

Review

Recent Research Progress of Polysaccharide Polymer Coatings for Improving Properties of Paper-Based Packaging Materials

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Abstract: With the increasing attention paid to environmental pollution, paper-based packaging materials have gradually gained favor among people. Paper-based materials are very environmentally friendly and renewable packaging materials. However, the mechanical properties and hydrophobicity of paper-based packaging materials are relatively poor, and they have high requirements for the usage environment and occasions. Therefore, the application of paper-based materials as packaging materials is greatly limited. Polysaccharide polymers, as coatings, have good biocompatibility and are environmentally friendly. They have certain potential in improving the hydrophobicity and mechanical properties of packaging materials. This review article introduces the four kinds of most used polysaccharide polymers, elaborates on their characteristics, and discusses their advantages in enhancing the performance of paper-based packaging materials. It also explores methods such as chemical modification to improve the hydrophobicity of polysaccharide polymers as coatings. Finally, this review discusses the combination of polysaccharide polymer coatings with paper-based packaging materials and provides prospects for the future.

Keywords: coating; polysaccharide polymer; packaging materials; chemical modification



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1. Introduction

In recent years, due to the gradual attention to environmental pollution, the safety and environmental friendliness of packaging materials are becoming more and more important. Currently commonly used packaging materials include plastic, cardboard, metal, glass and other materials [1,2]. Among them, plastic packaging is widely used in processing, transportation, storage and packaging because of its low price, high strength and corrosion resistance [3]. However, due to the characteristics of plastics that make them difficult to degrade, a large number of plastic products cause serious environmental pollution. In addition, the recycling of plastics is also limited for technical and economic reasons, resulting in a global plastic recycling rate of only 9%. Global plastic production is increasing year by year, and more than 90% of plastics are extracted and synthesized from fossil fuels, with less than 10% of plastics coming from mechanical recycling. Therefore, there is an urgent need to find a green, environmentally friendly, renewable and recyclable packaging material to replace plastics, which is one of the ways to solve the problem of plastic pollution [4].

Paper and paper-based materials are a light, recyclable and biodegradable green materials. They have opened up a new road for the green and sustainable development of packaging materials [5]. Paper is a kind of porous bio-based thin layer material with a three-dimensional network structure and excellent mechanical properties. Its main constituent element is fiber; the fiber surface is rich in a large number of active hydroxyl groups [6], which not only makes the paper show hydrophilic properties, but also because of the high reactivity of the hydroxyl group provides the possibility for the functional modification of paper [7]. Paper-based materials play an important role in the national economy and are widely used in many fields such as culture and education, packaging, construction and electricity. However, due to the large number of hydroxyl groups contained in cellulose, its main component, paper-based materials are more sensitive to moisture. In addition, its porous three-dimensional mesh structure also allows water to easily penetrate the surface of the paper [8,9]. This strong hydrophilicity greatly limits the application potential of paper-based materials in fields such as barrier packaging and oil–water separation.

In order to improve the properties of paper-based materials and broaden their application fields, more and more scholars are trying to use coating technology to enhance the barrier function of paper-based materials [10]. The former strategy involves the creation of a composite structure consisting of different organic layers with different functions, usually achieved through coating or bonding techniques. The main strategies adopted include the integration of multi-layer composite materials and inorganic nanomaterials. Among them, biopolymer coatings with biodegradability and biocompatibility are gradually being favored by scholars. This class of materials significantly improves the properties of paper by combining polysaccharide polymers that have been chemically modified or adding functional additives to the paper substrate. Commonly used polysaccharide polymers include natural cellulose, chitosan, starch and seaweed derivatives. Bio-based polymers have attracted much attention because of their unique versatility and specific chemical structure. They not only have good biodegradability and biocompatibility but also are easy to obtain. Starch and cellulose, in particular, have become the most popular choices due to their being abundant resources, renewable, low cost, and their excellent biodegradability and biocompatibility. Applying these bio-based polymers to cardboard or paperboard surfaces through a coating process can provide excellent antimicrobial properties, enhance mechanical strength and form an effective barrier against moisture, oil and oxygen on paper-based materials without increasing their weight. At present, the development of environmentally friendly paper-based materials with biopolymers as the main components has become a research hotspot and development trend in this field.

