinity. Typically, self-standing CNC films possess a dense and highly crystalline structure, which contributes to their excellent oxygen-barrier performance under low-humidity conditions. However, CNC films tend to be brittle due to their rigid rod-like morphology and limited network entanglement. In contrast, CNFs have a high aspect ratio and an inherent ability to form entangled and interconnected networks, which enhances mechanical flexibility and structural integrity. As a result, CNF-based films generally exhibit better mechanical properties and improved oxygen-barrier performance under high-humidity conditions compared to CNC films (Las-Casas & Arantes, 2024). Nevertheless, owing to the intrinsically hydrophilic nature of cellulose, oxygen permeability increases sharply when the relative humidity exceeds 65 %, as moisture-induced swelling (exacerbated by clustering and capillary effects) disrupts the hydrogen-bonded network structure (Wang et al., 2018). Therefore, efficient strategies to mitigate moisture sensitivity in CNM-based films are essential for practical multilayer packaging applications.

1.3. CNM-plastic multilayer films for high-barrier performance packaging

Multilayer systems with CNM as core and plastic films as out layer

have been proposed to the most promising strategies to mitigate CNF oxygen barrier moisture sensitive challenge, pairing strong oxygen barrier properties with the moisture resistance (Chen, Wang, et al., 2024; Melendez-Rodriguez et al., 2021; Wang et al., 2020; Yu et al., 2022). In this system, the unique hydrophilic nature of CNMs may enable water-assisted delamination, as swelling of the CNM layer can weaken interfacial bonding with adjacent plastic layers and facilitate separation (Chen, Wang, et al., 2024; Ji et al., 2023). However, this mechanism has mainly been demonstrated under laboratory conditions and may be influenced in practice by adhesive residues, tie-layer chemistry, and industrial recycling parameters. Additionally, paper packaging offers stiffness, printability, and light barrier properties while also high recycling rate (Bauer et al., 2021). When coated with nanocellulose and then laminated with plastic films, paper-based materials may gain enhanced oxygen and water vapor barrier. Importantly, these layers could potentially be separated and recycled, aligning with circular economy goals. Therefore, multilayer film employing CNMs as core and commercial plastic films as out layer by directly laminating or coating CNM on paper substrates then laminating with plastic film provides a potential practical pathway toward commercialization and industrial adoption of environmentally friendly, high-performance multilayer packaging.

1.4. The scope of this review

Despite growing research in utilization of CNMs as an oxygen barrier, a comprehensive review that summarizes fabrication techniques, including lamination methods such as hot-roll, hot-pressing, extrusion, and thermo-compression, along with various coating methods, and critically examined regarding barrier performance, while linking these developments to challenges in industrial-scale implementation and recyclability is still lacking. Although such technologies are well established in traditional multi-plastic multilayer films, the practical challenges, necessary modifications, and limitations associated with integrating moisture-sensitive CNM layers into plastic multilayer architectures or transition from conventional multi-plastic multilayer film to CNM-plastic multilayers have not been systematically analyzed. Accordingly, this review provides a brief overview of preparation technologies, the oxygen and water vapor barrier properties of CNM-plastic multilayer packaging materials (focusing on protection of CNM layer at high humidity), and their role in contributing to sustainability. By doing this, we will compile insights into the benefits and challenges of CNM-plastic multilayer food packaging to guide future research to facilitate a transition towards a sustainable and circular economy. Achieving this requires increasing awareness of what constitutes recyclable packaging, as well as understanding the potential negative effects that may arise when redesigning circularity to improve the sustainability of specific product packaging while maintaining required performance.

2. Multilayer film preparation strategies

Generally, the production of multilayer packaging films primarily relies on extrusion or lamination processes. While extrusion or coextrusion is commonly used to combine materials with similar processing conditions, such as melting point and viscosity, lamination is essential for combining polar polymers with non-polar polymers (Bauer et al., 2021). In addition to these two primary methods, coatings offer the opportunity to integrate even more beneficial properties into a packaging film. Coatings can form a thin layer of material that is deposited directly onto a surface, and can be applied in liquid form through immersion, homogeneous spinning, spreading, or spraying. For the production of nanocellulose and hydrophobic polymer multilayer films, lamination and different types of coating is typically employed (Jahangiri et al., 2024). For paper based packaging, coating methods are usually used.

However, compared with conventional multi–plastic multilayer films, integrating CNM introduces additional processing requirements. CNMs are typically processed as aqueous dispersions with low solids content and relatively high viscosity due to strong hydrogen bonding and entangled fibrillar networks (Jonoobi et al., 2015; Wang et al., 2018). These characteristics influence coating uniformity, drying energy demand, interfacial adhesion, and defect tolerance. In addition, CNMs cannot be directly melt-processed because of their thermal sensitivity and hydrophilic nature, which limits compatibility with conventional extrusion-based multilayer fabrication (Bauer et al., 2021; Wang et al., 2018). Therefore, each preparation strategy must be evaluated not only in terms of barrier performance but also in terms of processing window, moisture control, adhesion reliability, scalability, and recyclability implications (Bauer et al., 2021; Tamizhdurai et al., 2024). The key advantages and limitations of lamination and coating approaches with regard to these factors are summarized in Table 1.

2.1. Lamination

Lamination has proven to be an effective strategy for producing multilayer films incorporating CNM layers as the core and plastic films as outer protective layers for food packaging applications (Chen, Wang, et al., 2024; Le Gars et al., 2020; Wang et al., 2018). In this configuration, the inner CNM layer is protected by the outer plastic layers from moisture and environmental degradation (*e.g.,* mechanical or chemical). Laminating plastic films with CNM films, either with or without an adhesive tie layer—significantly reduces both oxygen and water vapor transmission rates (Chen, Wang, et al., 2024). Commonly used lamination techniques include hot-roll lamination (Fig. 1 A), hot and pressure lamination (Fig. 1 B), which include hot-pressing lamination **(a)** and thermo-compression lamination **(b)**, and extrusion lamination (Fig. 1 C).

However, multilayer performance strongly depends on interfacial integrity. In practice, imperfect adhesion, microvoids, incomplete wetting, and thickness non-uniformity may create preferential diffusion pathways for oxygen and water vapor, reducing barrier performance below ideal values. In addition, residual stress generated during lamination or cooling may induce interfacial debonding or microcracking under mechanical loading or thermal cycling. Long-term exposure to elevated relative humidity may also promote CNM swelling, interfacial fatigue, or gradual delamination, particularly when adhesive layers are not optimally designed. Therefore, interfacial compatibility between hydrophilic CNMs and hydrophobic plastics often necessitates surface treatment or adhesive tie layers (Huang et al., 2003; Saremi et al., 2020).

From a processing perspective, successful lamination requires careful control of temperature (generally 80-170 °C depending on polymer), pressure (0.2-0.6 MPa typical in reported CNM laminations), residence time, and the moisture content of the CNM layer prior to lamination. Residual moisture in CNM films may generate interfacial voids or weaken adhesion during heating (Wang et al., 2018).

2.1.1. Hot-roll lamination

Hot-roll lamination is a scalable and industry-relevant method for producing CNM-plastic multilayer films. Chen, Wang, et al. (2024) fabricated tri-layer polylactic acid (PLA)/CNC/PLA films using a hot-roll laminator at 90 °C, 0.276 MPa and 50 cm/min (Fig. 2 (A)). To improve compatibility and processability, CNCs were modified by nonionic, polyvinyl alcohol (PVA), and anionic kappa-carrageenan (CG). These films exhibited 70 times lower oxygen permeability than pure PLA film (13,515 g μm/(m^2·day·KPa)), and 7 times lower water vapor permeability than the pure CNC (1000 cm^3 μm/(m^2·day·atm)) film at 50 % RH. Wang et al. (2020) laminated CNC or CNF films with biaxial oriented polypropylene (BOPP) film using a polyurethane (PU) adhesive at 90 °C, 0.55 MPa, and a lamination speed of 60 cm/min (Fig. 2 (B)). The resulting BOPP/PU/CNC/PU/BOPP or BOPP/PU/CNF/PU/BOPP multilayer films had oxygen transmission permeability (OP) of 40 (CNC) and 24 (CNF) times lower than the BOPP/PU/BOPP films (376.4 cm^3

Table 1
Comparative analysis of preparation strategies for CNM–plastic multilayer films.

Manufacturing Strategy	Typical Configuration	Key Advantages	Main Limitations & Commercial Bottlenecks	Recyclability Implications
Hot-roll lamination	Pre-formed CNM film laminated between polymer films	• Compatible with existing roll-to-roll lines • Continuous processing • Good thickness control (Alias et al., 2022; Bauer et al., 2021)	• Requires adhesive/tie layer • Interfacial defect sensitivity • Adhesion reliability under high RH • Delamination vs durability trade-off (Fraldi et al., 2014; Wang et al., 2018)	• Adhesive layers (*e.g.*, PU, AR) may hinder mono-material recycling; controlled delamination possible if designed intentionally (Bauer et al., 2021; Ji et al., 2023; Tamizhdurai et al., 2024)
Hot-pressing/Thermo-compression	CNM film compressed between thermoplastic layers under heat and pressure	• Strong interfacial bonding • Reduced voids • Good lab-scale control Fitoussi et al. (2022)	• Batch process, limited scalability • Energy intensive • Risk of CNM thermal degradation • Not fully compatible with roll-to-roll manufacturing (Fitoussi et al., 2022; Fraldi et al., 2014)	• Often requires tie layers; crosslinked adhesives may reduce recyclability and compostability (Bauer et al., 2021; Tamizhdurai et al., 2024)
Co-extrusion (with CNM insert or hybrid design)	Polymer melt processing with CNM layer incorporated as insert or hybrid coating	• High industrial scalability • Continuous melting processing • Uniform thickness control (Azzaz et al., 2025; Huang et al., 2003)	• CNM cannot be melt-processed directly • Thermal sensitivity of CNM • Polar/non-polar incompatibility • Narrow process window (Bauer et al., 2021; Tamizhdurai et al., 2024)	• Multi-material structures complicate mechanical recycling; compatibilizers affect recyclate purity (Bauer et al., 2021; Tamizhdurai et al., 2024)
Solution coating (bar, slot-die, gravure)	CNM dispersion coated onto plastic or paper substrate	• Precise thin-layer control (<5 μm possible) • Adaptable to existing coating lines • Low material consumption (Saremi et al., 2020; Wang et al., 2018)	• High viscosity at low solids limits coating speed • High drying energy demand • Defect sensitivity (pinholes, cracking) • Moisture sensitivity during storage (Fotie et al., 2017; Saremi et al., 2020; Wang et al., 2018)	• Easier to design delamination pathways; however, added tie layers or surface treatments may limit recyclability (Saremi et al., 2020; Yang et al., 2024)

