

# Plastic-Free Bioactive Paper Coatings, Way to Next-Generation Sustainable Paper Packaging Application: A Review

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## Abstract

Hydrocarbon-derived polymers have been utilized in various packaging applications, such as pouches, films, foamed containers, rigid containers, and multiple components for medical, food, and other uses. However, mounting environmental considerations increased knowledge of the harmful consequences of greenhouse gas emissions, landfills, and disposal difficulties. Rising oil prices are forcing researchers and businesses to produce environmentally friendly packaging. These new sustainability requirements are particularly suited to biomass-based products, instead of petroleum sources; sourced from biomass entities. More functional and performance-oriented packaging is necessary despite the widespread usage of bio-based materials like paper. As a result, the transition to eco-friendly packaging will necessitate the improvement of existing bio-derived packaging and the development of new bio-derived materials like biopolymer paper coatings. The goal of this brief study was to give a synopsis of the present status of bio-derived packaging and an insight into ongoing and prospective developments in sustainable next-generation paper coatings for the packaging industry.

## Keywords

Biopolymers, Paper, Coatings, Starch, Itaconic Acid

## 1. Introduction Paper Coating

Because it is compostable and fully safe for the environment, paper is often utilized in packaging. Paper is a low-cost, renewable material that can be used in place of plastics in situations where short-term use is preferred. Because of their fibrous structure, paper substrates are porous [1]. Furthermore, the cellulose

backbone chain contains hydroxyl groups, making the paper very hydrophilic [2]. Because hydrophilic hydroxyl groups and porous structures absorb water and grease substances, uncoated paper is less valuable when plastic replacement is required [3] [4]. However, there is a scarcity of hydrophobic and oleophobic developments in the paper packaging business. As a result, there is an urgent need to develop strategies for enhancing paper's grease and water resistance while managing to stay recyclable and cost-effective [3] [5] [6]. High surface energy coatings also improved oil resistance due to a larger quantity of polar components, resulting in stronger grease resistance. After applying the coating, the mechanical characteristics of the paper improved as well. These naturally generated compounds provide an alternative to the fluoride-containing materials currently utilized in the market to increase paper wettability [7]. To address the aforementioned concerns, the coating industry today uses hydrocarbon-based polymers such as polyvinylidene chloride, low-density polyethylene, ethylene acrylic acid, and others to produce oil- and water-resistance paper via surface coating [8]. Due to their low recyclability in current practices, alternatives to present procedures are in great demand [9]. Other extensively used coating techniques include fluorinated compounds, which negatively impact human health and aquatic habitats [10].

Given that, food and packaging containers account for around 45% of trash shipped to landfills, with papers and paper products accounting for the vast majority, these chemicals voice issues about microplastic contamination [11]. As health and environmental concerns regarding wax-based, fluorochemical-based, and extrusion-based paper coatings have grown, so has an interest in bioresorbable and repulpable substitutes for generating water- and oil-repellent coatings. This work describes a plastic-free, fluorine-free, and cost-effective grease and water-resistant paper coating process that uses novel chitosan-graft-polydimethylsiloxane copolymer and corn starch blends [12]. Scientists are presently focused on bio-based substances like chitosan, poly [lactic acid], wax-based, and protein-based (e.g., whey protein, and casein) and endow paper with specific properties like improved one's liquid-repugnancy or gas-barrier performances. However, because of several complexities associated with modifying these materials while preserving water impermeability features, these attempts have had limited success, particularly in controlling water blocking capabilities and the high price of the alternatives available [9].

## 2. Biobased Paper Coating

Global plastic manufacture has grown quickly since the 1950s, generating over 450 million tons of annual output. Despite the benefits and great value provided by plastics, environmental impacts over plastic waste have attracted international attention. Over 340 million tonnes of waste were generated worldwide, with the packaging industry accounting for approximately 46% of the total [13]. This waste is primarily the consequence of things with a shorter "in-use" life-