In this review, we focus on the mechanism of paper-based material coating barrier technology, as well as the classification of common polysaccharide polymer coatings, and finally we review the application types and research progress of polysaccharide polymers on paper-based packaging. This paper provides clues and ideas for further research on polysaccharide polymer-coated paper-based materials, provides a variety of solutions for improving the barrier properties of paper-based materials, puts forward the existing problems in the barrier aspects of polysaccharide polymers in the existing research, and the challenges facing the future.

2. Mechanism of Coating Barrier Technology

A coating is a continuous and closed film that is formed by evenly coating a specific material on the surface of the substrate, capable of binding tightly to the substrate [11]. The thickness of the coating is usually much smaller than that of the substrate, ranging from a few tenths of a nanometer to several microns [12]. The application of coating technology on paper-based materials can give it excellent barrier properties, effectively preventing gases

(such as oxygen, water vapor), liquids (such as water, grease) and other low molecular weight substances from penetrating into the substrate and diffusing. The rate and degree of diffusion of these substances not only depends on the physical and chemical structure of the coating, but also is closely related to the characteristics of the diffused substance itself and its interaction with the coating.

2.1. Hydrophobic Mechanism

The models of interaction between solid surfaces and droplets mainly include the Young model [13], Wenzel model [14] and Cassie–Baxter model [15]. Figure 1a–c show these three models, respectively. Among them, the Young model is only suitable for idealized smooth and flat surfaces. However, most solid surfaces in practice are not perfectly smooth, so the application of this model is limited. In order to solve this problem, the Wenzel model and Cassie–Baxter model were subsequently proposed. The Wenzel model states that increasing the roughness of a solid surface will enhance hydrophilic or hydrophobic properties, but its applicability is mainly concentrated in moderately hydrophilic and hydrophobic regions [16]. However, the Cassie–Baxter model emphasizes that under certain rough structural conditions, the contact between the liquid and the solid forms a composite interface, resulting in the inability of the liquid to fully wet the solid surface, resulting in superhydrophobic phenomena.

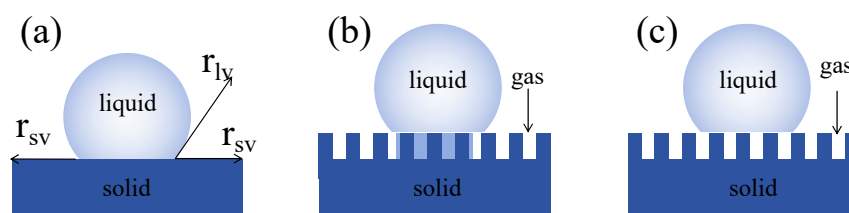


Figure 1. (a) Young model; (b) Wenzel model; (c) Cassie–Baxter model.

In practical applications, the preconditions of the above three models only hold true when the droplets are in equilibrium. However, due to the inhomogeneity of the real solid surface, the droplets will not usually be in an ideal state, so the contact angle hysteresis effect needs to be considered. At the gas–liquid–solid three phase contact line, the researchers define static friction as hysteresis resistance [17]. This resistance only causes the droplet to roll off when the solid surface reaches a certain angle of inclination, known as the rolling angle α . A smaller roll angle makes it easier for the droplet to roll off the solid surface [18], while a larger roll angle makes the roll more difficult.

For bio-based polysugar molecular chains, they often contain a large number of polar groups (for example, hydroxyl, amino and carboxylic groups) that exhibit strong hydrophilic properties [19]. When directly applied to paper-based materials, it is not only difficult to improve their hydrophobic properties, but may weaken their hydrophobic effects. Therefore, in order to improve this situation, it is usually necessary to perform hydrophobic modification treatment on polysaccharide polymers, which reduces their hydrophilicity by blocking these polar groups, thus enhancing the hydrophobic properties of paper-based materials. For example, Yang et al. [20] employed hydroxyethyl cellulose as the base material and introduced methyl methacrylate, butyl acrylate, glycidyl methacrylate and dodecyl methacrylate through a graft copolymerization process. This process led to the successful development of a hydrophobic-modified hydroxyethyl cellulose-methyl methacrylate copolymer emulsion, which was subsequently applied as a coating on paper surfaces. The findings revealed that this coating treatment significantly elevated the paper's water contact angle from an initial 0° to 114.2° , markedly improving its hydrophobic characteristics.