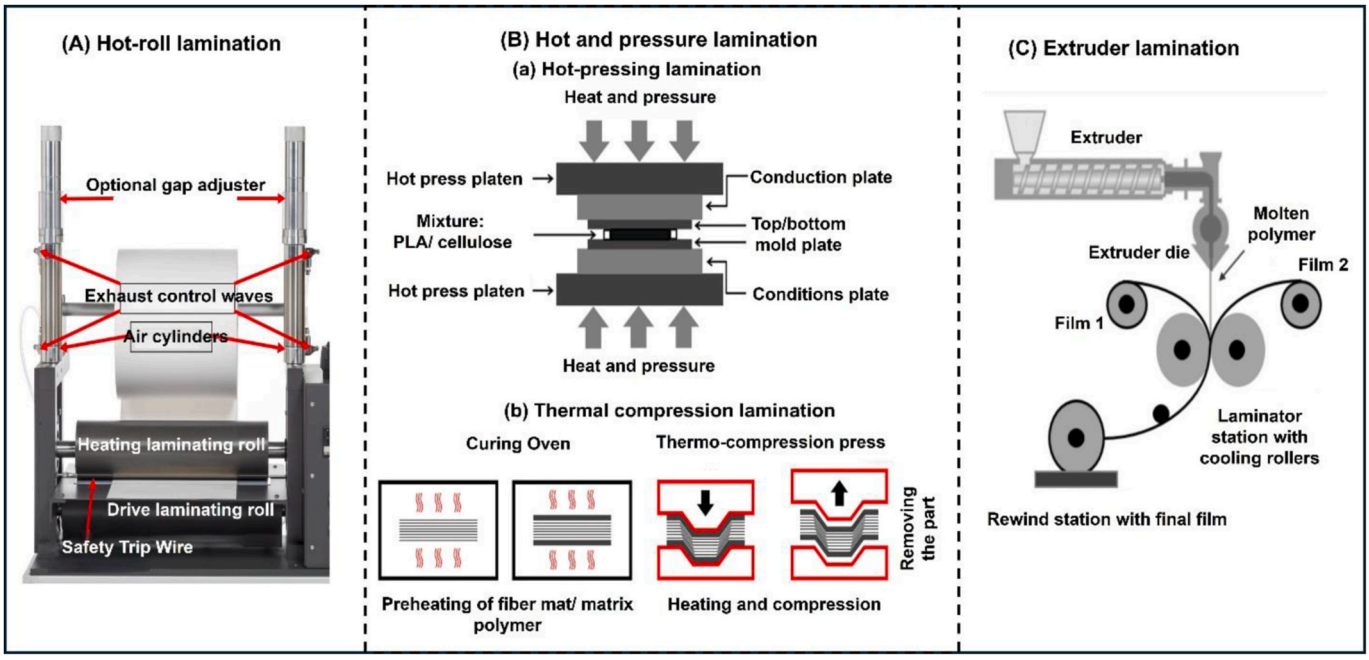

Fig. 1. Mechanical schematic of hot roll laminator **(A)** (https://www.cheminstruments.com/content/media/wysiwyg/HLManual_v10_1.pdf), hot and pressure lamination **(B)** (including hot-pressing lamination **(a)** (Penjumras et al., 2016) and thermal-compression model **(b)** (Fitoussi et al., 2022)), and extrusion lamination **(C)** (Azevedo et al., 2022). Reuse permission was obtained from each cited reference.

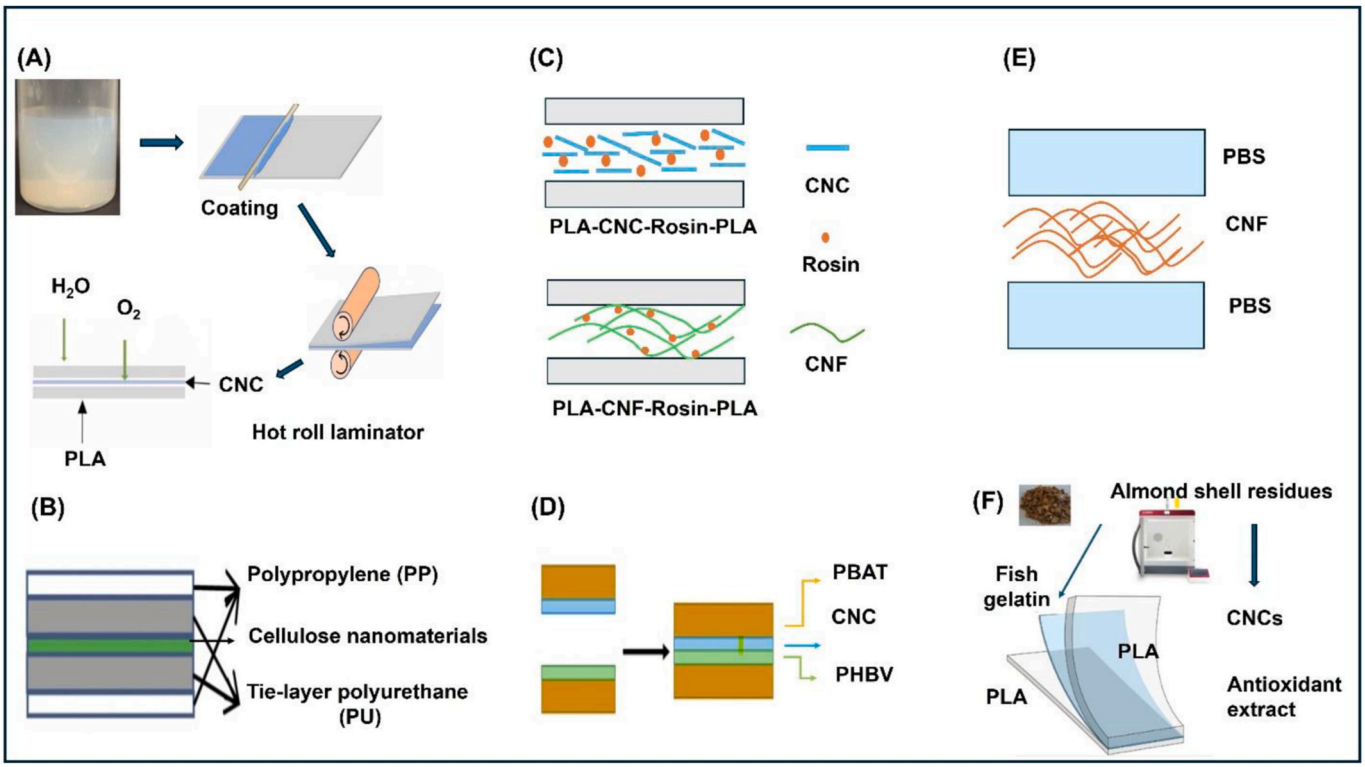

Fig. 2. The multilayer nanocellulose-polymer based film made by Hot roll laminator **(A)** (Chen et al., 2024) and **(B)** (Wang et al., 2020), hot-pressing. **(C)** (Le Gars et al., 2020) and thermal compression **(D)** (Melendez-Rodriguez et al., 2024), **(E)** (Melendez-Rodriguez et al., 2021) and **(F)** (Valdés et al., 2021).

μm/(m^2·day·atm)) and were 9.2-21.1 (CNC) and 2.3-6.2 (CNF) times lower than that of the neat CNC (19.3 cm^3 μm/(m^2·day·KPa)) and CNF (13.4 cm^3 μm/(m^2·day·KPa)) films, respectively. The water vapor permeability (WVP) decreased by over 500 times for both BOPP/-PU/CNC/PU/BOPP or BOPP/PU/CNF/PU/BOPP compared with pure CNC (4741.5 g μm/(m^2·day·KPa) or CNF (5529.5 g μm/(m^2·day·KPa) film. Importantly, the CNMs layers maintained an excellent oxygen barrier at 80 % RH due to the assistance of the PU tier layer. Mechanical strength remained comparable to the control laminates.

Barrier performance under high humidity (80 % RH) indicates that lamination alone is insufficient; interfacial sealing and adhesive chemistry play decisive roles in moisture resistance. Therefore, hot-roll lamination performance depends not only on temperature and pressure but also on: CNM film density and porosity, adhesive curing kinetics, line speed versus bonding time, long-term moisture equilibration.

2.1.2. *Hot-pressing/thermo-compression*

Discontinuous heat and pressure laminating techniques include hot-pressing and thermo-compression, both of which apply heat and pressure to promote polymer bonding between layers. In practice, hot-pressing and thermo-compression are often used interchangeably to describe this process, typically involving the use of heated platens to achieve consolidation and bonding. A pre-curing step (*e.g.*, oven drying) is not inherently required for this process but may be employed depending on the specific material system or processing conditions (Fig. 1 (B) **a** and **b**). Processing temperatures reported for CNM multilayers range from 110 to 170 °C, with pressures between 0.5 and 4 MPa and dwell times from 20 s to 10 min (Le Gars et al., 2020; Melendez--Rodriguez et al., 2024; Valdés et al., 2021). Le Gars et al. (2020) produced three-layer films by hot-pressing dry rosin modified CNF or CNC films between two PLA sheets at 170 °C and 0.5 MPa for 10 min (Fig. 2 (C)). The oxygen permeability decreased comprised between 84 and 96 % and between 44 and 50 % for neat nanocelluloses and nanocelluloses with rosins as the inner layer at 23 °C and 50 % RH, respectively. Films containing rosins also showed ~20 % radical scavenging activity. Similarly, Melendez-Rodriguez et al. (2024) laminated poly(butylene adipate-*co*-terephthalate) (PBAT)/CNC/Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) multilayer films at 140 °C for 20 s at a speed of 5 m/min using a Reliant Powerbond device (thermo-compression), achieving a 91.5 % OTR reduction at 0 % RH compared with without CNC one. The same group later prepared PHBV/CNC/3-hydroxyvalerate (HA)/PHBV multilayers with significantly reduced OTR and WVTR (Fig. 2 (D)) (Melendez-Rodriguez et al., 2021). Using a layered spray-coating approach, Nabels-Sneiders et al. (2023) incorporated 1–20 NFC layers between Polybutylene succinate (PBS) films, followed by compression molding at 112 °C for 2 min. A single NFC layer (0.35 wt %) reduced WVTR 5.5-fold, although excessive layering (>10) led to poor adhesion and structural defects. Repeated deposition–drying cycles may lead to drying-induced shrinkage stresses and capillary forces within the nanocellulose network, which can accumulate across layers and promote microcracking or interfacial weakening (Tyagi et al., 2021; Wang et al., 2018). In addition, increasing the number of layers may amplify internal stress accumulation and mismatch in mechanical properties between adjacent layers, reducing interlayer cohesion (Saremi et al., 2020). When hydrogen bonding dominates interfacial interactions, as is common in CNM systems, excessive thickness may also limit effective stress dissipation and increase the likelihood of delamination under mechanical or humidity cycling (Habibi, 2014; Qin et al., 2019). (Fig. 2 (E)). Valdés et al. (2021) fabricated PLA/gelatin-CNC/PLA multilayers via thermo-compression at 110 °C, 4 MPa, and 3 m min^{-1}, without adhesives, achieving a 78.6 % OTR reduction compared to pure PLA (Fig. 2 (F)).

High temperature improves interfacial flow and adhesion but may induce nanocellulose embrittlement or thermal degradation if exposure time is prolonged (Habibi, 2014). Pressure must be sufficient to eliminate interfacial voids, but excessive pressure may cause layer thinning or stress-induced cracking. Although these methods provide precise laboratory control and strong bonding, their batch nature and relatively long

dwell times limit throughput and increase energy consumption compared with continuous roll-to-roll lamination (Bauer et al., 2021).

