

## **DEVELOPMENT OF WATER-RESISTANT PAPER USING NATURAL POLYMERS: A SCIENTIFIC AND TECHNICAL REVIEW**

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### **Abstract**

The development of water-resistant paper has become increasingly important due to the environmental concerns associated with synthetic coatings. Conventional waterproofing agents, such as polyethylene, waxes, and fluorochemicals, are non-biodegradable and hinder recyclability, contributing to long-term ecological issues. As a sustainable alternative, natural polymers such as starch, chitosan, cellulose derivatives, and proteins have gained significant attention due to their renewability, biodegradability, and film-forming capabilities. These biopolymers improve the hydrophobicity of paper surfaces through mechanisms like hydrogen bonding, cross linking, and barrier layer formation. Various coating techniques, including spray coating, dip coating, and layer-by-layer deposition, have been employed to apply these materials effectively. Characterization methods such as Cobb value, contact angle measurements, and FTIR analysis help evaluate coating performance. Despite their potential, challenges such as coating uniformity, printability, durability under humid conditions, and economic scalability remain major obstacles to industrial application. This review presents a comprehensive analysis of natural polymer-based coatings for water-resistant paper, covering material chemistry, application techniques, performance characteristics, biodegradability, and environmental impact. The paper also discusses recent advancements, future trends involving bio-nanocomposites and smart coatings, and the need for standardized testing protocols. Continued interdisciplinary research and collaboration between academia, industry, and regulatory bodies are crucial for commercial viability and environmental sustainability.

**Keywords:** Water-resistant paper, Natural polymers, Biodegradable coatings, Chitosan, Starch, Sustainable packaging, Barrier properties

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### **Introduction**

Paper has been a staple material in human civilization for centuries, widely utilized in applications ranging from writing and printing to packaging and hygiene products. Its natural origin, ease of processing, low cost, and recyclability have contributed to its enduring utility. However, one critical limitation of paper is its inherent hydrophilicity, which renders it unsuitable for applications where moisture or liquid contact is expected. When exposed to water, paper readily absorbs moisture, loses mechanical strength, and undergoes structural deformation. These characteristics significantly limit its utility in sectors such as food packaging, outdoor labeling, medical wraps, and other moisture-sensitive applications. To overcome these limitations, water-resistant paper technologies have historically relied on synthetic coatings. These include petroleum-based polymers such as polyethylene (PE), polypropylene (PP), and polyvinylidene chloride (PVDC), as well as waxes and fluorinated compounds that form hydrophobic barriers over the paper surface. These treatments effectively prevent water penetration by reducing surface energy and blocking pore structures in paper fibers. While these technologies are functionally robust and have been adopted on an industrial scale, they pose significant environmental concerns. The non-biodegradable nature of many synthetic coatings impairs paper recyclability, leads to microplastic pollution, and contributes to the growing problem of plastic waste in landfills and aquatic ecosystems [1,2]. As global attention shifts towards sustainable development and circular economies, the paper and packaging industries are under increasing pressure to develop eco-friendly alternatives.

In this context, natural polymers, derived from renewable biological sources, have emerged as promising candidates for water-resistant paper coatings. These biopolymers including starch, chitosan, alginate, soy protein, gelatin, cellulose derivatives, and polylactic acid (PLA) offer the advantages of biodegradability, biocompatibility, and, in many cases, compostability. They can be engineered to form thin, continuous films over paper surfaces that impart water resistance through mechanisms such as surface hydrophobization, pore blockage, and moisture diffusion barrier formation [3,4]. The development and application of natural polymer-based coatings are inherently interdisciplinary, involving principles from materials science, polymer chemistry, food technology, and environmental engineering. Researchers are exploring various processing techniques, including dip coating, spray coating, blade coating, and extrusion lamination, to apply these bio-based materials onto paper substrates. In addition, recent innovations in nanotechnology such as the incorporation of nanocellulose, nanochitosan, or clay nanoparticles have further enhanced the mechanical strength and barrier performance of these coatings without compromising biodegradability [5,6]. Furthermore, there is a growing interest in multifunctional coatings that not only provide water resistance but also incorporate antimicrobial properties, oxygen barriers, or controlled-release systems for food preservation. For instance, the integration of essential oils, natural antioxidants, and silver nanoparticles has led to the development of active paper packaging capable of prolonging shelf life and ensuring food safety [7,8]. Despite these promising developments, several challenges remain. Natural polymers often suffer from poor adhesion to paper surfaces, limited water resistance under prolonged exposure, and variability in performance due to environmental humidity and temperature. Additionally, the economic feasibility of

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scaling up bio-based coatings for industrial use particularly in comparison to inexpensive and readily available synthetic alternatives continues to be a major barrier to commercial adoption. This review aims to comprehensively evaluate the current landscape of natural polymer-based coatings for water-resistant paper applications. It explores the fundamental scientific principles behind water repellency, various types of biopolymers used, coating technologies employed, and performance evaluation methods. Through critical analysis of recent literature and technological advancements, the paper also identifies key limitations and future research directions necessary for the development of commercially viable, sustainable water-resistant paper products. Recent reviews also emphasize the role of sustainable biopolymer coatings for packaging applications due to their excellent barrier properties [9]. By consolidating current knowledge, this review intends to serve as a reference for researchers, manufacturers, and policymakers working at the intersection of material innovation and environmental sustainability.