In addition to natural cellulose, nanocellulose, as a key hydrophobic modification material for cellulose, has also received extensive attention. Nanocellulose refers to cellulosic materials that are less than 100 nanometers in size in at least one dimension and can be divided into cellulose nanocrystals (CNCs), cellulose nanofibers (CNFs) and bacteria cellulose (BC) [21,22], according to their preparation methods and sources. Compared with traditional cellulose, nanocellulose not only retains its basic advantages, but also shows a larger specific surface area, stronger deformation resistance and higher Young's modulus and chemical activity [23]. Using the self-polymerization properties of tannic acid, Xiang et al. [24] successfully fixed octadecylamine (ODA) on the CNC surface through the Schiff base or Michael addition reaction between polytannic acid-coated CNC (PTA@CNC) and ODA and prepared an ODA-PTA@CNC composite material. After being dispersed in ethanol and sprayed on the surface of a paper substrate, the composite material can effectively impart superhydrophobic properties to the paper. The hydrophobicity of chitosan is closely related to its acetyl group content and molecular weight. Studies have shown that a higher degree of deacetylation and larger molecular weight can help improve the hydrophobicity of chitosan. Catto et al. [25] increased the acetyl content of 2% chitosan to 48% by acetylation treatment, and the treated chitosan-coated paper showed better hydrophobicity and mechanical properties. This is because the heterogeneous deacetylation treatment makes the molecular groups distributed in blocks, which reduces the solubility of chitosan in water.

Ion crosslinking is an effective means to improve the hydrophobicity of alginate. Alginate has a lot of negative charge; through the bridge between the anion and bivalent cation, the molecular chain can be tightly bound, thus enhancing the hydrophobicity of alginate coating. Zhu et al. [26] developed a unique coating with hydrophobic and oil-repellent properties for paper. This coating was composed of sodium alginate, hydroxypropyl methylcellulose, polyvinyl butyral and hydrophobic silica nanoparticles. Due to the hydrogen bond between sodium alginate and hydroxymethylcellulose, as well as the crosslinking between sodium alginate and Ca^{2+} ions in the base paper, a dense and tough coating is formed on the paper, which significantly improves the hydrophobic and oil-resistant properties of the paper.

2.2. Gas Barrier Mechanism

The molecular chains of polysaccharide-based polymers, including cellulose, starch, chitosan and alginate, contain an abundance of polar functional groups like hydroxyl, amino and carboxylic groups. These groups facilitate the formation of a coating that efficiently impedes oxygen permeation. It has been pointed out that this excellent oxygen blocking property is mainly due to the interaction between polar molecules [27], especially the formation of hydrogen bonds. Due to the highly hydrophilic nature of these polar functional groups, the polysaccharide polymer coating exhibits inadequate water vapor barrier properties, making it challenging to satisfy the water vapor barrier demands in food packaging applications.

The gas permeation process in the coating can be divided into four steps [28]: first, the gas molecules will be adsorbed on the surface of the coating; next, these molecules dissolve and enter the interior of the coating; they then spread through the coating; and finally, desorb from the other side of the coating. The process of small gas molecules passing through the film is shown in Figure 2. According to solution-diffusion theory, the permeability coefficient of a gas is the product of its solubility and diffusion coefficient, i.e., $P = S \times D$ (where P stands for permeability coefficient, S for solubility coefficient, and D for diffusion coefficient). These parameters are directly affected by the properties of the coating material itself, especially the cohesion energy density and free volume fraction. In

order to reduce gas permeability, the coating material needs to have a high cohesion energy density and a low free volume fraction.