2.1.3. Co-extrusion

Co-extrusion may be a potential route to produce CNM–plastic multilayer films, but only when the CNM is blended with an extrudable polymer, as CNMs as a single component cannot be processed by co-extrusion alongside another traditional plastics. Due to processing incompatibility, only limited amounts of CNF are likely to be able to be incorporated into one of the co-extrusion phases as co-extrusion typically requires materials with compatible processing conditions, such as similar melting points and viscosities (Bauer et al., 2021). There were no readily available references that examined co-extrusion with a CNM-rich component. An example of utilization of co-extrusion to produce

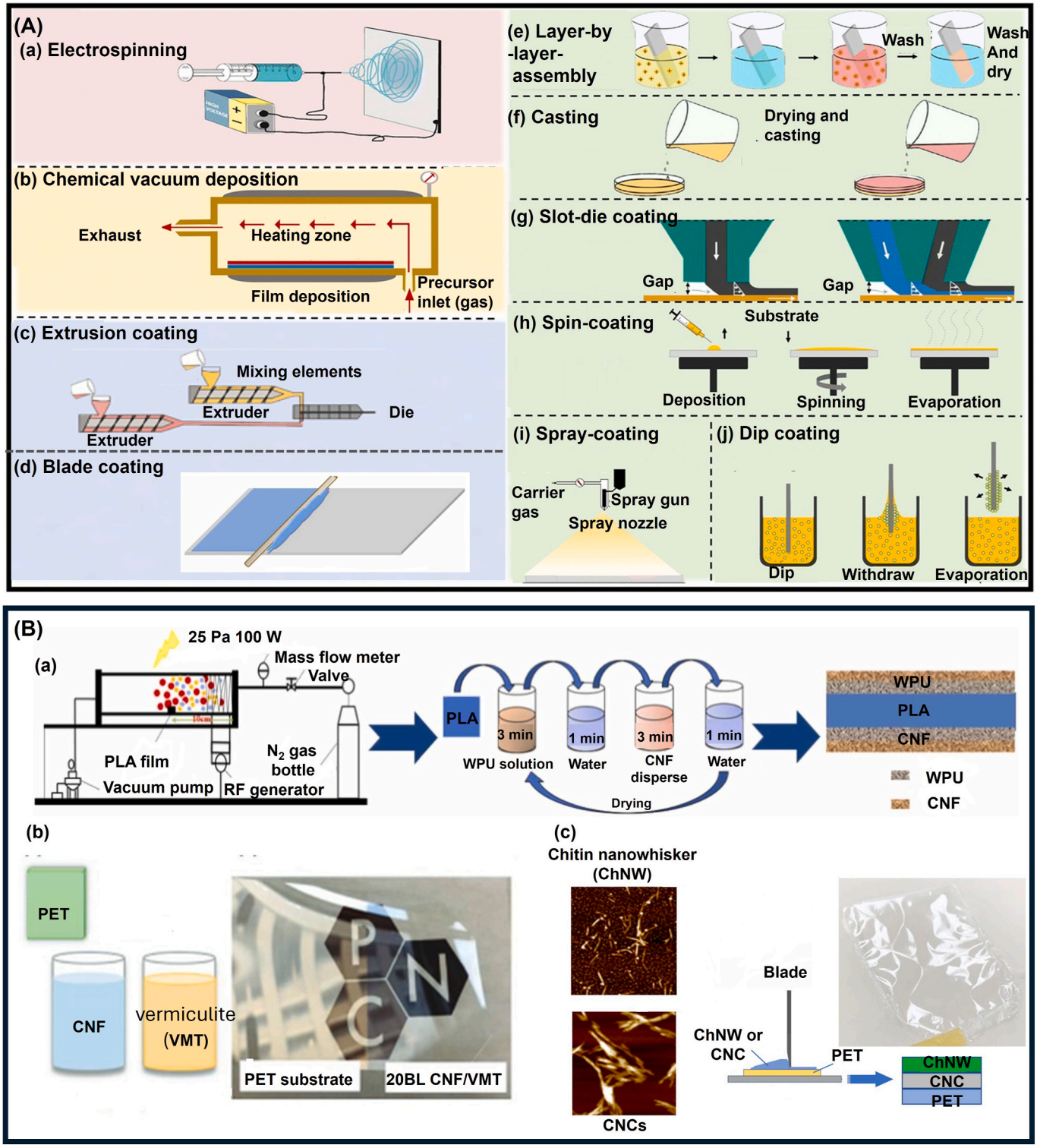

Fig. 3. Different coating techniques for the preparation of single-layer or multilayer coatings **(A)** (Chen, Wang, et al., 2024; Jahangiri et al., 2024). CNM-plastic multilayer structure formation by dip coating **B (a)** (Yang et al., 2024) **and B (b)** (Qin et al., 2019), Dr. blade coating **B (c)** (Yu et al., 2022). Reuse permission received from each cited reference.

multilayer packaging films is the traditional approach laminating EVOH with LDPE. In such structures, EVOH provides an oxygen barrier, and LDPE serves as a water vapor barrier. One interesting study utilizing co-extrusion with cellulose (but not specifically CNMs), is a study by Barros et al. (2025). In the study, the authors propose the potential route to complying with European restrictions on EVOH content in LDPE films by adding nanoclays (NC) and microcrystalline cellulose (MCC) to EVOH. The film with 1.0 wt% CNF achieved OTR reductions of 45.3 % (0 % RH) and 29.0 % (60 % RH). The films were co-extruded at 180–200 °C with a blow-up ratio (BUR) of 1.7, take-up ratio (TUR) of 13.8, and a total thickness of 60–70 μm. This study did not use CNMs but a similar approach to using coextrusion could be taken with CNMs if a co extrudable polymer can be composited with CNMs in a meaningful manner in term of oxygen barrier reduction.

2.2. Coating

Coating processes provide more flexibility for integrating various layered structures and properties into packaging films. There are various coating techniques including vacuum deposition, solution coating (*e.g.*, layer-by-layer assembly, slot-die coating, brush/bar/blade coating, spray coating, spin coating and dipping), electrohydrodynamic processing (*e.g.*, electrospraying, electrospinning), and melt extrusion coating (Fig. 3 (A)) (Chen, Wang, et al., 2024; Jahangiri et al., 2024). Among them, blade, slot die, or spray coating are the most scalable. Layer-by-layer approaches (Qin et al., 2019; Yang et al., 2024; Yu et al., 2022) require multiple deposition–drying cycles, which may limit industrial throughput unless adapted to high-speed automated systems.

Coating involve applying thin layers onto one or both sides of bio-based or conventional materials such as paper or paperboard. Compared with lamination, coating are more versatile in controlling multilayer thicknesses, typically ranging from 100 nm to several micrometers depending on solids content and coating gap settings (Tyagi et al., 2021). CNM dispersions for coating commonly contain 0.5–3 wt.% solids; higher solids (>4–5 wt.%) significantly increase viscosity and may limit slot-die or blade coating speeds to below 10–50 m/min depending on rheology (Jonoobi et al., 2015; Wang et al., 2018). Drying is a critical step. Water evaporation requires significant energy, and drying temperatures of 60–120 °C are commonly used in laboratory studies, while industrial IR or hot-air dryers enable shorter residence times (seconds rather than minutes) (Fotie et al., 2017; Tyagi et al., 2021). Rapid drying may induce cracking due to capillary stresses; controlled humidity drying may reduce defect formation.

Before coating, remote plasma treatment is often used to improve the surface adhesion of nanocellulose materials. Remote plasma treatment is a surface functionalization technique that can introduce oxygen-containing groups (*e.g.*, –OH, –COOH) or other functionalities onto plastic substrate surfaces, increasing surface energy and improving interfacial adhesion with CNMs. Lu et al. (2018) reported the use of cold plasma treatment on biaxially oriented polypropylene (BOPP)/low-density polyethylene (LDPE) films to enhance the attachment of CNF coatings and improve oxygen-barrier performance. Pre-treating the plastic film with cold plasma increased surface hydrophilicity and facilitated better CNF coating adhesion. CNF-coated films on plasma-treated BOPP/LDPE showed significantly lower oxygen transmission rates (OTR) compared to uncoated films, demonstrating how plasma surface activation improves CNM–plastic multilayer interactions and barrier properties.

Coating methods have been applied to construct multilayer nanocellulose-based films. Yang et al. (2024) assembled alternating layers of cationic waterborne polyurethane (WPU) and carbonylated CNF on plasma-modified PLA films, achieving 99.5 % and 63.2 % reductions in pressure-normalized OTR (permeance) (from 695 $cm^3/(m^2 \cdot day \cdot bar)$ to 3.15 $cm^3/(m^2 \cdot day \cdot bar)$) and WVTR (from 150.50 $g/(m^2 \cdot day)$ to 55.41 $g/(m^2 \cdot day)$) at RH 51 %, respectively, with 12 layers with thinness about (106 μm transverse and 108 μm longitudinal) (Fig. 3 B (a)). Qin et al. (2019) developed CNF/vermiculite (VMT) nanocoating through layer-by-layer deposition; a 20-bilayer film (136 nm thickness) formed a "nanobrick wall" structure with high transparency, flame resistance, and an OTR of 0.013 $cm^3/(m^2 \cdot day)$ at 50 % RH (Fig. 3 B (b)). Yu et al. (2022) used doctor-blade coating to layer chitin nanowhiskers (ChNWs) with chitosan (CS) (27 μm), and CNCs (26 μm) on PET via electrostatic and hydrogen bonding, yielding excellent oxygen barrier performance (OTR 21.8 $cm^3/(m^2 \cdot day)$) even under 80 % RH (Fig. 3 B (c)). For a sandwich multilayer system with CNMs in the center, the most likely method of large-scale manufacturing would be to take a polymer film and coat (blade, slot die, spray) the CNMs onto said film as a suspension and dry the coating, then proceed to hot roll laminate another polymer layer onto the other side of the CNM layer in a roll-to-roll fashion. The most difficult aspect is drying the CNM layer at a high rate. CNM suspensions have very high viscosities and therefore typically must be applied as a low solid (less than 10 %) suspension to be coated. This means that significant water must be removed from the coating via evaporation. Even the academic studies that aim to move towards a continuous process still only coat at line speeds up to 3 m/min. In order to make packaging materials cost-effective and competitive with existing solutions, they must be processed at significantly higher rates (hundreds of m/min). Therefore, significant improvements in this area are needed. This may come in the form of modifying CNMs to allow them to dry faster, additives to reduce viscosities at high solids, or even new methods of drying.

3. Water and oxygen barrier properties and food shelf-life applications

Loss or gain of oxygen and water is a major cause of food deterioration (Choskit, 2023). To address this issue, high-quality packaging that prevents moisture and oxygen transmission between products and their environment is essential. Traditional synthetic plastics (polyolefins and polyesters) generally exhibit excellent water vapor barrier but poor oxygen barrier properties (Natarajan et al., 2022; Trinh et al., 2023). Multilayer films with CNM as the core layer for oxygen barrier functionality and plastic outer layers for water resistance represent a promising strategy, providing comparable barrier performance with conventional barrier systems while enabling improved recyclability or reduced material intensity. This configuration combines strengths of both components, providing superior oxygen and water barrier performance, while the hydrophobic outer polymer can help maintain CNM performance even under high humidity. Here, we summarize recent advances in the oxygen and water barrier properties of nanocellulose–plastic polymer multilayer films, emphasizing strategies that overcome CNM limitations in high-humidity conditions. A comparative analysis of reported nanocellulose-polymer multilayer systems is presented in Table 2, noting that performance values vary depending on testing methods, CNM type, outer layer polymers, tie layers, and test conditions. These factors and their effects on gas barrier properties are discussed in the following sections.

3.1. Gas permeation mechanism and standard testing methods

Various standards are used to measure the gas transmission rate (GTR) of polymeric films, but few studies have compared their accuracy or representativeness. As materials and structures evolve, these methods may require modification. A solid understanding of gas permeation mechanisms is essential for developing reliable, standardized tests that accurately evaluate barrier performance under relevant conditions.

3.1.1. Gas permeation mechanisms

The barrier performance of polymeric films is governed by their ability to restrict the transport of small molecules through mass transport mechanisms, primarily permeation (Lagaron et al., 2004). The most widely applied model for dense polymers is the solution–diffusion

Table 2
Oxygen and water barrier properties of CNM-plastic multilayer films.