### **Necessity and market drivers for natural polymer-based water-resistant paper coatings**

The increasing environmental concerns over synthetic plastic-based coatings in paper products, such as polyethylene (PE), polypropylene (PP), and fluoropolymers, have catalyzed research into sustainable alternatives. These synthetic coatings are non-biodegradable, impair recyclability, and contribute significantly to microplastic pollution in landfills and aquatic ecosystems [1,2]. Despite their cost-effectiveness and industrial scalability, the long-term ecological impact of synthetic coatings is unsustainable, particularly in the context of growing global emphasis on circular economy practices and sustainable development goals.

**Global market demand:** The global paper packaging market is estimated at approximately 400 million tons annually, with food packaging accounting for about 30% of this volume [9]. The market share of natural polymer-based coatings is currently below 5% worldwide, mainly due to technological and economic challenges [9]. However, consumer demand for environmentally friendly, biodegradable packaging solutions, as well as government regulatory pressure, is expected to expand this market share significantly in the coming years.

**Why natural polymers are needed:** Natural polymers such as starch, chitosan, cellulose derivatives, and proteins offer several significant advantages:

- **Biodegradability and Compostability:** These polymers fully degrade within 60–90 days under industrial composting conditions, meeting standards like ASTM D6400 and ISO 17088, in contrast to plastics that persist for centuries [1,37].
- **Regulatory Drivers:** Regulations such as the EU SUP Directive (2019/904) banning single-use plastics and stricter food packaging requirements are major driving forces. Several major brands, including Nestlé and Unilever, have voluntarily committed to replacing plastic-based coatings with sustainable alternatives [1].
- **Lower Environmental Footprint:** Life cycle assessment (LCA) studies have shown that natural polymer coatings reduce carbon footprint and non-renewable energy usage by approximately 45–60% compared to synthetic coatings [37].

**Cost vs. benefit:** Although raw materials for natural polymers are abundant (e.g., corn starch, shrimp shells for chitosan), the cost of coating paper with natural polymers is typically 20–50% higher than plastic-based alternatives [15]. This cost is mainly due to additional processing steps such as chemical crosslinking, purification, and incompatibility with existing high-speed PE coating lines. Nevertheless, the environmental advantages and regulatory compliance benefits justify this investment, especially in food packaging applications where biodegradability and food safety are critical.

**Industrial Challenges:** Applying natural polymers on industrial-scale paper coating machines faces several challenges:

- Poor wetting and adhesion to cellulose-based paper substrates due to their hydrophilicity.
- High viscosity and gelation of some biopolymer solutions make them incompatible with conventional coating equipment.
- Lack of standardized evaluation methods limits comparability and regulatory acceptance [33,34].

Recent advances include enzymatic fiber treatments, hybrid polymer blends, and optimized crosslinking strategies that aim to improve adhesion and barrier performance [25,26].

**Advantages in recycling:** Natural polymer-coated paper does not interfere significantly with recycling processes, unlike synthetic PE coatings, which contribute to problematic sticky residues (“stickies”) during pulping that degrade recycled paper quality [1]. Furthermore, natural polymers do not release persistent organic pollutants (POPs) and degrade safely under composting conditions.

### **Additional market and technical considerations for natural polymers**

**Regulatory and safety requirements:** Food-grade paper coatings must meet stricter regulatory and safety requirements compared to industrial water-resistant papers. Regulations such as FDA CFR Title 21 and EU Regulation 10/2011 mandate that all materials in contact with food must be non-toxic, free of migration risks, and capable of withstanding wet food contact conditions without degrading. In contrast, industrial water-resistant papers primarily focus on mechanical barrier properties and durability without stringent food safety standards [15,37].

**Printing performance comparison:** Natural polymer coatings influence the printability of paper by altering surface energy and absorption properties. Research indicates that chitosan-coated papers generally exhibit improved ink adhesion and color intensity compared to polyethylene (PE)-coated papers, due to the hydrophilic nature of chitosan increasing ink wettability [26]. However, some studies report slightly higher surface roughness, which may affect fine-detail printing resolution [15]. Measurement of print density, adhesion strength, and drying time are critical parameters that should be compared under standardized testing protocols.