span, often six months or less, contrary to items used in the construction and engineering sectors, with an average lifespan of thirty years [14]. Even though plastic recycling rates have increased from 0% in the nineties to 19.5% in recent estimates, most packaging materials, particularly pliable packaged food, are non-recyclable. Roughly 95% of conventional plastic wrapping materials made of polyethylene terephthalate and polyolefins are non-recyclable and end up in landfills after one use, causing a yearly economic loss of \$80 - 120 billion to the world economy [15]. These difficulties are induced by polymers' required characteristics, including lighter weight, specific strength, intricate structures, and contaminating from physical touch with foodstuffs [16]. Because of their ecologically favourable compostability, biodegradable polymers have sparked tremendous scientific and industrial interest. For the benefit of the market economy and recurrent environmental risks, biodegradable materials should play a larger part in plastic packaging, which today account for 60% of plastic items. The development of biodegradable polymers provides a solution to these problems by degrading such plastic into CO<sub>2</sub>, H<sub>2</sub>O, and biomass within six months; however, conventional synthetic plastic materials, even after burning or landfilling, remain for millennia. With the rise of processed foods in modern society, food packaging has played an essential part in the packaging sector. By 2025, the global market is anticipated to be worth USD 411.3 billion [17]. Because of its strong barrier and gas selectivity, plastic packaging provides food stability, shelf-life extension, and safety and protection during transit and storage. Biodegradable polymers that can be degraded or composted after use are excellent substitutes for non-biodegradable packing. It can also work as a selective functional membrane or barrier against gas, humidity, and odour. Although several scientific research attempts to encourage the use of bio-based polymers in packing, few bio-based polymers in the market can meet modern society's high demand for food packaging [18].

### 3. Fillers in Biobased Coatings

Because many bio-derived polymers are hydrophilic, their gaseous barrier properties and engineering capabilities are incredibly reliant on ambient humidity, limiting their usefulness as a packaging material compared to conventional polymers. Furthermore, their molar mass, rheological properties, and mechanical characteristics such as time-dependent crystallization behavior might cause problems and necessitate adjustments in processing techniques. As a result, bio-derived polymers should be blended with other polymer matrices or micrometers to nanometer-scale fillers to improve hydrophobicity and processing efficacy. The use of nanotechnology in papermaking science has been introduced in some areas. It has the potential to enhance the behavior of biopolymers and provide new capabilities to paper coatings. Currently, inorganic pigments [19], minerals [20], and ceramics [21] are the most often utilized nano-scale fillers in paper coatings, with bio-derived nanomaterial such as Nanocellulose [22] and

Nanoclays [23] being studied frequently. Antimicrobial paper [24], microfluidic paper devices [25], bioactive papers for drug delivery [26], flame-retardant papers, and self-healing characteristics for cotton fabric [27] are only a few of the fascinating functions achieved by using nanoparticles.

## 4. Bio-Polymers for Paper Coatings

### 4.1. Polylactic Acid

Paper is a biodegradable and hence ecologically beneficial material commonly utilized in packaging applications. The use of hydrocarbon derivatives as the coating, such as polyethylene, waxes, and fluor-derivatives, frequently regulates papers' barrier resistance and wettability. Because of this coated layer, protective paper packaging loses its biodegradability and recyclability. Because of their poor recyclability and non-biodegradability, petroleum-based polymers account for most world trash. As an alternative, organically renewable biopolymers can be used as barrier coatings over paper packaging materials. Polylactide (PLA) is among the most promising polymers due to its bio-compatibility, bio-degradability, and ability to produce bio-based feedstocks [28].

### 4.2. Itaconic Acid

It was suggested that polyvinyl alcohol (PVA)/Itaconic acid (IA)/acrylamide (AM) might generate a hydrophobic protective coating with a spatial structural framework on the paper's surface, increasing the count of hydrogen bonds amongst the copolymer and the fiber. Further, when coated on the paper's surface, the copolymer exhibited a superior film feature and a decreased penetration capacity through the paper compared to PVA. The strength of the paper reduces as the number of recycled paper increases. Surface scaling was widely recognized as the most efficient approach for improving the surface strength and hydrophobicity of paper and board [29] [30] [31] [32]. Starch and its derivatives were employed in the paper business for many years. Because of its low cost and ubiquitous availability, starch has indeed been recognized as among the most acceptable renewable biopolymer materials. Enzyme-modified starch, for example, is often employed to improve the interfacial strength of paper. Excess starch, on the other hand, may increase chemical oxygen demand (COD) and biological oxygen demand (BOD) during the effluent treatment process of the paper industry [33] [34] [35] [36] [37]. PVA was created as a biodegradable material by polymerizing vinyl acetate monomer. This provided the paper with large interfacial strength, low ink uptake, and exceptional film-forming capabilities. Nonetheless, the water-resistance of the bio-based PVA film had been lower than that of petroleum-derived polymers such as polyamide epoxy epichlorohydrin (PAE) [38] [39] [40] [41].