Fabrication	Multilayer films	Thickness (μm)	OTR (cm^3/(m^2·day)	OP (cm^3·μm/(m^2·day·atm)	OP Test conditions	WVTR (g/m^2·day)	WVP (g·μm/m^2·day·kPa)	WVP Test conditions	References
Lamination	PBAT-PHBV/CNC/PBAT/PHBV	105	4.3772	459.9	OxTran MOCON, ASTM D3985, 23 °C, 0 % RH	0.79	21.8	PERMATRAN-W 398 Mocon,30 °C, 90 % RH	*Melendez-Rodriguez* et al. *(2024)*
	PLA/CNC+15PVA/PLA	82-84	2.2-4.4	183-368	OxTran MOCON, ASTM D3985, 23 °C, 50 % RH	23	1364	ASTM E96/E96M-16, 23 °C, 100 %−50 % RH, 2.81 KPa	*Chen, Wang,* et al. *(2024)*
	PLA/FG/ASE/CNC/PLA	118	346.36	40870	Oxygen permeation analyser, 23 °C, 50 % RH	/	/	/	*Valdés* et al. *(2021)*
	OPP/CNC/PE	106	0.06<	<6.36	Isostatic permeabilimeter, 23 °C, 80 % RH		/	/	*Fotie* et al. *(2020)*
	OPP/CNC/PLA	56	320	17920		/	/	/	
	PET/CNC/PE	88	1.107	97.42		/	/	/	
	PET/CNC/PP	53	0.06<	<6.36		/	/	/	
	PLA/CNC + Rosin/PLA	94	0.08	7.89	Systech Illinois M8001 ASTM-F	0.04	2.51	T448 om-09 TAPPI, 23 °C and	*Le Gars* et al. *(2020)*
	PLA/CNF + Rosin/PLA	88	0.12	1.51	1927 23 °C, 50 % RH	0.04	2.76	50 % RH	
	PP/PU/CNC/PU/PP	74	5.7-6.1	1053.78	Oxtran MOCON, ASTM D3985, 23 °C, 80 % RH	0.9-1.0	82.0-83.5	ASTM E96/E96M-16, Mason jar, 23 °C, 80 % RH, 2.81 Kpa	*Wang* et al. *(2020)*
	PP/PU/CNF/PU/PP	78	10.5-24.3	1904.91-4275.92		0.8-1.0	106.0-111.5		
	bio-HDPE/CNF/bio-LDPE	78	8	624	Oxtran MOCON, ASTM D3985, 23 °C, 50 % RH	1.6	37.8	Permatran-W MOCON, 38 °C; 50 % RH	*Vartiainen* et al. *(2016)*
	bio-HDPE/CNF/bio-LDPE	78	490	38220	OxTran, MOCON, ASTM D3985, 23 °C, 80 % RH	/	/	/	
Coating	CNF/WPU/PLA/WPU/CNF	/	3.15	/	Gas Transmission rate tester GB/T1038, 23.4 °C, 51 % RH	/	/	/	*Yang* et al. *(2024)*
	PET/CNC + ShNW/CS	27	19.13917	516.758	OxTran, MOCON, ASTM D3985, 23 °C 50 % RH	/	/	/	*Yu* et al. *(2022)*
	PET/CNC + ShNW/CS	27	21.76611	587.685	MOCON, ASTM D3985, 23 °C 80 % RH	/	/	/	
	20-layer CNF/VMTclay/PET	179.136	0.013	2.32877	OxTran, MONCON, ASTM D3985, 23 °C, 50 % RH	/	/	/	*Qin* et al. *(2019)*
	CNF/wax/LPChNF	45	3	135	Systech Illinois M8001, 23 °C, 50 % RH	/	/	/	*Pasquier* et al. *(2022)*
	CNF/wax/LPChNF	45	55	2475	Systech Illinois M8001, 23 °C, 80 % RH	/	/	/	
	Stearate C18/CNF/stearate C18	60	40-60	2400.00-3600.00	Oxygen Permeation Analyzer 8011, ASTM, 23 °C, 80 % RH	/	/	/	*Willberg-Keyriläine* et al. *(2017)*
	PET/SCNF/FP	53.5	0.28	14.98	OxTran, MOCON, 23 °C, 0 % RH	/	/	/	*Kim et al. (2020)*
	$(GNPs\text{-}CNC)_7$/PE-coated smart paper	0.0932	100-150	9.32-13.98	Oxygen Headspace Analysis, ASTM F2714-08, 23 °C 50 % RH	/	/	/	*Chemin* et al. *(2019)*
	BOPP/LDPE-nanofibrillated cellulose	76.5	24.02	1837.53	OxTran MOCON, ASTM D3985, 23 °C 50 % RH	/	/	/	*Lu* et al. *(2018)*
	HDPE/TCNF/LDPE	79	2	158	8001 Oxygen Permeation Analyzer, 23 °C, 0 % RH	1.62	60.9	The gravimetric cup method, 23 °C; 75 % RH	*Vähä-Nissi* et al. *(2017)*
	HDPE/TCNF/LDPE	79	400	31600	8001 Oxygen Permeation Analyzer, 23 °C, 80 % RH				

(*continued on next page*)

Table 2 (*continued*)

Fabrication	Multilayer films	Thickness (μm)	OTR (cm^3/(m^2·day)	OP (cm^3·μm/(m^2·day·atm)	OP Test conditions	WVTR (g/m^2·day)	WVP (g·μm/m^2·day·kPa)	WVP Test conditions	References
	CNF/10TEOS-20GPTS	24.1	0.0084<	0.2026<	A coulometric cell Systech 8001, 23 °C, 70 % RH	16.1	117	Electrolytic P2O5 sensor 38 °C, 50 % RH	*Karasu* et al. *(2018)*
	PHBV/LCNF/PHBV	141.5	2.63	372.15	M8001 from Systech Illinois, 25 °C, 60 % RH,	4.0926	183	ASTM 2011 gravimetric method, 25 °C, 100 % RH	*Cherpinski* et al. *(2018)*

Note: the following are full name of abbreviations in Table 2.
MCC: cellulose microcrystals, PBAT: (Polybutylene adipate terephthalate), PHBV: poly(3-hydroxybutyrate-co-3-hydroxyvalerate), CG: kappa-carrageenan, ASE: antioxidant extract, FG: fish gelatin, OPP: Oriented polypropylene, PP: polypropylene, PU: polyurethane, WPU: waterborne polyurethane, ShNW: short chitin nanowhiskers, CS: chitosan, LhNW: Long chitin nanowhiskers, *VMT*: vermiculite, LPChNF: lignin nanoparticles nucleated on chitin nanofibrils, FP: Fluoropolymer, SCNF: succinylated cellulose nanofibers, GNPs: gibbsite nanoplatelets, BOPP: Biaxially oriented polypropylene, CNF/TEOS: tetraethylorthosilicate, GPTS: 3-glycidoxypropyltrimethylenesilane.

mechanism (Fig. 4 A), which involves adsorption of gas molecules at the upstream surface, diffusion through the polymer matrix, and desorption at the downstream side (Las-Casas & Arantes, 2024). Gas permeability under this model depends on polymer properties such as molecular weight, crystallinity, and chain mobility, as well as gas characteristics and environmental conditions including temperature and humidity.

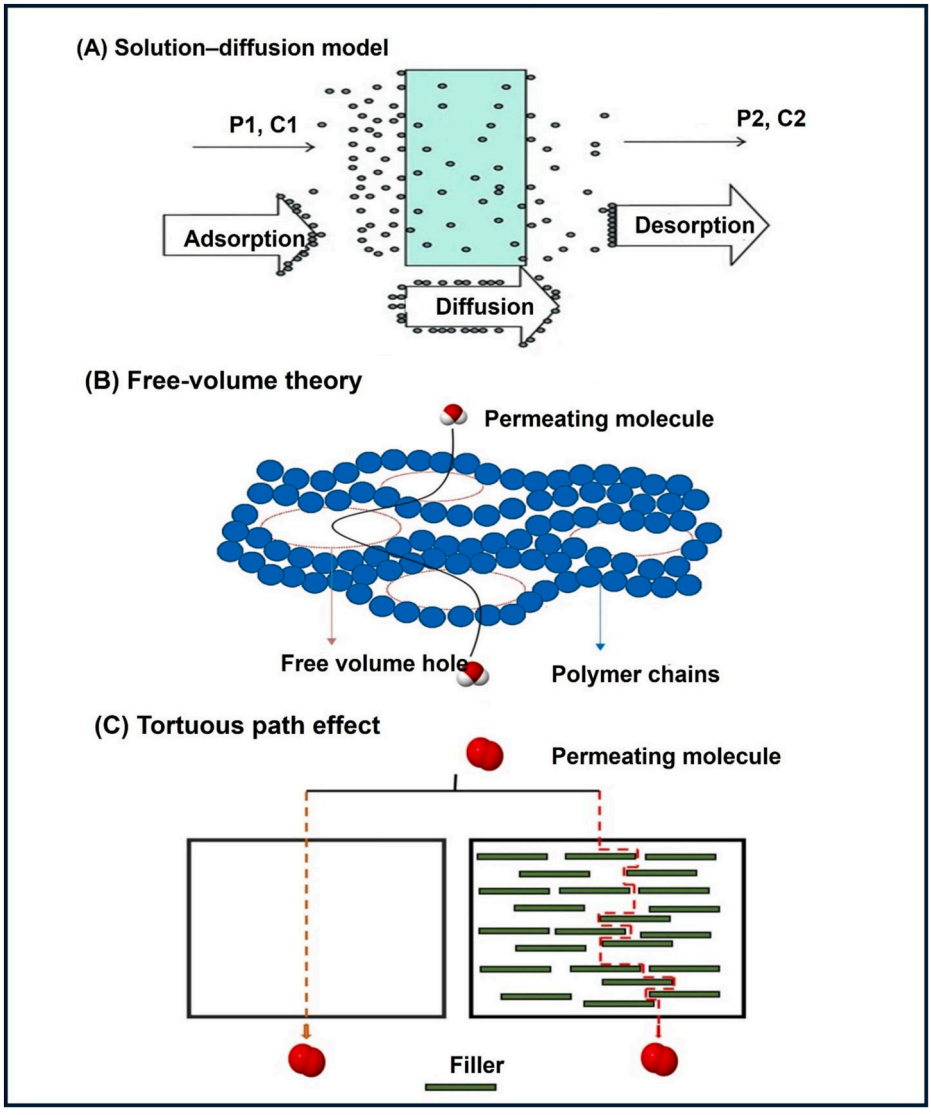

Fig. 4. Illustrative concepts related to gas permeability. **(A)** Overview of the general mechanism of gas permeation through a plastic film (Siracusa, 2012). Here, P1 and P2 represent different permeant pressures, while C1 and C2 denote varying permeant concentrations across the film. **(B)** The diffusion of a permeating molecule through the voids between the polymeric chains, known as free volume (Las-Casas & Arantes, 2024). **(C)** The 'tortuous path' effect resulting from the addition of fillers within a polymeric matrix (Las-Casas & Arantes, 2024). Reuse permissions were obtained from cited references.

Complementary models further describe gas transport in semi-crystalline polymers and nanocomposites. The free volume theory (Fig. 4 B) attributes permeability to transient voids between polymer chains, with permeability increasing exponentially with fractional free volume (Siracusa, 2012); temperature and moisture can enhance chain mobility and thus gas diffusion. The tortuous path mechanism (Fig. 4 C), often invoked in nanocomposites and multilayer systems, describes how well-dispersed, high–aspect ratio fillers can increase diffusion path length and thereby reduce permeability (Natarajan et al., 2022; Karkhanis et al., 2018; Idris et al., 2022). However, CNMs do not universally behave as impermeable fillers. Under elevated relative humidity, swelling of the hydrophilic nanocellulose network can disrupt packing density and reduce effective tortuosity. Moreover, poor dispersion, aggregation, or excessive loading may lead to percolation pathways or microvoid formation that counteract barrier improvements. Therefore, the tortuosity-based barrier enhancement in CNM systems strongly depends on humidity, dispersion quality, and interfacial compatibility.

There are several key aspects of CNMs that work together to give them the ability to perform well as an oxygen barrier. First, cellulose is a polar material due to the large number of hydroxyl groups. This means that the non-polar oxygen molecules will have lower solubility in cellulose. Lower solubility decreases the ability of oxygen to absorb into the cellulose layer. Secondly, hydrogen bonding is the main form of bonding between cellulose chains. Hydrogen bonding is often stronger than other common forms of bonding in polymeric systems such as van der Waals bonding. Lastly, CNMs are characteristic of their highly crystalline domains. Within these crystalline domains, there is very low molecular mobility and free volume. Research has shown that adding CNMs to other polymer systems at low loading levels can increase tortuosity (Karkhanis et al., 2018; Karkhanis et al., 2021) but the decreases in oxygen permeation are not significant enough to make the composite material a useful oxygen barrier (Wang et al., 2018). Therefore, researchers have suggested that the most effective utilization case of CNMs is in a dense layer where they are the primary component and can bond to each other (Wang et al., 2018). This points to the integration approach of creating a CNM layer and then laminating it with other polymers as opposed to compounding CNMs into a polymer matrix.