**Extra cost to consumer:** The additional cost incurred for natural polymer-coated paper compared to conventional PE-coated paper is typically in the range of \$0.05 to \$0.15 per square meter, depending on the polymer type and processing

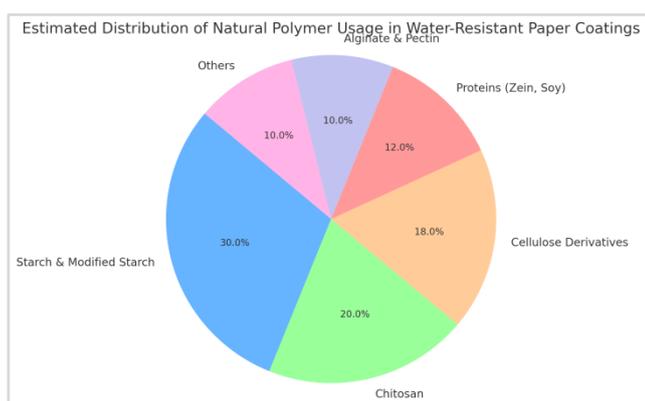
method [15]. This price difference arises due to higher raw material costs (e.g., chitosan extraction), the need for specialized cross linking steps, and slower industrial coating line speeds due to higher viscosity. Despite the higher cost, the environmental benefits and regulatory compliance make it a worthwhile investment, particularly for food packaging applications where biodegradability and non-toxicity are critical. To better understand the environmental and economic implications of switching from synthetic to natural polymer-based coatings, a comparison of key life cycle assessment (LCA) parameters is summarized in Table 1. The table highlights that natural polymer coatings such as chitosan and starch offer significantly lower carbon footprints and faster biodegradation times compared to polyethylene (PE)-based coatings. Furthermore, they do not interfere with recycling processes, unlike synthetic coatings that generate problematic "stickies." Although natural polymer coatings incur an additional cost of approximately \$0.05–\$0.15 per square meter, this is justified by the environmental and regulatory benefits [37,15].

**Table 1. Life Cycle Assessment (LCA) comparison**

Parameter	Polyethylene Coating	Chitosan/Starch Coating
Carbon Footprint (kg CO <sub>2</sub> eq/kg paper)	~5.0	~2.5
Time to Biodegradation	>500 years	~60–90 days
Recyclability Impact	Impairs recyclability	No significant interference
Cost Difference	Baseline	+\$0.05–\$0.15/m <sup>2</sup>

## Overview of natural polymers suitable for paper coating

Natural polymers commonly used in water-resistant paper coatings include starch, chitosan, cellulose derivatives, proteins, and lignin. These materials are renewable, biodegradable, and offer functional properties that allow them to form hydrophobic barriers when applied to paper surfaces [9,15]. Starch and chitosan are particularly widely studied due to their film-forming ability and low cost.



**Figure 1. Estimated usage distribution of key natural polymers in water-resistant paper coatings**

Figure 1 shows the estimated usage distribution of key natural polymers in the development of water-resistant paper coatings, highlighting starch and chitosan as the most widely utilized materials due to their film-forming and barrier properties, based on a survey of recent literature [1, 2, 3].

### Starch and modified starch

Starch is one of the most abundant and cost-effective biopolymers, derived from sources like corn, potato, and

cassava. Native starch, although biodegradable and widely available, is inherently hydrophilic. Therefore, chemical modifications (e.g., esterification, acetylation) or physical blending with hydrophobic agents are employed to improve its water-resistant properties. Cationic or oxidized starches have been used to enhance bonding with paper fibers while contributing to film formation.

### Chitosan

Chitosan is a cationic polysaccharide derived from the deacetylation of chitin, primarily found in crustacean shells. It is known for its excellent film-forming, antimicrobial, and barrier properties. Its ability to form strong hydrogen bonds with cellulose fibers makes it a preferred choice for paper coating. Chitosan-coated papers demonstrate notable resistance to moisture, grease, and microbial degradation.

### Cellulose Derivatives

Various cellulose-based materials such as carboxymethyl cellulose (CMC), hydroxypropyl methylcellulose (HPMC), and ethyl cellulose have been utilized for coating applications. These derivatives exhibit good adhesion to paper fibers and can form transparent, flexible films. Some derivatives, especially ethyl cellulose, provide enhanced hydrophobicity compared to native cellulose.

### Proteins (Zein, Soy Protein, Casein)

Proteins are biodegradable polymers capable of forming cohesive films with moderate to good barrier properties. Zein, a prolamin protein extracted from corn, is particularly water-insoluble and forms glossy, durable coatings. Soy protein isolate and casein have also been studied; however, they often require plasticizers or cross-linking agents to enhance water resistance.

### Polyhydroxyalkanoates (PHAs)

PHAs are microbial polyesters synthesized by bacteria under nutrient-limiting conditions. Among them, polyhydroxybutyrate (PHB) is the most widely studied. These polymers are inherently hydrophobic, biodegradable, and can serve as effective barriers against water vapor and grease. However, the cost of production and brittleness are current limitations for widespread use.

### Alginate and Pectin

Extracted from seaweed and fruit peels respectively, alginate and pectin are water-soluble polysaccharides. While their hydrophilicity limits water resistance, blending with hydrophobic agents or crosslinking (e.g., with calcium ions) can enhance barrier performance. They are frequently used in food-contact paper products due to their biocompatibility.