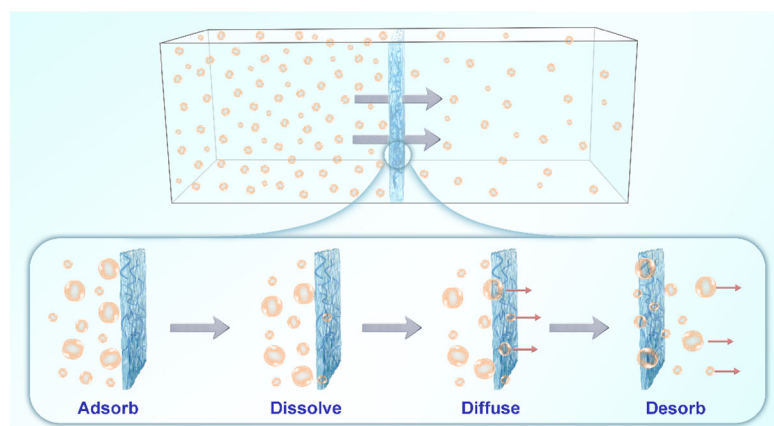


Figure 2. Process of small gas molecules passing through the film.

Although the permeability mechanism of water vapor and oxygen is similar, the actual performance is different due to the polarity difference between the two. For hydrophilic materials, they are more sensitive to water vapor, and the diffusion coefficient has a lot to do with the water vapor concentration, rather than a fixed value. In addition, the solubility of water vapor in polar materials is usually higher than that of non-polar materials, so water vapor permeates faster in polar materials.

In order to resist the gas, small molecules going through the polymer matrix, in addition to reducing the adsorption, dissolution and diffusion of the polysaccharide matrix, and in order to improve the barrier performance of the polysaccharide, vapor and oxygen can also be added to the polysaccharide polymer by increasing the transmission path of small molecules, reducing the diffusion coefficient of small molecules in the chitosan matrix. The nano packing curve path theory is a widely accepted theory that uses a composite material to improve performance. Shown in Figure 3 is the curve path model diagram of gas (water vapor and oxygen) through the poly nanocomposite.

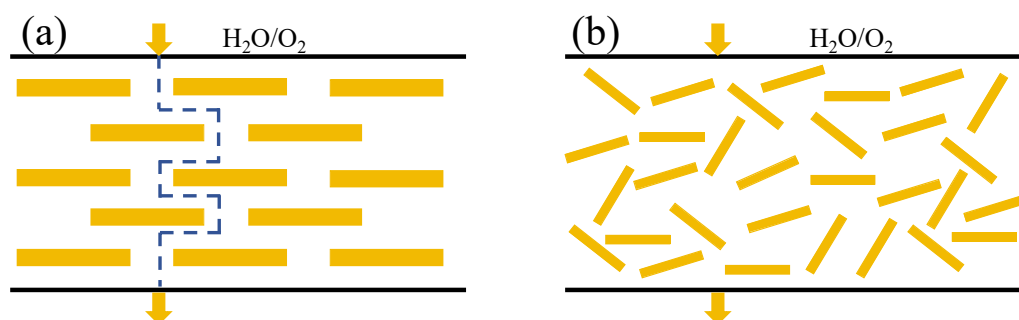


Figure 3. (a) Nielsen model; (b) Bharadwaj revised model.

For Liu et al. [29], the permeability rate of the uncoated paper-based material was $6.3 \mu\text{m}\cdot\text{pa}^{-1}\cdot\text{s}^{-1}$, while the permeability of the composite material using a chitosan-coated fiber paper-based material with only 0.5 g/m^2 was reduced to $0.86 \mu\text{m}\cdot\text{pa}^{-1}\cdot\text{s}^{-1}$.

Despond et al. [30] utilized paper-based materials, chitosan, and carnauba wax as the surface coating of the material; multilayer biodegradable materials capable of handling the gas barrier were prepared, and the effects of molecular weight and the concentration of chitosan solution on the barrier properties were investigated. The study showed that a layer of carnauba wax was deposited on the surface of the double layer of chitosan, and the water absorption of the multilayer film was reduced due to the hydrophobicity of the outer

layer. In the water and state, the permeability coefficients of carbon dioxide and oxygen are also greatly reduced.