3.1.2. Standard testing methods

Table 2 summarizes multilayer films incorporating CNMs and their reported OTR and WVTR under various conditions and testing standards. OTR is typically measured using a columbic sensor type system (*e.g.*, MOCON OxTran) following ASTM D3985 (Barros et al., 2025; Melendez-Rodriguez et al., 2024), ASTM F1927 (Fotie et al., 2020), or GB/T1038 (Yang et al., 2024) at 23 °C and 0-80 % RH. Additionally, ISO standards 15105-1 and 15105-2 describe two common methods: differential pressure system (change in pressure on vacuum side opposite a high-pressure oxygen side) or a coulombic sensor system (consumes oxygen in an electrochemical reaction resulting in measurable current). In general, the coulombic sensor type system will have better resolution than a differential pressure method when testing very high oxygen barrier materials. In contrast, WVTR is typically measured using gravimetric cup methods, electrolytic sensors, or modulated infrared sensors (*e.g.*, MOCON Permatran W) adhering to standards such as ASTM E96/E96M (Barros et al., 2025), TAPPI T448-om-09 (Fotie et al., 2020), or ASTM F1249 (Tyuftin et al., 2022), with conditions ranging from 23 °C to 38 °C and relative humidity (RH) up to 100 %. Despite widespread use, limited research compares the accuracy of these methods. Test standards or methods and conditions must align with the intended application conditions of the packaging film to ensure relevant and reproducible results. Additionally, comparing oxygen permeability (OP) across studies is further complicated by variations in testing methods, environmental conditions and inconsistent reporting units. Standardization in testing protocols and data reporting is therefore critical.

An often-overlooked factor in multilayer systems is the moisture equilibrium of the CNM layer under high humidity. Since water vapor can slowly permeate through outer plastic layers, the CNM core may not reach equilibrium within short test times. Without accounting for this, tests conducted over hours may underestimate long-term oxygen permeability relevant to food packaging applications. Future studies should include time-dependent evaluations and clear criteria for moisture equilibrium. Equilibrium of an internal hygroscopic layer is not well understood in scientific literature. How the moisture content within the external hydrophobic layers corresponds to the moisture content within the hygroscopic layer is an area where fundamental understanding is needed. Moisture transfer modeling may be a useful tool in developing this fundamental understanding.

3.2. Types of laminating polymer layers

As listed in Table 2, polymers commonly used for laminating with cellulose nanomaterials (CNMs) in research mainly include polypropylene (PP), polyethylene (PE), polyethylene terephthalate (PET), low-density polyethylene (LDPE), high-density polyethylene (HDPE), and polylactic acid (PLA). The interactions between polymers and CNMs vary across materials (Fotie et al., 2020). Sealable and hydrophobic plastics such as PP, PE, and PET generally create better oxygen barriers when laminated with CNMs under 50 % and 80 % relative humidity (RH) compared to PLA, which is more moisture sensitive.

However, in studies focusing on biopolymer-based laminates, PLA is most often used as the outer layer to produce fully biobased multilayer films. Among biobased and biodegradable polymers, PLA is currently produced at the largest scale at an estimated 476,790 metric tons/year in 2024, making it the most cost-effective option (MarketResearch.com, 2025). PLA is widely used in films, thermoformed containers, and short shelf-life bottles. However, its poor water vapor barrier (Fig. 5 (A)) and brittleness (elongation typically less than 7 %) (Fig. 5 (B)) limit its packaging performance. To overcome these limitations, alternative biodegradable polymers such as poly(butylene succinate) (PBS), poly(butylene succinate-*co*-butylene adipate) (PBSA) or polyhydroxyalkanoate (PHA) are gaining attention. PBS and PBSA exhibit good flexibility, processability, and mechanical properties similar to LDPE, HDPE, PP, but have lower melting points compared to other bioplastics such as PLA and poly(hydroxybutyrate-*co*-3-hydroxyvalerate (PHBV) (Li et al., 2025; Nath et al., 2025). PHAs, a family of polymers including polyhydroxyvalerate (PHV), poly(hydroxybutyrate-*co*-3-hydroxyvalerate (PHVB), generally show greater ductility, improved water vapor barrier properties and full compostability compared to PLA (Naser et al., 2021). The main limitation of PHAs remains their production scalability, as current manufacturing capacities make PHAs significantly more expensive than PLA. However, future scale-up could make PHAs viable for CNM-based biodegradable packaging with strong oxygen barrier performance.

Overall, the WVTR and high humidity OTR of the CNM-plastic multilayers depend on the outer-layer's ability to protect the CNM layer from moisture. However, the equilibrium moisture behavior of inner CNM layers and the timescales for moisture equilibration remain poorly understood in laminated multilayer films. Beyond moisture protection, properties such as strength and flexibility are also critical for packaging applications. Future research is needed to evaluate the protective effectiveness of different plastic outer layers when used in CNM-based multilayer structures and to better understand the mechanisms underlying these differences. Particularly, it must be determined if the water vapor barrier properties of specific polymers, namely PLA, are sufficient to protect CNM layers from external humidity. This knowledge is critical for selecting optimal materials for commercial-scale production and ensuring long-term barrier performance in real-world packaging applications.

3.3. Types of CNM layer

Cellulose nanomaterials (CNMs) can be derived from various sources

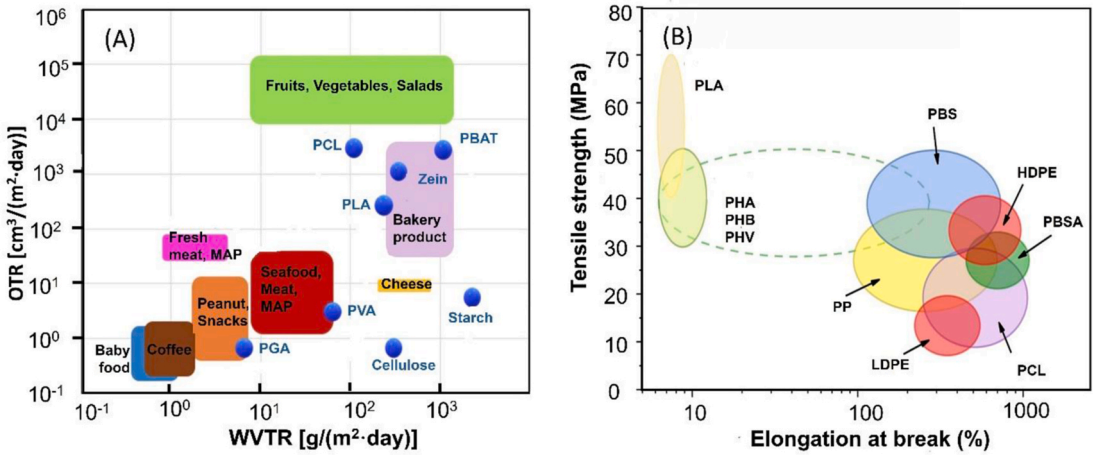

Fig. 5. The barrier properties **(A)** (Trinh et al., 2023) and tensile properties of different biodegradable polyesters and non-biodegradable commodity plastics **(B)** (Reproduced with permission from reference (Platnieks et al., 2021).

using different isolation methods, resulting in materials with distinct aspect ratios, crystallinities, and surface chemistries that significantly influence their performance. Evaluating and comparing these CNM types is therefore essential to identify the most suitable candidate for gas barrier applications in packaging. A recent review by Las-Casas and Arantes summarized the gas barrier properties of CNMs in the form of individual films, reinforcing agents, and coatings under different RH conditions, with particular focus on low RH (Las-Casas & Arantes, 2024). This section compares the oxygen and water vapor barrier performance of CNM individual films with the performance of CNM layers laminated with external polymers at high humidity, aiming to highlight recent research advances, identify remaining challenges, and propose future directions to overcome those challenges, particularly those related to humidity sensitivity.

Du et al. (2021) prepared transparent films with a symmetric sandwich structure using layer-by-layer assembly of biaxially oriented polypropylene (BOPP) and acrylic resin (AR) followed by a cellulose nanoparticle (CNP) layer. The WVTR remained similar across TEMPO-mediated oxidized microcrystalline cellulose (T-MCC is nanoscale, but MCC is microscale), TEMPO-mediated oxidized cellulose nanofibrils (T-CNF), CNC, and TEMPO- mediated oxidized cellulose nanocrystals (T-CNC) at 0.3174, 0.3232, 0.3251, and 0.3252 g/(100 in^2·day), respectively. This indicated that the WVTR was primarily decided by the BOPP layers, which masked the effects of CNM aspect ratio and carboxyl groups. In contrast, the OTR varied significantly (11.83, 18.68, 2.36, and 5.59 cc/100 in^2·day), respectively at 23 °C and 0 % RH, showing that oxygen barrier performance depended mainly on CNM structure and orientation. The CNP layer exhibited smooth and dense cross-sections formed through cooperative orientation and hydrogen bonding during drying. During the drying, CNPs adjusted their orientation cooperatively and drew closer with water evaporation and eventually formed hydrogen bonds to get dense structure for better barrier performance. Moreover, the aspect ratio of CNPs influenced the microstructures of the cellulose layer, displaying a network or parallel structure or a combination of both. These different microstructures showed different tortuous pathways affecting the resultant gas barrier properties (Fig. 6 (A)).

Generally, CNC-based films or coatings exhibit superior gas barrier

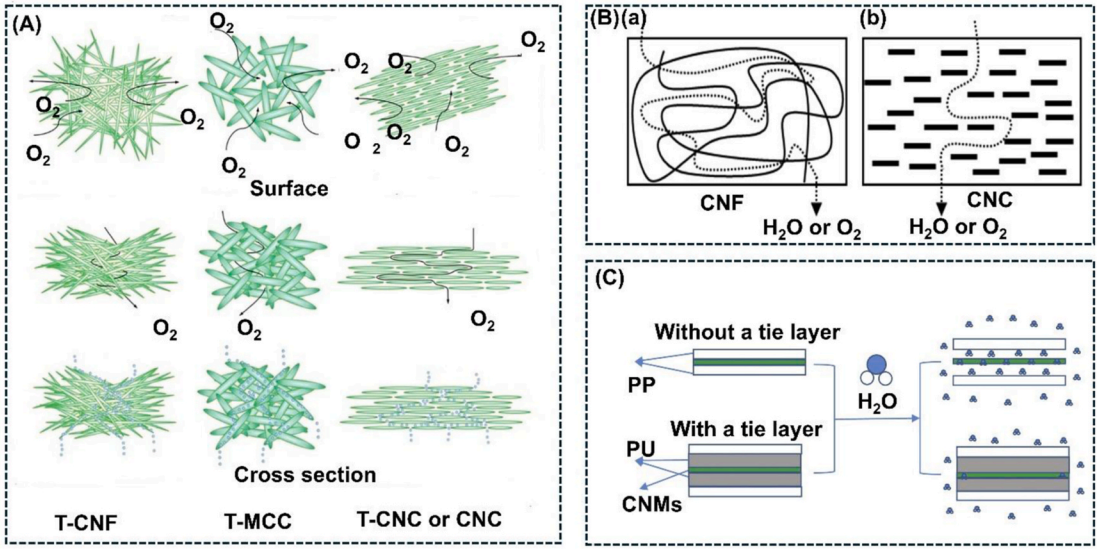

Fig. 6. Schematic representation illustrating the microstructures of CNMs layers of varying morphologies and their influence on the permeation of oxygen and water vapor molecules at 50 % RH **(A)**. Adapted from Du et al. (2021). T-CNF: TEMPO-oxidized cellulose nanofibrils; T-MCC: TEMPO-oxidized microcrystalline cellulose; T-CNC or CNC: TEMPO-oxidized cellulose nanocrystals or cellulose nanocrystals. **(B)** Scheme of a gas passing through CNF (a) and CNC (b) individual or laminated with plastics films via cross sections at high humidity (RH 80 %) (Wang et al., 2020). A scheme explaining how an adhesive layer protects CNM films from being swollen by moisture **(C)** (Wang et al., 2020). Reuse permission was obtained from the cited reference.

performance under low humidity due to their higher crystallinity and density, whereas CNF-based films or coatings perform better under high humidity, attributed to their entangled fibrillar networks creating tortuous diffusion paths. For instance, a study comparing oxygen barrier performance of CNC and MFC-coated PET films found that PET-CNC films exhibited lower oxygen permeability at RH < 42 %, while PET-MFC films perform better at 60–80 % RH (Fotie et al., 2023). This is attributed to MFC's two-phase (amorphous and crystalline) structure which provide flexibility and a tortuous diffusion path for oxygen molecules. Wang et al. (2020) found that at 80 % RH, CNF films exhibited significantly lower OTR and OP (31.3–65.8 cm^3/m^2·day and 1124.7–1357.8 cm^3 μm/(m^2·day·kPa)) than CNC films (55.2–126.4 $cm^3/(m^2$·day) and 1955.6–2026.5 cm^3 μm/(m^2·day·kPa)) (Wang et al., 2020) due to its more tortuous microstructure (Fig. 6 B **(a)** and (b)). However, when laminated with polypropylene (PP) using a polyurethane (PU) adhesive layer, the gas barrier behavior reversed: PP/PU/CNC/PU/PP outperformed PP/PU/CNF/PU/PP. This finding suggests that additives and tie layers can significantly alter gas permeation pathways in laminated structures. Thus, the gas transport mechanisms in individual CNM films and additive-free multilayer laminates seem similar under high humidity, but they diverge when additives are introduced. Further investigation is needed to clarify these mechanisms and their implications for packaging design.