### Lignin and Tannins

These plant-derived polyphenols have inherent hydrophobic characteristics. Lignin, a by-product of the paper pulping industry, can be used in coatings to reduce water absorption. Tannins, due to their high phenolic content, also show promise in forming cross-linked networks when combined with other polymers. A comparative summary of natural polymers used in coatings is presented in Table 2.

**Table 2. Comparison of selected natural polymers used in paper coating applications**

Polymer	Source	Barrier Type	Pros	Cons
Chitosan	Crustacean shells	Water, microbes	Antimicrobial, good adhesion	Costly, pH-sensitive
Starch	Corn, potato	Water	Inexpensive, available	Hydrophilic
Zein	Corn protein	Water vapor	Hydrophobic, biodegradable	Brittle
CMC	Cellulose	WVTR	Good adhesion, printable	Limited hydrophobicity

## Natural polymers for water resistance

The molecular structure and functional groups of natural polymers influence their performance in coating applications.

- Starch: Offers high film-forming capacity but is inherently hydrophilic. Crosslinking strategies are often required to improve water resistance and mechanical strength [15].
- Chitosan: A cationic polysaccharide with good film-forming ability and intrinsic antimicrobial properties. Its ability to form hydrogen bonds with paper fibers enhances adhesion and barrier performance [26].
- Cellulose Derivatives (e.g., Carboxymethyl Cellulose – CMC): Improve coating uniformity due to good solubility in water, but require blending or crosslinking to enhance barrier properties [13].
- Proteins (e.g., Gelatin, Casein): Provide high mechanical strength and good gas barrier properties but are sensitive to moisture and require chemical modification for water resistance [14].
- Lignin: Provides hydrophobicity and UV resistance, yet suffers from poor solubility and processability, making its application in coating formulations challenging [15].

While individual properties of natural polymers are well documented in the literature, there is a critical lack of research integrating these insights into scalable industrial processes. Most studies focus on laboratory-scale formulations without addressing challenges such as compatibility with high-speed coating lines, process reproducibility, and standardization of barrier property testing. Bridging this gap is crucial for advancing industrial adoption of natural polymer coatings.

## Coating techniques using natural polymers

Various techniques are employed to apply natural polymers onto paper substrates. The choice of method affects coating uniformity, penetration, water resistance, and final paper performance. A simplified representation of these techniques is provided in Figure 2 categorizing the approaches into surface application, film forming methods, crosslinking strategies and lamination technologies.

Various techniques are employed to apply natural polymers onto paper substrates. The choice of method significantly affects coating uniformity, penetration, water resistance, and final paper performance.

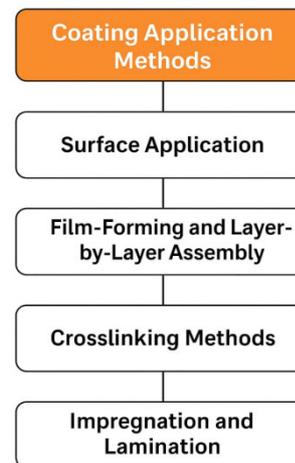
Figure 3 illustrates a general process flowchart of common coating application methods used for natural polymer coatings on paper substrates.

These methods can be broadly categorized as:

- Surface Application Techniques (e.g., rod coating, spray coating, dip coating), where the polymer solution is spread over the paper surface to create a thin barrier layer.

- Layer-by-Layer (LbL) Assembly, involving alternate deposition of oppositely charged natural polymers, creating nanolayered structures that improve barrier performance and reduce porosity.
- Crosslinking Methods (thermal, enzymatic, or chemical), which create a three-dimensional polymer network, enhancing water resistance and adhesion.
- Lamination Techniques, where pre-formed biopolymer films are laminated onto the paper to provide a continuous hydrophobic barrier and improve mechanical and barrier properties.

These techniques are chosen depending on the desired coating performance, process scalability, and material compatibility [13,14,25].



**Figure 2. Schematic flowchart illustrating common coating application methods used for natural polymer coatings on paper substrate**

### Surface application (Rod coating, spray coating, dip coating)

These traditional methods involve spreading a polymer solution onto the paper surface. Rod coating is precise and suitable for lab-scale testing, while spray and dip coating are cost-effective for scalable processing. Thickness and uniformity can be controlled by adjusting concentration and drying parameters [13].

### Film-Forming and Layer-by-Layer (LbL) Assembly

LbL involves alternate deposition of oppositely charged polymers (e.g., chitosan and alginate), creating nanolayered structures with enhanced barrier properties. These assemblies reduce porosity and enhance hydrophobicity [14].