Meanwhile, PVA may easily infiltrate the inside of the paper. As a result, it limited PVA used in paper production. As a result, the weakness of PVA should be accounted for this through grafting with some other synthetic biodegradable

polymers. Many chemical strategies have been discovered to increase PVA's water resistance. The water-resistance of PVA has been enhanced via grafting of acrylonitrile monomer with PVA. Some studies developed PVA/polyacrylic acid mixtures to reduce PVA's hydrophilic characteristic [42]-[52]. A cross-linked esterification process occurred between polyacrylic acid and PVA molecules. PVA molecules' hydrophilic hydroxyl groups were transformed into hydrophobic groups. As a result, the water resistance of the PVA film was substantially enhanced. Biodegradable materials have gained popularity as a result of environmental concerns. One of the merits of IA is renewability. Furthermore, it has the potential to reduce people's dependency on petrochemical products such as acrylic acid and maleic anhydride when picking raw materials for polymer synthesis [53]-[58]. This study used free radical polymerization to produce a PVA/IA/AM copolymer having excellent water resistance and durability. PVA's hydroxyl groups can create ester bonds with IA molecules' carboxyl groups. It can increase the hydrophobicity of PVA. The double bond of itaconic acid may polymerize with an acrylamide monomer to form a structural matrix. Thereby, this could restrict PVA diffusion into the interior of the paper. Mini-table software was used to design the PVA/IA/AM copolymer experiment. It sought to enhance PVA's hydrophobicity, increase surface strength, and decrease PVA penetration. PVA/IA/AM can establish additional hydrogen bonding with fibers and has better film-forming characteristics over PVA [59] [60].

### 4.3. Starch

Starch and its compounds have indeed been largely used in the paper industry because of its complete biodegradability, abundant availability, and relatively inexpensive. Surface coating of starch-derived goods is a well-established commercialized procedure for acquainting paper with desirable qualities. As a raw material, starch is exceptionally versatile. First, starch is a natural polymer with a large molecular mass that can be precisely depolymerized. It is also a hydrophilic polymer, meaning it dissolves in water and creates hydrogen bonds with cellulose fibers and pigment. Third, starch contains hydroxyl groups, which allow for a variety of substitution or oxidation methods to change its rheological characteristics and prevent retrogradation. Starch graft co-polymerization can generate innovative materials that combine the advantages of natural and synthesized polymers [32] [61]-[68]. Coated paper is a form of paper that consists of a base coat and a top layer. The coat color is applied to the base paper during the coating process to improve its qualities. Coat color is primarily made up of binders, pigments, and other additives. Binders are the second most common coating component after pigments [69]. Natural binders including starch and protein or synthesized latex such as styrene-butadiene, poly (vinyl acetate), and poly-acrylates are employed. When compared to synthetic latex, starch is a relatively low-priced binder. However, nature-derived starch applications are restricted because of its insolubility in cold water, tendency to retrograde, and decrease in viscosity and thickening power during cooking and storage [70]. However, too

much starch in a coating formulation might reduce drying rate [71] [72], reduce picking resistance [73], and maximize cracking area [74] [75]. The ultimate skill is finding a middle ground between quality and price. Starch grafted copolymers can be used instead of starch and synthesized latex. Graft co-polymerization is indeed a flexible method for combining the characteristics of starch and synthetic polymers [76] [77].