In their study, Afra et al. [31] explored how CNF coatings alter the physical, mechanical and barrier characteristics of paper. They assessed the influence of various CNF concentrations and multiple coating applications. It was found that as the CNF concentration and the number of coating layers increased, there was a significant reduction in surface porosity. Additionally, higher CNF concentrations or more layers gradually improved air resistance, surface strength, stiffness and tensile strength. A key observation was that a single 3% CNF layer provided comparable mechanical and barrier performance to two layers of 1.5% CNF.

2.3. Antimicrobial Mechanism

The antimicrobial mechanism of polysaccharide polymer coating is mainly divided into the following: (1) inhibiting bacteria cell adhesion; (2) damaging or increasing the permeability of bacteria cell membranes and cell walls; (3) inhibiting the synthesis of bacteria nucleic acids and proteins; (4) affecting bacteria metabolism; (5) inhibiting biofilm formation or destroying the biofilm; and (6) hindering the absorption of nutrients. Figure 4 shows the antibacterial mechanism of the polysaccharide polymer coating. Six antimicrobial mechanisms are described below:

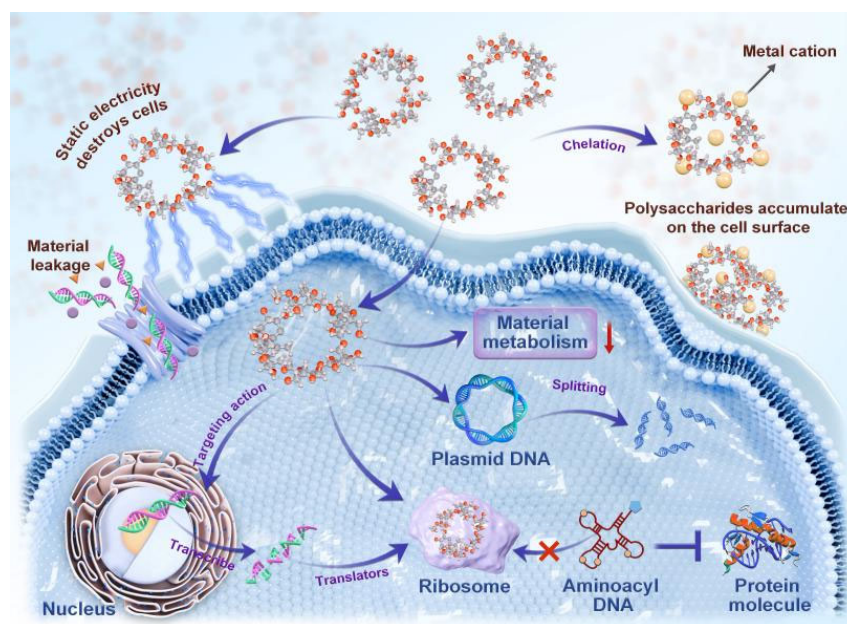


Figure 4. Antimicrobial mechanism of polysaccharide polymer coating.

2.3.1. Prevent Bacteria Cells from Sticking

The process by which bacteria infect their host depends on their adhesion to the surface tissues of the host. This binding process is typically facilitated by the interaction between carbohydrates and proteins, specifically between adhesins on the bacteria surface and those on the host cell surface [32]. Adhesins are among the bacteria virulence factors that play a crucial role in enabling bacteria to colonize within the host organism. Studies have shown that certain carbohydrate-rich polysaccharides have structures similar to host cell receptors and are able to bind to bacteria, thereby preventing adhesion of adhesins to host cell receptors and exerting significant antimicrobial effects.

2.3.2. Damage the Bacteria Cell Membrane or Cell Wall

Positively charged chitosan can interact electrostatically with anionic lipopolysaccharides in the outer membrane of *Gram-negative bacteria*, negative charges on the surface

of fungal cell membranes and lipoteichoic acid in the cell wall of *Gram-positive bacteria*, resulting in damage to the cell membrane or cell wall, thereby triggering intracellular component leakage and bacterial dysfunction, and eventually bacterial death [33].