### Crosslinking Methods (Thermal, Enzymatic, Chemical)

Crosslinking is key to improving water resistance by forming a three-dimensional network. Citric acid, glutaraldehyde, or enzymes like laccase can be used to crosslink starch or

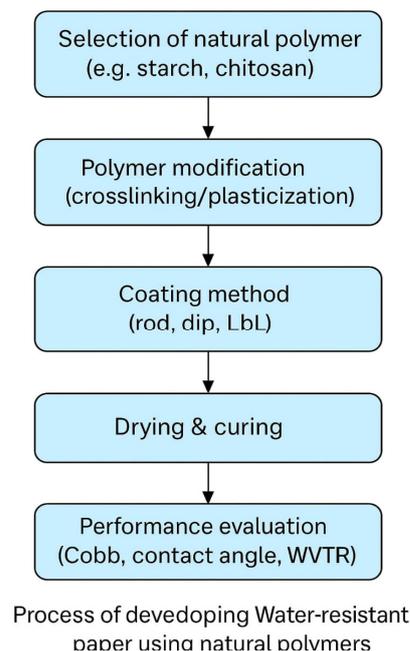
chitosan, improving resistance to water solubility and mechanical deformation [10].

### Impregnation and Internal Sizing

Instead of surface coatings, some biopolymers are impregnated into the fiber matrix. This is common with starch and CMC, and helps in forming hydrogen bonds with cellulose, improving internal bonding and wet strength.

### Lamination with Biopolymer Films

Laminating paper with pre-formed biopolymer films (e.g., PLA or chitosan) adds a waterproof barrier and can improve visual and tactile properties of paper for packaging and printing.



**Figure 3. Process flowchart illustrating common coating application methods used for natural polymer coatings on paper substrates, including surface application, layer-by-layer assembly, crosslinking strategies, and lamination techniques**

### Comparative performance analysis of natural polymer-based coatings

The effectiveness of natural polymer coatings is generally evaluated by comparing their barrier, mechanical, and environmental properties against conventional synthetic coatings (e.g., polyethylene, waxes, fluorochemicals). This section offers a systematic comparison based on recent scientific studies.

#### Water Resistance (Cobb Value and Contact Angle)

Water resistance is commonly assessed using Cobb60 values and static contact angle measurements. Studies show that:

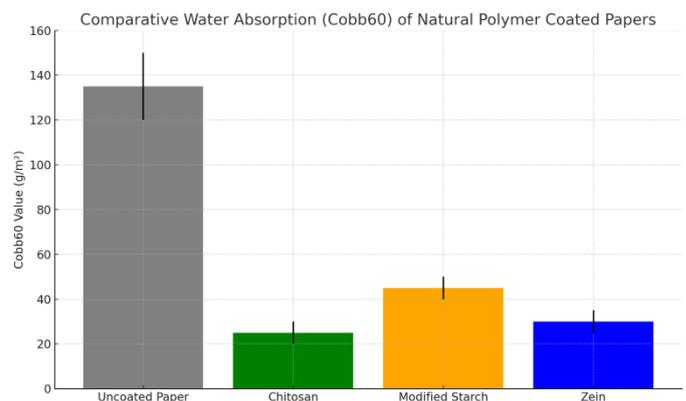
- Chitosan-coated papers have Cobb values as low as 20–25 g/m<sup>2</sup> compared to uncoated kraft paper (~120–150 g/m<sup>2</sup>) [15]. Uncoated bleached Kraft paper was used as a substrate for coating experiments, as bleached Kraft paper is typically preferred in food packaging due to its purity and appearance.

- Starch-based coatings, when crosslinked with citric acid, reduce Cobb values by up to 60% [16].
- Contact angles of over 90° have been achieved using zein and beeswax-based coatings, indicating high hydrophobicity [17].

The water resistance performance is detailed in Table 3.

**Table 3. Comparative water resistance of uncoated and coated papers using natural polymers**

Coating Type	Cobb60 (g/m <sup>2</sup> )	Contact Angle (°)
Uncoated Paper	120–150	35–60
Chitosan	20–30	90–100
Modified Starch	40–50	70–80
Zein	25–35	95–110



**Figure 4. Comparison of Cobb60 Values and Contact Angles of Uncoated and Natural Polymer-Coated Paper Samples**

#### Mechanical Strength

Mechanical performance such as tensile strength, burst index, and folding endurance must be maintained post-coating:

- Cellulose nanofiber (CNF) coatings improve tensile index by 20–30% due to hydrogen bonding [18].
- Starch coatings may reduce flexibility unless plasticizers (e.g., glycerol) are added [16].
- Protein-based coatings, like soy and zein, generally maintain paper strength and are compatible with printing inks [19].

#### Barrier Properties {Water Vapour Transmission Rate (WVTR) and Oxygen Transmission Rate (OTR)}

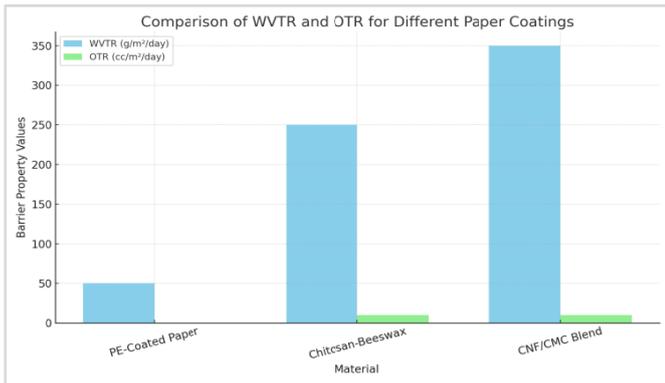
Water vapor and oxygen barrier properties are essential for packaging:

- Chitosan + beeswax coatings reduce WVTR to ~250 g/m<sup>2</sup>/day, close to commercial PE films [20].
- Nanoclay-starch composites exhibit 2–3× better oxygen resistance than uncoated paper [21].