Additionally, due to their nanoscale dimensions and high crystallinity, even thin CNC coatings (~1 μm) can outperform much thicker EVOH or PVDC coatings (~3–4 μm) in oxygen barrier performance, making them especially promising for packaging oxidation-sensitive foods (Wongthanaroj et al., 2022). However, CNCs' hygroscopic nature led to swelling and disruption of their crystalline structure at high RH, which dramatically reduces their barrier performance (Fotie et al., 2017). To address this issue, Fotie et al. successfully introduced ester groups onto a mixture of CNCs extracted from wood pulp by sulfuric acid (CNC_{SO3H}) and cotton linters by ammonium persulfate (CNC_{COOH}) with a 2-to-1 weight ratio through in-situ esterification, creating CNC_{COOR} (Fotie et al., 2020). The resulting lamination of polymers coated with esterified cellulose nanocrystals were the best in terms of reducing the gas permeability, even in higher relative humidity, followed by laminated polymers coated with CNC_{COOH} and CNC_{SO3H}. These modified CNCs, when laminated between water-repellent layers, retained their crystallinity and exhibited significantly improved oxygen barrier performance at 80 % RH compared with unmodified CNCs laminated structures (Table 3). This study demonstrates that chemical modification combined with rational multilayer architectural design can effectively mitigate CNC moisture sensitivity.

Table 3
Effect of in-situ esterification of a 2-to 1 wt ration mixture of wood pulp extracted by sulfuric acid (CNC_{SO3H}) and cotton linters by ammonium persulfate (CNC_{COOH}) on oxygen permeability (OP) of multilayer film formed by CNC laminated with plastic film (Fotie et al., 2020).

Types of CNC in CNC-plastic laminates	OP ($cm^3/(m^2 \times 24h \times bar)$)	
	50 % RH	80 % RH
Unmodified CNC_{SO3H} (acid extracted)		
OPP-CNC_{SO3H}/PE	<0.06	1.2 ± 0.25
OPP-CNC_{SO3H}/PLA	40 ± 0.15	320 ± 3.5
PET-CNC_{SO3H}/PE	0.25 ± 0.15	1.11 ± 0.25
PET-CNC_{SO3H}/PP	<0.06	<0.06
Unmodified CNC_{COOH} (ammonium persulfate extracted)		
OPP-CNC_{COOH}/PE	<0.06	<0.06
OPP-CNC_{COOH}/PLA	12.41 ± 3.5	201.11 ± 3.5
PET-CNC_{COOH}/PE	<0.06	<0.06
PET-CNC_{COOH}/PP	<0.06	<0.06
Esterified CNC_{COOR} (CNC_{SO3H} and CNC_{COOH} mixture)		
OPP-CNC_{COOR}/PE	<0.06	<0.06
OPP-CNC_{COOR}/PLA	3.78 ± 3.5	103.55 ± 3.5
PET-CNC_{COOR}/PE	<0.06	<0.06
PET-CNC_{COOR}/PP	<0.06	<0.06

Chemical modification has often been proposed as a potential route to reduce the hygroscopic nature of cellulose nanomaterials. The basic principle is the reduction of hydroxyl groups via a range of different chemical processes such as esterification (acetylation being most common), silylation, etherification, and polymer grafting, just to name a few (Missoum et al., 2013; Wei, Agarwal, et al., 2017). Additionally, the hygroscopic nature of CNMs can be reduced via crosslinking, either through covalent crosslinking (*e.g.*, carboxylic acids) or through ionic/electrostatic interactions (*e.g.*, metal ions in carboxylated CNMs) (Habibi, 2014). Although these methods have been shown to provide some level of reduction in hygroscopic nature there are two limiting factors that arise when trying to move CNMs towards commercialization. First, CNMs (in their current state) are relatively expensive to produce. Therefore, any additional chemical modification would only make them more expensive. Secondly, crosslinking will likely make what is already considered a brittle material even more brittle. This may have impacts on durability of CNM layers in thin films. Crosslinking with citric acid or metal ions makes CNMs more resistant to moisture but may also cause embrittlement.

Nevertheless, despite the clear potential of such strategies, the literature specifically addressing those modified CNMs within multilayer film architectures remains very limited. To date, studies directly investigating chemically modified CNMs in true multilayer packaging architecture are lacking, underscoring a significant research gap and opportunity for further development.

3.4. *Effect of adhesion tie layer on permeability*

Incorporating an adhesive tie layer is crucial for integrating CNM films with plastic layers into a mechanically stable and high-performance multilayer structure. Due to the porous nature of CNM layers, the absence of a soft adhesive layer can result in poor interfacial adhesion, allowing water vapor to penetrate the interface between the hydrophobic plastic and the hydrophilic CNM. This moisture ingress weakens the bond and causes swelling of the CNM layer, reducing its oxygen barrier performance, especially under high relative humidity (Fig. 6 (C)). An adhesive tie layer mitigates these issues by sealing surface pores and limiting moisture diffusion, which preserves structural integrity and maintains barrier efficiency under humid conditions. For example, CNC (5.7–6.1 $cm^3/(m^2$·day)) and CNF (10.5–24.3 $cm^3/(m^2$·day)) individual films (14 μm) laminated with polypropylene (PP) using polyurethane (PU) tie layers (50 μm) demonstrated significantly lower OTR at 80 % RH (Wang et al., 2020). In contrast, a thinner CNC coating (1.44 μm) applied onto PP with a 1.58 μm acrylic resin (AR) tie layer resulted in a PP/AR/CNC/AR/PP multilayer film exhibiting an OTR of 86.7 $cm^3/(m^2$·day) (Du et al., 2021). The difference in barrier performance may be attributed to variations in CNC layer thickness, tie-layer chemistry, interfacial compatibility, and total laminate architecture. These results highlight the need for systematic studies evaluating the influence of adhesive type and tie-layer thickness on gas barrier performance. It should also be pointed out that the barrier performance of laminated structures could theoretically be predicted based on the barrier properties of the individual layers (Outer layers, tie layers, and core layer) (Vercasson et al., 2025). If the outer and core layer thickness are maintained constant, increasing tie layer thickness will then decrease the overall permeant transmission rate. Notably, laminates incorporating defined adhesive tie layers generally exhibited lower OTR values than some plasma-treated systems without dedicated tie layers, such as bio-HDPE/CNF/bio-LDPE (490 $cm^3/(m^2$·day)) and HDPE/TCNF/LDPE (400 $cm^3/(m^2$·day)) (Vähä-Nissi et al., 2017; Vartiainen et al., 2016) (Table 2). Further OTR reductions were observed in OPP-CNC/PE (<0.06 $cm^3/(m^2$·day)), PET-CNC/PE (1.107

$cm^3/(m^2 \cdot day)$), and PET-CNC/PP (<0.06 $cm^3/(m^2 \cdot day)$) films that combined PU tie layers with plasma surface treatment (Fotie et al., 2020). The plasma surface treatment helps the coating coat more evenly and the tie layer helps prevent gaps between layers that moisture and oxygen can permeate through mor easily. Better coatings and better adhesion both appear to help barrier properties.

These findings highlight the importance of selecting appropriate tie layers to enhance oxygen barrier performance in CNM-based multilayer films and prevent moisture ingression through film edges before sealing. However, the commonly used tie layer like PU and AR tie layers can introduce recyclability and compostability challenges depending on their chemistry and crosslinking structure. Further discussion on tie-layers as it pertains to interlayer adhesion is contained in Section 4.2 of this manuscript. Future research should focus on identifying and optimizing biobased or biodegradable tie layers to align with sustainability goals while maintaining high-performance gas barrier properties and balance delamination for recycling.

3.5. Laminating for humidity resistance

Constructing multilayer coating has been proposed as a strategy to improve the humidity stability of CNM-based barrier packaging. A fundamental question that must be answered is whether sandwiching the hydrophilic CNM between two outer hydrophobic layers can protect it from external humidity. A very limited number of studies have reported some success in doing this. Interestingly, most of these studies also report water vapor transmission rates through the film that are non-zero, indicating that moisture is moving through these types of films. If moisture is moving through the film, then the internal CNM layer should eventually reach a moisture equilibrium. However, the concept of a full equilibrium with elevated moisture is not fully addressed in these studies. Wang et al. (2020) appear to do the best at describing their high-humidity oxygen transmission testing protocol. The researchers set the relative humidity to 80 % on both sides of the double outer layer of PP sandwiched film and performed repeated 1-h tests with a convergence criterion of permeation difference over the last five test cycles of less than 1 %. What could also have been helpful to report was how long the test took, *e.g.*, was it the first 5 h or did it take a month to equilibrate? How can one be certain that equilibrium can occur at a rate faster than 5 h? Additionally, the total thickness of the laminated constructions appeared to be in the range of 100-200 μm, which is quite thick for a thin film application. The thickness of outer protective layers must be considered when considering equilibrium. The other studies that show success at high-humidity oxygen barrier testing of laminated structures are less rigorous in their experimental methods reporting. Yu et al. (2022) measured the oxygen transmission of a CNC-coated PET, but notably only set the elevated (80 %) humidity on one side of the film while the other side was set at 0 % RH. One must assume the CNC coating was on the dry side and being constantly dried out. This could be the real-world case when packaging a dry product, but when packaging a wet product, humidity may be on both sides of the film, and equilibrium must be understood. Fotie et al. (2020) also reported that lamination is a method to protect CNMs from humidity. The researchers tested CNC laminated with a range of different non-polar polymers, and in certain cases showed minimal increase in OTR when moving from 50 % RH to 80 % RH. However, the description of experimental methods is lacking in that it states that the elevated humidity was only applied on one side of the film (*e.g.*, the other dry side can dry out the CNM layer), and there is no mention of how long these tests were performed. The timescale of testing must be considered in order to determine if the CNM layer is reaching equilibrium.

3.6. Food shelf-life applications

Different applications require substantially different barrier levels, ranging from low oxygen resistance (*e.g.*, breathable produce packaging) to very high oxygen barrier performance (*e.g.*, ready-to-eat meals or oxygen-sensitive foods). CNM-based multilayer films represent a tunable platform whose design must be aligned with product-specific humidity exposure, required shelf life, mechanical performance, and recyclability considerations. Specifically, we now discuss how CNM–plastic multilayer systems can be tailored as follows based on Fig. 5 and Table 2:

Fresh produce, such as fruits and vegetables, typically tolerates relatively high oxygen transmission rates to prevent anaerobic respiration, with OTR values often exceeding 10^2–10^3 $cm^3 \cdot m^{-2} \cdot day^{-1}$. For these applications, extremely dense CNC-based barrier cores are not necessary. Instead, thinner CNM layers combined with breathable polymer substrates may provide sufficient oxygen regulation while maintaining moisture balance.