On the other hand, starch is a biopolymer with a large molecular mass and a high viscosity in an aqueous medium at low concentrations. The starch's molecular weight must be decreased before it may be adequately grafted in co-polymerization [78]. Starch-based bio latex is a newly developed water-swollen, a cross-linked starch nanoparticle that commercially replaces synthetic latex [79] [80] [81]. Biolatex is sometimes referred to as starch-based bio-latex, biobased latex, bio binder, and a bio latex binder. Biolatex is a huge step forward in starch-based resins for paper coatings. On an industrial scale, bio latex can try replacing hydrocarbon-based synthesized latex by up to 50% or more [82]. For that reason, biolatex has enormous potential in the manufacturing of coated paper. A polymer blend is the physical mixing of two different polymers that may or may not interact chemically [83]. Compared to pure polysaccharide components, blends of multiple polysaccharides can form a new category of materials with improved mechanical and barrier properties. Starch/PVA mixes have been utilised to improve the characteristics of coated paper. A thin coating of starch/PVA film was applied to paper. The coated paper demonstrated good organic solvent barrier characteristics. The better barrier properties are due to hydrogen bonding formed by hydroxyl groups of starch and PVA molecules. The hydrogen bonding on the paper surface form a tight thin layer, which increases the barrier characteristics of starch/PVA films [84]. Ethylene vinyl alcohol and starch mixture Ethylene vinyl alcohol (EVOH) copolymer is a semi-crystalline synthetic random copolymer composed of ethylene and vinyl alcohol units [85]. The vinyl alcohol molecule has good air barrier qualities, whilst the ethylene component has great damp resistance, thermal and mechanical properties, and easy handling [86] [87] [88]. Furthermore, EVOH is more biodegradable than normal PVA [89]. Blends of EVOH and hydrophobically altered starch may increase the permeability performance of flexible packaging paper.

#### 4.4. Cellulose

Nanocelluloses, which have lately been developed, have provided a new polymer coating option. Micro-fibrillated cellulose (MFC) is being used as a coating pulp for paper products. The MFC manufacturing technique was discovered and patented in 1983 by Herrick *et al.* [90] and Turbak *et al.* [91]. MFC, a cellulose fiber subdivision, has diameters of 10 - 50 nm and lengths of several micrometers [92] [93]. MFC had significant reinforcing potential and was employed in nano-composites because of its high aspect ratio, nano-scale dimensions, entangled network, and intrinsically good mechanical characteristics [92].

Many researchers were intrigued by MFC's capacity to manufacture films because MFC films' barrier and mechanical properties are comparable with those of existing high-quality polymer matrices [94]. MFC coating and its mixture with petro- and bio-polymer films have attracted the attention of researchers by dramatically improving the barrier properties of the original films [95]. In contrast to these uses, the use of MFC with cellulosic materials is very new and remains uncommon. The first investigation of MFC coating onto paper was conducted in 2009 [94]. As ecologically friendly alternatives, many forms of cellulose nanofibres derived from cellulosic fibers have been used [96] [97] [98] [99].

Cellulose nanofiber (CNF) has been discovered to improve the properties of various composites and papers when used as a reinforcing element in wet-end applications [96]-[102]. Aulin *et al.* [103] demonstrated that utilizing CNF as a coating agent significantly reduces the air barrier capability of paper by generating a thick top layer. Hult *et al.* [101] investigated the mixture of CNF and shellac, deposited as a one-layer or multi-layer coating on paper products using a bar coater and a spray coating technique. The results revealed that paper permeability to water vapor, air, and oxygen decreased dramatically after coating. CNF improved the surface and barrier characteristics of coated sheets, according to Ridgway *et al.* [104]. The combination of CNF and clay improved the printing properties of paper, such as ink absorption rate and print density, according to Hamada *et al.* [105]. It was demonstrated that partially replacing NFC for the conventional co-binder carboxymethyl cellulose reduced the permeability of the coating material to water [106]. Andrade *et al.* studied the impact of several chemicals on the wettability of gelatin-derived edible coatings on banana and eggplant epicarps, discovering that cellulose-nanofibers increased coating wetting [107].

#### 4.5. Lignin

Papers made from recycled fibers have poor mechanical and barrier qualities due to insufficient inter-fiber bonding. The coating is a methodology of interest for creating appealing items from this type of paper. On 100% recycled paper, an oxypropylated lignin-based coating was investigated. Instead of propylene oxide, propylene carbonate, a non-volatile and biodegradable solvent, was employed as a reagent. One of them is the creation of lignin-based coatings for paper and paperboard applications, which is based on the hydrophobic property of lignin polymer [108] [109]. Some research has previously been conducted on bio-based coatings using cellulose derivatives, starch, and other polymers, [110] as well as a combination of lignin and cellulose nanocrystal or esterified lignin for paperboard use [110] [111] [112]. The subsequent research was of particular interest since they dealt with the esterification of lignin. Another method for lignin modification is oxypropylation, which has been extensively researched to produce novel polyol for polyurethane foam applications [112]. Lehnen *et al.* [113] investigated oxypropylation with propylene carbonate as a non-volatile and biodegradable solvent rather than propylene oxide, a possible human carcinogen.