**Table 4. The barrier properties achieved using different biopolymer coatings.**

Material	WVTR (g/m <sup>2</sup> /day)	OTR (cc/m <sup>2</sup> /day)
PE-Coated Paper	~50	<1
Chitosan-Beeswax	250	~10
CNF/CMC Blend	300–400	5–15

**Table 4 Oxygen and water vapor barrier properties of biopolymer-based coated paper**



**Figure 5. Oxygen Transmission Rate (OTR) and Water Vapor Transmission Rate (WVTR) of Biopolymer-Based Coated Papers Compared to PE-Coated Paper**

### Biodegradability and Environmental Impact

Unlike synthetic coatings, natural polymer films decompose within 60–90 days under composting conditions [22]. Many are also:

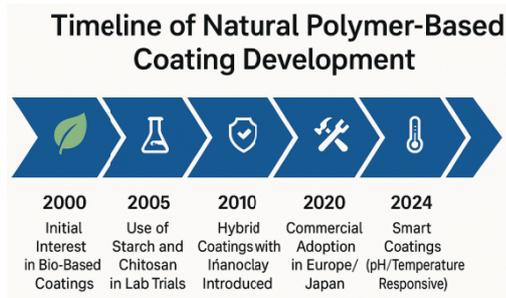
- Derived from agricultural waste (e.g., shrimp shells for chitosan, corn for starch).
- Free from persistent organic pollutants (POPs) and do not contribute to microplastic load.

### Cost and Industrial Scalability

Although raw materials are abundant, natural polymer processing and coating uniformity remain costly. Comparative analysis:

- Starch and cellulose coatings are most cost-effective.
- Chitosan and zein are currently expensive due to purification and processing needs [15,17].
- Innovations in spray and curtain coating techniques are improving scalability.

This timeline summarizes key research and industrial milestones in the development of water-resistant coatings derived from natural polymers, including innovations in formulation, application techniques, and smart coating functionalities.



**Figure 6. Timeline of Natural Polymer-Based Coating Development for Water-Resistant Paper (2000–2024).**

### Case Studies and Recent Research Trends

Case Studies and Recent Research Trends the application of natural polymers in developing water-resistant paper has

advanced considerably in recent years. This section highlights key case studies and emerging research trends that showcase the versatility and effectiveness of biopolymer coatings across different contexts.

### Chitosan-Based Coatings for Packaging Applications

Chitosan, derived from chitin, has emerged as a powerful film-forming and antimicrobial agent for paper coatings. A study by Rhim et al. (2013) demonstrated that paper coated with chitosan exhibited not only improved water resistance (contact angle  $>90^\circ$ ) but also antimicrobial activity against *E. coli* and *S. aureus*, making it suitable for food packaging [15]. The addition of beeswax or essential oils has also been explored to further enhance barrier properties [16].

### Starch-Based Coatings

Enhanced with Crosslinkers Native starch is hydrophilic, limiting its direct use in water-repellent coatings. Recent advancements have focused on chemically modified starches, such as oxidized or hydroxypropylated starch, often crosslinked with citric acid, borax, or glyoxal [17]. For instance, Xu et al. (2018) achieved a 65% reduction in water absorptiveness (Cobb value) using crosslinked corn starch films [18].

### Protein-Based Coatings

Zein and Soy Zein, a corn protein, forms hydrophobic films and has been used to coat kraft paper. Compared to uncoated samples, zein-coated papers demonstrated up to 50% reduction in water vapor transmission rate (WVTR) [19]. Soy protein isolates, when blended with nanoclays or lipids, showed improved flexibility and water resistance while retaining biodegradability [20].

### Hybrid and Composite Coatings

Several studies have integrated multiple polymers for synergistic effects. For example, a blend of cellulose nanofibers (CNF) and carboxymethyl cellulose (CMC) with chitosan created a robust coating with low water vapor permeability and good adhesion to paper substrates [21]. These hybrid coatings have applications in barrier packaging, disposable cups, and eco-friendly labels.

### Nanotechnology

Integration Nanoparticles (e.g., nano-silica, nanoclay, or silver nanoparticles) are being incorporated into biopolymer matrices to enhance coating performance. Nanoclay in particular reduces porosity and improves thermal stability and water resistance [22]. Such combinations also offer potential for smart coatings, such as moisture-indicating labels or self-healing surfaces [23].

### Recent Trends in Industrial Application

Recent commercialization efforts have been seen in Europe and Japan, where companies are replacing polyethylene coatings in paper cups and wraps with biodegradable alternatives such as PLA-chitosan or CMC-based films [24]. These papers pass regulatory standards (e.g., FDA/EFSA for food contact) while being compostable under industrial conditions.