In contrast, modified atmosphere packaging (MAP) systems for meat and seafood require significantly lower oxygen transmission, generally below 10 $cm^3 \cdot m^{-2} \cdot day^{-1}$, to preserve product quality and extend shelf life. Multilayer configurations such as PET/CNC/PE or OPP/CNC/PE (Table 2) demonstrate OTR values within this low-barrier range under controlled humidity conditions, indicating that CNC-based cores are particularly suitable for oxygen-sensitive refrigerated products.

For dry food products including bakery goods and snacks, oxygen barrier performance remains critical, whereas internal humidity is typically low. Laminated systems such as PLA/CNC/PLA and PP/PU/CNC/PU/PP provide effective oxygen reduction while offering adequate moisture resistance under moderate relative humidity conditions.

In high-humidity internal environments, such as wet or semi-moist foods, oxygen permeability of cellulose increases sharply above approximately 65 % RH due to moisture-induced swelling. Under these conditions, CNF-based multilayer systems laminated with hydrophobic polymers (*e.g.*, HDPE/CNF/LDPE) are more appropriate, as their entangled fibrillar network provides improved mechanical stability and humidity tolerance compared to brittle CNC films.

Finally, flexible pouch applications impose additional mechanical requirements. As illustrated in the tensile strength–elongation map (Fig. 5 (B)), polymers such as LDPE, PBAT, and PBS provide high elongation suitable for flexible packaging. In such systems, CNF cores are advantageous due to their interconnected network structure, which enhances flexibility and structural integrity relative to CNC-based layers.

4. Adhesion between polar nanocellulose and non-polar polymers

A key challenge in laminating CNMs is achieving strong adhesion between the polar CNM layer and the typically non-polar polymer layer that provides water resistance. Adequate interlayer adhesion is essential to prevent delamination during converting or product use. However, in some cases, controlled delamination is desirable during recycling to facilitate the separation of individual layers. This dual requirement—strong adhesion during service and easy delamination at end-of-life—makes understanding interfacial adhesion mechanisms critical for optimizing laminate performance and recyclability.

4.1. Existing studies of CNM laminate adhesion

Few studies have quantified the adhesive strength between CNM layers and plastic substrates. Nabels-Sneiders et al. (2023) tested the interlayer adhesion strength of micro/nano fibrillated cellulose derived from hemp laminated with PBS using a T-peel test. Without chemical modification or tie layer, a steady state peel strength of around 0.04 N/mm with a peak strength of 0.08 N/mm were achieved. The micro-scale fibers likely contribute to mechanical interlocking with PBS, improving the joint strength. Although standard adhesion requirements for thin-film food packaging are not well documented, the values reported by Nabels-Sneiders et al. are comparable to those aluminum-based multilayer packaging (Jesdinszki et al., 2012; Paul

et al., 2022). Bamps et al. (2021) suggested 0.5 N/mm as an optimal peel strength for packaging that must be delaminated by the user to open. Also, Liang et al. (2015) achieved close to 1 N/mm of peel strength between polypropylene and aluminum by chemically modifying the polypropylene, *e.g.,* it is possible that the values reported by Nabels-Sneiders et al. are either within the acceptable range or perhaps one order of magnitude lower. Saremi et al. (2020) performed peel testing of a mechanically fibrillated CNF laminated with both smooth sheets and woven fabrics made of cellophane, PET, and PA6,6. The researchers reported peel strengths on the order of 0.1 N/mm for smooth substrates with slightly better adhesion to cellophane than the PET or PA 6,6. The peel strengths were higher at 0.3 N/mm when laminating with woven polymer fabrics, suggesting that mechanical interlocking aids in bonding. The researchers also showed that peel strength was improved by treating the surfaces of the plastic substrates with either polyethylenimine or a copolymer of glycidyl methacrylate (GMA) and oligo (ethylene glycol) methacrylate before coating with nanocellulose. Additionally, d'Eon et al. (2017) examined the adhesion of CNCs on a polypropylene substrate. The substrate was treated with UV-ozone to enhance surface energy to improve the wetting and adhesion of the CNC suspension during spin coating. Adhesion was examined via a scratch test with a 10 mm stainless steel ball that traverses 30 mm along the surface while linearly increasing the downward force up to 5 kg. With the UV-ozone-treated substrate, the adhesion was strong enough that the CNC film was not delaminated during the scratch test, while other samples with a primer designed to reduce adhesion did delaminate. Surface treatments such as ozone or corona plasma appear to enhance interfacial adhesion, but the effects of such treatments have been known to dissipate over time, which may affect adhesion over time. Also, one limitation to using a scratch-based adhesion test is that it is hard to compare to a peel-based test. Using a peel-based test allows for values to be compared quantitatively in peel force per unit width of sample.

4.2. Chemical modification and tie layers

Adhesion between polar wood fibers and non-polar polymers has long been an issue in wood-plastic composites (Matuana, 2009). A traditional approach involves the use of coupling agents that form a chemical bridge or modify the wood fibers surface to improve compatibility. Typical coupling agents include isocyanates, anhydrides, silanes, and anhydride-modified copolymers (Li & Matuana, 2003; Lu et al., 2000; Matuana et al., 1999). Isocyanates are effective but high toxic (Pawlak et al., 2024), prompting efforts to find safer alternatives. Silanes can effectively hydrophobized CNMs (Habibi, 2014; Jo et al., 2021; Missoum et al., 2013) but their toxicity and higher costs limit large-scale use. Another approach is to use organic or polycarboxylic acids (*e.g.,* citric, oleic, lactic, maleic acid) or canola oil fatty acid methyl ester (Wei, Agarwal, et al., 2017, Wei, Luo, et al., 2017) for esterification, improving CNMs more compatibility with non-polar polymers. These acids could provide a cost-effective route to compatibility and provide the dual purpose of making the CNM layer more resistant to humidity. However, the challenge is adding organic acid directly to CNM suspensions can alter the viscosity, affecting the coating process.

In flexible films, interlayer adhesion is often achieved using a thin tie layer that can bond with both layers. This is the standard approach for laminating polar ethylene vinyl alcohol (EVOH) with non-polar polymers. Anhydride-modified polyolefins are typically used for tie layers. For instance, maleic anhydride–grafted polyethylene can bond EVOH to polyethylene (Azzaz et al., 2025; Huang et al., 2003). The maleic anhydride groups grafted onto polyethylene interact with the hydroxyl groups of EVOH, while the polyethylene sides bond with the other polyethylene layer. It is hypothesized that a similar approach of using a maleic anhydride modified polymer as a tie layer could adequately bond cellulose to an external laminating polymer through similar mechanisms. Because adhesion occurs only at the interface, the tie layer can remain very thin, requiring minimal chemical modification. However, its presence may still influence polymer recyclability after delamination.

Several studies have applied tie layers to improve CNM-plastic adhesion. Wang et al. (2020) used a polyurethane tie layer to laminate CNMs with polypropylene, improving adhesion and reducing interfacial moisture penetration. Du et al. (2021) utilized an acrylic resin as a tie layer between CNC and polypropylene, achieving good adhesion without delamination. While anhydride-modified tie layers have not yet been reported for CNM laminates, maleic anhydride–grafted polypropylene has proven effective in wood–polymer composites, suggesting promise for future CNM–polymer systems (Lu et al., 2000).

4.3. Methods of adhesion testing

Research on the interlayer adhesion requires method of quantification of the adhesion strength (Fig. 7). The International Organization for Standards (ISO) and the American Society of Testing and Materials (ASTM) have established standards for peel testing. ISO 11339 specifies a T-peel test for flexible adherends and ISO 8510 describes a 180° peel test where one arm of the adherends is fixed to a stiff aluminum plate. ASTM has a corresponding T-peel test, ASTM D1876, which is similar to the ISO T-peel (11339), and has standard ASTM D903, which is like the ISO 180° peel test (8510). Most studies that have examined thin film packaging materials have utilized a 180° peel test (Bichler et al., 1997; Jesdinszki et al., 2012; Paul et al., 2022), as a stiff substrate can provide added control for the system. For CNM-based laminates, fixing the brittle CNM layer to a rigid plate and peeling the flexible polymer layer is preferred to avoid film fracture and to isolate interfacial adhesion. Careful observation is required to confirm that failure occurs at the bondline (adhesive failure) rather than within the materials (cohesive failure). For mechanically refined CNF, larger microscale fibrils can lead to mechanical interlocking with the laminating polymer. The contributions of chemical adhesion and mechanical interlocking to system performance should be elucidated when relevant. Additionally, the effect of defects at the interface, as well as how long-term moisture swelling cycles (fatigue) can alter bond integrity, must be systematically evaluated. As with any type of mechanical characterization of a cellulose film, sample conditioning (humidity) will need to be controlled and documented.

4.4. Recycling

Recent researchers have begun examining how to separate different layers of multilayer films to produce mono-materials for recycling (Fraldi et al., 2014; Loukodimou et al., 2024; Tamizhdurai et al., 2024). Using a CNM layer as an oxygen barrier could help. The CNM could be removed during recycling by soaking and mechanical action in water. The hydrophilic CNMs would swell and disperse, leaving behind a single plastic layer. The process would be similar to how pulp fibers are dispersed in paper recycling. Ji et al. (2023) examined this concept for the coating of chitin nanowhiskers and cellulose nanocrystals on a PET substrate. The CNCs were removed in pH 7 water and the chitin was removed in pH 3 acetic acid. The recovered nanoparticles were then coated onto the PET substrate for a second cycle, and the oxygen barrier performance of the system only saw a slight reduction compared to the originally coated material. The chitin was used as the first layer on which the CNCs were coated, but no chemically modified tie layer was used, and the adhesion was not measured. Al-Gharrawi et al. (2021) examined a CNF layer between polyethylene (PE) and paper. More paper fibers were recovered during aqueous recycling because they were not embedded into the laminating polyethylene layer, *e.g.,* the thin CNF coating (4 g/m^2) served as a delamination layer, allowing for pulp fibers to be easily removed. 180° peel testing showed a tenfold reduction in peel load. It was proposed that this reduction is due to the difference in mechanical interlocking. The PE flows around the much larger pulp fibers better than it does into CNFs.

Several unexplored questions arise from the a forementioned

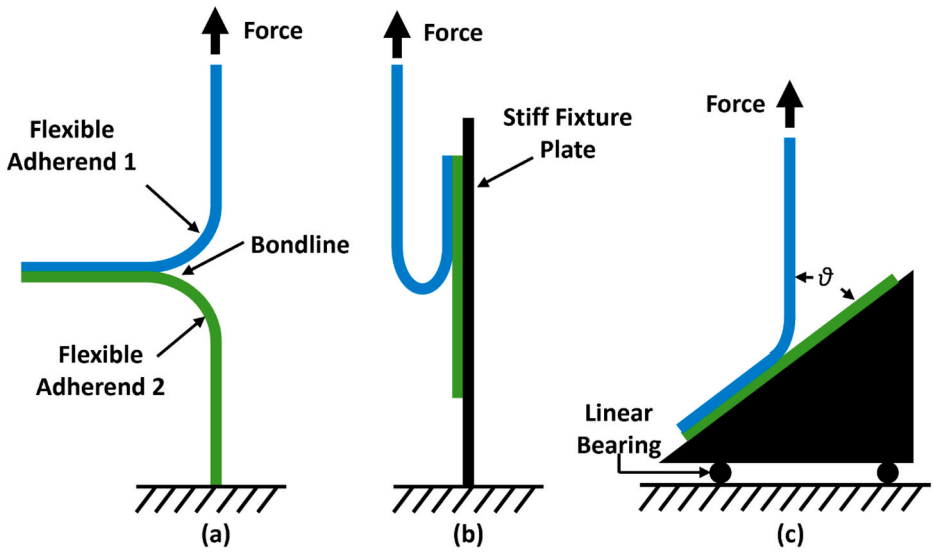

Fig. 7. Schematics of the various peel testing methods discussed: (a) T-peel of flexible adherends (ISO 11339 and ASTM D1876), (b) 180° peel test with fixed plate (ISO 8510 and ASTM D903), and (c) Fixed angle (θ) peel test for energy analysis described in protocol by Kawashita et al. (2006).

recycling concept and existing studies. First, if the CNMs are sandwiched between two hydrophobic polymers, can the aqueous medium adequately penetrate between the layers to force the CNMs to swell, *e.g.,* what type of mechanical prepossessing (*e.g.,* shredding, perforation) is required? Secondly, how does the addition of a tie layer alter the delamination process during recycling? A tie layer may result in more fibers staying adhered to the plastic substrate. It then becomes a question of the acceptable limit of fiber impurities in the mono-material polymer. Lastly, there exists a desire to increase the water resistance of CNMs in order to improve their poor oxygen barrier properties at high humidities. If the water resistance is improved, will it limit the material's ability to be redispersed in an aqueous recycling process? These questions should be the subject of further research in this area.