An oxypropylated lignin-based coating was explored in this work to enhance the general characteristics of recycled paper. Propylene carbonate was employed as an oxypropylation reagent and a solvent in the coating formulation. This enables immediate use of the mixture at the end of the reaction, reducing the number of process stages to one.

#### 4.6. Soya

Protein-based biopolymer films have been researched for usage as food wrapping and coating materials. They've been frequently employed in food systems like oil, gaseous, and mechanical barriers to extending processed foods' shelf life [112] [113] [114]. Zein was used as an oxygen barrier layer in peanut products to avoid oxidation [115]. Casein coatings might also be utilized on less processed fruits to enhance their shelf life [116]. Trezza and Vergan [117] observed that zein-coated paper does have a high prospective for use as an oil-barrier paper bag in fast-food eateries. Isolated soy protein (ISP) also has a remarkable film-forming capacity [118] [119] [120]. Soy protein sheets have traditionally been used as a digestible covering for veg and meat mixtures that are then fried in oil. ISP-based films beat zein and wheat gluten-based films in terms of oxygen barrier performance [121]. Grease resistance is critical for packing items that include oils or fats.

Polyethylene and other polymers are considered effective grease barriers. On the other hand, the polyethylene content makes material segregation, recycling, and regeneration difficult [122] [123]. Papers coated with biodegradable polymers may meet packaging criteria as an oil barrier for commodities with a very short lifespan, like as fast-food restaurant sandwiches. ISP, a key by-product of soybean-oil extraction, on the other hand, is a low-cost natural polymer that competes against polyethylene. Mechanical and barrier performance of protein-based coatings are controlled by factors like plasticizer content and pH, solvent, and other additive choices [124]-[130]. As plasticizer content increases in wheat gluten films, flexibility improves while water vapor, oxygen barrier, and puncture strength qualities diminish [125] [126] [127]. Gennadios *et al.* [119] produced protein films from an ISP-wheat gluten blend. They investigated the impact of varying the pH of the produced film solution on properties such as water vapor permeability and mechanical strength. Brandenburg *et al.* [120] discovered that alkali-treated ISP films had enhanced film appearance and elongation characteristics. ISP films did not seem clear due to the inclusion of insoluble particles [128]. High pH and temperature treatments of ISP resulted in more soluble, hydrolyzed, and denatured chemicals [129]. Protein denaturation is known to change the structure of a protein from circular to extended chain geometry. Increased protein-protein linkages, in principle, would lower gas permeability while increasing tensile strength. Park *et al.* [130] revealed that different polyethylene glycol and glycerol plasticizer mixtures had a substantial influence on the mechanical properties and water vapor permeabilities of pro-



tein-based membranes.

## 5. Conclusions

Bio-polymer based coating on paper packaging materials are very important systems for the future improvement of food packaging. They have potential environmental advantages over conventional petroleum-based paper coatings. A literature survey indicates that bio-based polymers have a great potential for replacing hydrocarbon-produced synthetic polymers as a sustainable solution for paper coating applications. Not only can biopolymer paper coating provide biodegradability and renewability, but numerous methods for increasing the functioning of coated paper surfaces have been proposed. An appropriate mix of chosen ingredients (bio-derived polymer and fillers) and surface structure, according to the review, has the ability to offer an entirely impenetrable bio-based paper coating. From the presented review, appropriate bio-based polymers like starch, polylactic acid, itaconic acid, lignin, soybean protein and cellulose, provide a fully protective bio-based paper coating.

Finally, the difficulties associated with most biopolymers, such as hydrophilicity, crystallization behavior, brittleness, or melt instabilities, prevent their full industrial exploitation. This study cites investigations in which it was observed that mixing with other biopolymers, compatibilizers, and plasticizers might assist in alleviating this bottleneck. But extensively research will be needed in the paper packaging industry towards insubstantial materials for reduction of raw material use, conveying costs, minimizing the amount of waste and improvement in the performance properties of paper coatings. Interest in sustainable materials combined with improvement in performance properties of paper coating will continue to growth.

## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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## **Abbreviations**

PLA: Polylactide

PVA: Poly vinyl alcohol

IA: Itaconic acid

AM: Acrylamide

COD: Chemical oxygen demand

PAE: Polyamide epoxy epichlorohydrin

EVOH: Ethylene vinyl alcohol

MFC: Micro-fibrillated cellulose

CNF: Cellulose nanofiber

ISP: Isolated soy protein