## Challenges, Standardization, and Future Prospects

Despite significant progress in the development of natural polymer-based water-resistant paper coatings, several scientific, economic, and industrial challenges remain. Addressing these issues is essential for wider commercial adoption and for achieving environmentally sustainable paper products.

### Coating Uniformity and Adhesion

Natural polymers such as starch, chitosan, and proteins often face difficulties in achieving uniform and defect-free coatings, especially on high-speed commercial paper machines. This is due to:

- Inherent hydrophilicity of many natural polymers, which leads to poor wetting and adhesion on cellulose surfaces.
- Incompatibility with certain pigments, fillers, or inks, especially in multilayer coated boards.
- Need for surface modification of paper substrates or polymer functionalization, which adds complexity and cost.

Efforts to overcome these limitations include enzymatic treatment of fibers, crosslinking (e.g., with citric acid or genipin), and hybrid coatings incorporating natural waxes or nanoclays for improved barrier and adhesion properties [25,26].

### Coating Performance vs. Cost Trade-offs

While natural polymers are biodegradable and often derived from renewable sources, they frequently suffer from inferior performance compared to conventional coatings such as polyethylene, especially in:

- Water vapor resistance (WVTR)
- Grease/oil resistance
- Heat sealing and flexibility

Moreover, many bio-based coatings require additional plasticizers, emulsifiers, or crosslinkers, raising the formulation and processing costs. For example, pure chitosan coatings offer excellent antimicrobial and water resistance but remain cost-prohibitive for many applications [15]. Balancing cost-efficiency with functionality is a major challenge for industry adoption.

### Lack of Standardization and Regulatory Clarity

There is currently no universal standard for evaluating and certifying natural polymer-based coatings for paper products. Aspects needing standardization include:

- Test methods for biodegradability, migration, and barrier properties
- Regulations concerning food contact safety
- Certification for compostability and eco-labels

International harmonization of standards through organizations like ISO, ASTM, BIS, and FDA is crucial for market trust and global trade of such coated papers.

### Scalability and Processing Constraints

The scale-up of natural polymer coatings faces multiple bottlenecks:

- High viscosity and gelation of some bio-based formulations (e.g., alginate, gelatin), making them unsuitable for high-speed coaters.
- Sensitivity to moisture, temperature, and pH during storage and application.
- Incompatibility with existing PE/solvent coating lines, requiring specialized equipment.

Recent work on nanoemulsions, spray-drying, and extrusion coating with biopolymer blends is promising for enhancing industrial adaptability [27].

### Industrial Integration and Smart Functional Coatings

The integration of natural polymer coatings into industrial paper production faces multiple challenges but holds significant potential for sustainable innovation. Incorporating Industry 4.0 technologies, such as real-time data monitoring, digital twins, and AI-guided formulation optimization, enhances reproducibility and scalability of biopolymer coating processes. These advances enable adaptive control over coating parameters (e.g., polymer concentration, drying temperature) to achieve uniform layer formation and optimal barrier performance in high-speed manufacturing settings. Moreover, smart functional coatings represent a growing research frontier. Natural polymer coatings can be engineered to provide responsive properties such as pH-sensitivity, moisture indication, and antimicrobial activity, enabling active and intelligent paper packaging solutions. For example, incorporating natural pigments allows real-time detection of food spoilage through visible color changes, while moisture-sensitive agents provide visual cues for packaging integrity. The synergy of industrial integration with smart functionality aims to meet regulatory and market demands for sustainable, high-performance paper-based materials. Collaborations between industry, academia, and regulatory bodies are essential to develop standardized protocols, intelligent quality control systems, and multi-functional coatings that support both environmental sustainability and product performance [35, 36].

### Market Adoption and Consumer Awareness

Finally, large-scale market penetration will depend on:

- Consumer willingness to pay a premium for green products.
- Government incentives, eco-tax reductions, and stricter bans on plastics.
- Brand endorsement by major packaging and e-commerce companies.

As climate change and plastic pollution become urgent concerns, public and policy support for sustainable materials like bio-coated paper will likely grow. While several scientific and technical obstacles remain, advances in material science, nanotechnology, and process engineering are rapidly bridging the performance gap between natural and synthetic coatings. A collaborative approach involving academia, industry, regulatory bodies, and end-users is essential to ensure scalable, economically viable, and environmentally benign solutions. Despite the encouraging progress in utilizing natural polymers for water-resistant paper coatings, several technical and industrial challenges persist. These limitations span from material performance issues to process standardization and integration into advanced manufacturing systems.

## Coating Cost vs. Performance Trade-offs

One of the major challenges is balancing performance with cost-effectiveness. Natural polymers such as chitosan, starch, alginate, and cellulose derivatives, while biodegradable and sustainable, often exhibit variable barrier properties depending on molecular weight, degree of substitution, and blend compatibility [28,29]. For example, chitosan-based coatings provide excellent water vapor resistance but are significantly more expensive than conventional synthetic coatings [30]. Moreover, some natural polymers require chemical cross-linking or blending with additives to achieve desirable hydrophobicity, which increases formulation complexity and processing costs [31]. The scalability of natural polymer-based formulations is another concern. Uniform dispersion and consistent coating thickness on industrial paper machines are more difficult to control for bio-based formulations, especially under high-speed coating conditions [32]. These trade-offs between eco-friendliness, water resistance, and operational efficiency must be optimized to facilitate commercial adoption.