5. Opportunities and challenges

5.1. Opportunities

5.1.1. High oxygen and water vapor barrier packaging materials

The preparation of nanocellulose-plastic polymer based multilayer films offers diverse opportunities through lamination and coating techniques, enabling the development of high-barrier packaging materials. Lamination provides robust structural films where CNMs serve as oxygen barriers and polymer layers protect against moisture and mechanical damage. Coating technologies add further flexibility by enabling nanoscale control and integration of functional materials like chitin or chitosan. These methods demonstrate excellent performance in oxygen and water vapor barrier even in high humidity environments, particularly when adhesives or surface treatments are used or CNMs are hydrophobic functionalized to enhance interfacial adhesion. This multilayer structure has the potential to address the challenge of cellulose nanofiber (CNF)'s reduced oxygen barrier performance under high humidity and the limited oxygen barrier effectiveness of plastic polymers when used as monolayer films.

5.1.2. Enabling sustainability

Unlike current commercial multilayer barrier packaging materials that incorporate metallized films or ethylene vinyl alcohol (EVOH) coatings, CNM–polymer-based multilayer films have the potential to offer new routes of enhanced recyclability—particularly in configurations where cellulose nanomaterials (CNMs) are coated on one side of plastic substrates. The proposed concept is that upon immersion in water, the CNM layer swells and disperses into suspension, allowing it to be readily separated from the underlying polymer film. This separation enables both the CNM and polymer components to be individually recovered and potentially reused, thereby supporting sustainable material use and contributing to a circular economy. Currently, it is rare that even mono-material thin film polymers are recycled, but as recycling technologies advance this approach could become a useful strategy to achieving oxygen barrier performance with mono-material recyclability, which is not possible with current thin films.

5.1.3. Enabling fully biodegradable thin film solutions

Biodegradable polymers such as PLA, PHA, PBS, and PBSA offer significant potential to replace conventional plastics in the development of fully biodegradable multilayer films. When used in thin film packaging, these polymers will need the addition of an oxygen barrier layer. Given that cellulose is the industry standard by which other materials biodegradation is tested against, it appears that a CNM barrier layer could add performance both is oxygen barrier properties and biodegradation properties.

5.2. Challenges towards industrial translation

5.2.1. Scientific and technical knowledge gaps

5.2.1.1. Moisture equilibrium and long-term barrier stability. Most reported oxygen and water vapor barrier measurements are conducted over short testing periods. However, real-world applications (such as food packaging) require barrier performance to be maintained over extended shelf lives, often lasting several months. To date, there is a lack of published research investigating the long-term barrier properties of CNM-based multilayer films at elevated humidity, *e.g.,* how long-term moisture transmission pertains to the equilibrium moisture of a CNM layer sandwiched between two outer hydrophobic layers. Outer hydrophobic layers may protect the CNM layer over several hours (timescales in existing studies), but moisture may transport through outer layers over the course of months and have an impact on the CNM core layer. This represents a critical knowledge gap in evaluating their practical viability. Further studies are needed to better understand the equilibrium moisture content of CNM layers when laminated between hydrophobic outer layers under varying humidity conditions. In multilayer structures, differing humidity levels on either side of the film can influence the internal moisture distribution and, consequently, the barrier

performance. Specific food packaging applications will produce different humidity environments that must be accounted for. For example, a wet food product will produce a relative humidity close to 100 % inside the package, whereas dry food may have very low humidity within the package, and only the exterior humidity is relevant. This means researchers must account for where the humidity is and what type of water vapor barrier (material and thickness) is between the humidity and the CNM layer. Moreover, the time scale required to reach moisture equilibrium is a key factor in determining how long the packaging material can maintain its functionality in specific environmental conditions.

5.2.1.2. Interfacial adhesion vs delamination challenges. Adhesives, tie layers, surface treatments, or hydrophobic functionalization of CNMs are potential routes for enhancing interfacial adhesion in CNM–plastic multilayer films, thereby improving overall packaging performance. However, these approaches may also introduce challenges when trying to use the CNM layer as a delamination layer for recycling purposes. Despite its importance, there is currently a lack of research specifically focused on understanding the relationship between interfacial adhesion strength and delamination behavior in CNM multilayer films. Addressing this gap is crucial for optimizing both performance and long-term durability of sustainable barrier materials. These trade-offs highlight the need for further research to optimize interfacial design strategies that balance strong adhesion for product use with controlled delamination for end-of-life recycling.

5.2.2. Industrial and system-level challenges

5.2.2.1. Scalable processing and manufacturing. Despite promising laboratory results, industrial adoption of CNM-based multilayer films remains limited. Challenges include large-scale coating uniformity, drying energy requirements, rheological control, and integration into existing lamination lines. Some industrial efforts involve nanocellulose coatings on paperboard or flexible film substrates to improve printability, grease resistance, and oxygen barrier performance in niche applications (*e.g.*, food pouches, carton liners), but these products generally do not yet represent mainstream multilayer packaging solutions. For instance, companies such as CelluCoat and nanocellulose-coated paper products from Stora Enso (https://www.packaging-gateway.com/data-insights/stora-enso-gets-grant-for-gas-barrier-film-for-paper-packaging-with-microfibrillated-cellulose/?utm_source=chatgpt.com) and Mondi (https://www.mondigroup.com/products-and-solutions/flexible-packaging/functional-barrier-papers/?utm_source=chatgpt.com) have explored nanocellulose coatings for functional enhancements, and pilot programs have evaluated nanocellulose barrier layers in lamination with PET or PE films. These developments highlight growing industrial interest, but full integration of CNMs as dedicated barrier layers in commercial multilayer packaging remains limited. The high viscosity and typically low solids content of CNM dispersions restrict coating speed and increase drying time, making it difficult to match current industrial production rates. Additionally, achieving robust adhesion between polar CNM layers and non-polar polymer substrates presents further technical challenges. Overcoming these processing and interfacial limitations will be essential for enabling scalable manufacturing and successful commercialization of CNM-based multilayer packaging systems.

5.2.2.2. Environmental trade-offs and life cycle assessment. While water-triggered delamination of CNM–plastic multilayers is promising for recycling, its net environmental benefit depends on life cycle trade-offs. Existing LCA work shows that the environmental footprint of CNMs is highly sensitive to the production route (chemical inputs, electricity demand) and especially to drying/film formation energy, which can dominate cradle-to-gate impacts. From an end-of-life perspective, delamination can improve circularity by enabling higher-quality polymer recovery compared with conventional multi-material laminates, but the process may introduce additional burdens associated with water use, chemical aids (if any), and wastewater management. Therefore, future LCAs should evaluate CNM–plastic multilayers using functional units tied to packaging performance (*e.g.*, required shelf-life under realistic humidity), and explicitly compare scenarios such as mechanical recycling with delamination versus conventional multilayer disposal routes. In this context, aqueous-based separation and recovery approaches for bio-based barrier layers have been proposed as potentially scalable circular pathways, but robust comparative LCAs remain limited and are needed to quantify benefits and trade-offs.

5.2.2.3. Regulatory considerations. The translation of CNM-based multilayer films into food packaging applications requires alignment with regulatory frameworks governing food contact materials. In the United States, the Food and Drug Administration (FDA) regulates food-contact substances under the Federal Food, Drug, and Cosmetic Act. Materials intended for direct or indirect contact must be demonstrated as safe for their intended use, typically through a Food Contact Notification (FCN) or inclusion in the Generally Recognized as Safe (GRAS) list, supported by toxicological and migration data (FDA, 2023). In the European Union, Regulation (EU) No 10/2011 sets specific migration limits (SMLs) and criteria for food contact plastics and polymers, and any novel or engineered nanomaterial must undergo a case-by-case safety assessment before authorization (EFSA, 2020). Currently, cellulose-based nanomaterials are not listed explicitly in regulatory guidance, and there remains ongoing discussion regarding the adequacy of existing migration testing methods for nanoscale components. Both jurisdictions emphasize the need for robust characterization, migration testing under realistic conditions, and toxicological evaluation to ensure consumer safety. These regulatory contexts present challenges and opportunities for CNM-based packaging, underscoring the importance of early consideration of compliance as part of material design and commercialization strategies.

5.2.2.4. Economic feasibility and cost competitiveness. Moreover, the variability of CNMs—stemming from differences in raw material sources and production methods—complicates performance standardization and reproducibility. Their integration into industrial processing remains both technically and economically challenging, limiting scalability. Addressing these barriers is essential for the successful commercialization of CNM-based packaging solutions. In addition, the production of biodegradable polymers typically incurs higher costs compared to the synthesis of conventional, fossil-based polymers, further constraining their competitiveness in large-scale applications.

6. Conclusion

Nanocellulose-based multilayer films have emerged as a highly promising solution for developing sustainable, high-barrier packaging materials. Their exceptional oxygen barrier performance, particularly under dry conditions, along with their compatibility with biodegradable polymers, makes them strong candidates to replace conventional synthetic and metallized packaging films. Lamination techniques offer scalable routes for producing robust multilayer structures, while coating methods provide nanoscale control and functional customization. Despite these advantages, significant challenges remain. The inherent hydrophilicity of cellulose nanomaterials (CNMs) leads to compromised barrier performance under high humidity. Additionally, poor interfacial adhesion with non-polar polymers must be addressed. While tie layers and chemical modifications can enhance compatibility, they often raise concerns regarding recyclability, toxicity, and processing complexity. Additionally, the lack of standardization in CNM production, variability in performance metrics, and the technical difficulties in industrial

processing limit the widespread adoption of CNM-based multilayer systems. To fully realize the potential of plastic laminated CNM systems, future research should focus on improving moisture resistance without sacrificing recyclability, developing safe and effective adhesion strategies, and establishing standardized benchmarks for barriers and mechanical properties. Integrating life cycle assessment, economic feasibility, and scalability considerations will be crucial for translating laboratory advances into commercial applications. Addressing these challenges will pave the way for the next generation of environmentally responsible, high-performance packaging solutions that align with circular economic principles.

Availability of data and material

Data will be made available based on a reasonable request.

CRediT authorship contribution statement

Yufei Nan: Conceptualization, Methodology, Investigation, Formal analysis, Visualization, Writing – original draft. **Daniel J. Franke:** Conceptualization, Methodology, Investigation, Formal analysis, Visualization, Writing –review & editing. **Cody A. Schilling:** Formal analysis, Writing –review & editing. **Ronald C. Sabo:** Conceptualization, Methodology, Visualization, Supervision, Funding acquisition, Writing – review & editing. **Nicole M. Stark:** Conceptualization, Methodology, Visualization, Supervision, Funding acquisition, Writing – review & editing. **Laurent M. Matuana:** Conceptualization, Methodology, Visualization, Supervision, Funding acquisition, Writing – review & editing.

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Declaration of generative AI use

ChatGPT was used only to improve the English language of the text after the draft was completed.

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Data availability

Data will be made available on request.

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