## Lack of Standardization and Regulatory Frameworks

There is currently no universal standard for evaluating the performance of water-resistant coatings derived from natural polymers. Variability in testing methods such as Cobb sizing, water contact angle, and water vapor transmission rate across laboratories limits comparability of results [33]. Furthermore, no globally accepted certification exists for “biopolymer-coated” or “green-coated” paper products. This hampers consumer awareness and limits market differentiation based on sustainable attributes [34]. Developing standard testing protocols and certification systems specific to natural polymer coatings is critical. Collaborative efforts between academic institutions, paper industry stakeholders, and regulatory bodies (such as ASTM, ISO, and BIS) will play a pivotal role in this direction.

## Future Research Directions

To overcome current limitations, future research should focus on:

- Molecular tailoring of biopolymers for improved hydrophobicity.
- Hybrid systems (natural polymer + nano-fillers or waxes) for superior barrier properties.
- Biorefinery-based production of coating materials to reduce dependency on food-based sources.
- Life cycle assessment (LCA) and techno-economic analysis (TEA) to validate sustainability claims.

With the global push for plastic-free packaging, the commercialization potential of natural polymer-coated paper is significant. However, the path forward demands interdisciplinary innovation and policy support.

## Biodegradability and Environmental Impact

The environmental impact and end-of-life behavior of water-resistant papers made with natural polymers is a critical factor determining their long-term viability and acceptance. Natural polymer-based coatings are gaining traction because they align with circular economy goals, reduce plastic pollution, and support sustainable development targets.

*Comparative Life Cycle Assessment (LCA) vs Synthetic Coatings:* Life cycle assessment studies have demonstrated that biopolymer-based coatings such as starch, chitosan, and PLA (polylactic acid) significantly outperform synthetic coatings in terms of environmental footprint. For instance, Pires et al. [37] noted that chitosan-coated papers exhibited a 45–60% reduction in carbon footprint and non-renewable energy use compared to polyethylene or fluorochemical-coated alternatives. Similarly, Hubbe et al. [1] emphasized that the use of nanocellulose and polysaccharide-based coatings substantially reduces the total environmental burden when considering cradle-to-grave scenarios.

*Compostability and Recyclability:* Many natural polymers such as starch, chitosan, and proteins are inherently biodegradable and compostable under industrial or home composting conditions. Kumar et al. [38] observed that starch-chitosan coated papers degraded by over 80% within 30 days in composting trials, meeting ASTM D6400 and ISO 17088 standards. Furthermore, the recyclability of such coatings does not interfere significantly with pulping operations—a key barrier often encountered with

## Conclusion

The development of water-resistant paper using natural polymers presents a promising and sustainable alternative to traditional synthetic coatings such as waxes, fluoropolymers, and plastics. This review has explored the various types of biopolymers such as starch, chitosan, alginate, cellulose derivatives, and lignin that exhibit potential as eco-friendly coating agents. These polymers provide varying degrees of hydrophobicity and barrier properties, often improved by crosslinking, nanofillers, or blending techniques. A critical evaluation of coating techniques, including dip coating, bar coating, spray coating, and novel extrusion-based methods, highlights the importance of scalability, uniformity, and adhesion in industrial applications. Characterization tools such as Cobb test, water contact angle, SEM, FTIR, and WVTR are essential for assessing coating effectiveness and ensuring reproducibility. The transition towards bio-based water-resistant coatings is propelled by environmental concerns, regulatory pressure to reduce plastic use, and the increasing demand for biodegradable packaging. However, challenges remain in terms of cost-performance optimization, lack of standardized evaluation metrics, and limited industrial-scale implementations. The integration of Industry 4.0 technologies, intelligent quality control systems, and smart packaging features offers exciting prospects for next-generation sustainable paper coatings. Future research should focus on molecular modification of biopolymers to enhance hydrophobicity, development of hybrid coating systems, and conducting life cycle assessments to validate environmental claims. The creation of international standards and certification schemes for green-coated paper products will be instrumental in encouraging widespread adoption. In conclusion, the development of water-resistant paper using natural polymers is not only a technically feasible and environmentally preferable solution but also an essential step towards achieving circular economy goals in the paper and packaging industries.

## Abbreviations

LCA – Life Cycle Assessment  
PLA – Polylactic Acid

CNF – Cellulose Nanofiber  
 CMC – Carboxymethyl Cellulose  
 WVTR – Water Vapor Transmission Rate  
 OTR – Oxygen Transmission Rate  
 PE – Polyethylene  
 ASTM – American Society for Testing and Materials  
 ISO – International Organization for Standardization  
 FDA – Food and Drug Administration  
 EFSA – European Food Safety Authority

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