2.3.3. Inhibition of Bacteria Nucleic Acid and Protein Synthesis

Polysaccharides can bind to bacteria DNA targets or ribosomes and interfere with the bacteria genetic system, affecting its replication, transcription, or translation processes, thereby inhibiting protein synthesis. Chen's research [34] has shown that fucosans with low molecular weight can form a high molecular membrane on the surface of cells, disrupt metabolism inside the bacteria, and may have destructive effects on DNA and RNA.

2.3.4. Interferes with Bacteria Metabolism

Studies have shown that polysaccharides can play an antimicrobial role by interfering with the material metabolism of bacteria, reducing toxin production or inhibiting bacteria growth. Recent studies by Ji et al. [35] have shown that the growth of *E. coli* is closely related to the concentration of 1, 6-diphosphate fructose (FBP). Triloban polysaccharides can interfere with the conversion of fructose 6-phosphate to FBP, resulting in lower than normal FBP concentrations, thereby inhibiting *E. coli* proliferation by impeding the glucose phosphorylation process [36].

2.3.5. Inhibiting or Destroying the Biofilm

Biofilms enhance the bacteria's tolerance to harsh environments and give them stronger drug resistance and the ability to fight off the immune system [37]. Research has demonstrated that galactans do not affect the initial adhesion of bacteria to surfaces; however, they play a significant role in inhibiting the accumulation of bacteria biomass. Furthermore, galactans not only prevent biofilm formation but also disrupt existing biofilms to some extent [38].

2.3.6. Hinder the Absorption of Nutrients

Polysaccharides can indirectly play an antimicrobial role by chelating with metal ions and hindering the absorption of nutrients by bacteria. Iron is an essential element for bacteria growth, and the stronger the iron binding ability of the modified polysaccharide [39], the more significant its antimicrobial effect. Chung et al. [40] found that chitosan extracted from agricultural waste can chelate with metal ions, affecting the uptake of nutrients by bacteria.

Due to the mechanism of the antimicrobial effect of polysaccharide coating, selecting the appropriate coating method is a challenge that researchers need to face. Due to the lack of clear indicators of response in antimicrobial testing, direct comparison of antimicrobial properties between different coating technologies is hindered. In addition, the long-term stability of the coating, the specific mechanism of killing bacteria and the antimicrobial test method are all problems that researchers need to solve in the future.

2.4. Oil Resistance Mechanism

Polysaccharide polymers, such as starch and chitosan, can effectively close the micropores on the surface of the paper, forming a continuous and dense protective film. This protective film prevents oil droplets from penetrating the paper through the spaces between the fibers, thus preventing the grease from coming into direct contact with the base paper and achieving an oil-proof effect. In addition, in an acidic environment, the amino group in chitosan has a positive charge, which can adsorb grease and form a soluble ion structure, inhibiting the absorption of oil by paper, while the hydroxyl group shows a repulsive effect on the oil.

Ham-Pichavant et al. [41] explored coating a chitosan solution on kraft paper and compared its effect with a fluorinated oil repellent. The results showed that when the concentration of chitosan solution was 3% and the coating amount was 0.884 g/m^2 , the oil-proof performance of kraft paper reached grade 10, and the highest oil-proof grade was grade 12, which was equivalent to that of fluorine-containing flooding, but the cost doubled. In order to reduce the production cost, the researchers further explored the mixed-use scheme of chitosan and sodium alginate. The experiment showed that with the concentration of chitosan solution at 47% percent and the amount of coating at 2.164 g/m^2 , the same oil-proof effect of class 10 can be achieved at half the cost of using chitosan alone.

A study by Jin et al. [42] showed that when carboxymethylated CNFs were applied to the surface of the paper, the oil-proof grade of the paper could be improved by one level when the amount was 0.3%. However, with the increase of coating amount or degree of substitution, the oil resistance grade did not continue to improve.

Jing et al. [43] developed a non-fluorophobic oil-resistant material consisting of collagen, chitosan and polydimethylsiloxane. The material is coated on paper in two layers; the first layer is a complex formed by crosslinking collagen and chitosan via glutaraldehyde, and the second layer is a polydimethicone coating. When the chitosan content is 30% and the polydimethicone mass fraction is 5%, the paper has an oil resistance rating of 12.

In their study, Sheng et al. [44] devised an environmentally friendly, oil-resistant paper that is free from fluorine and toxicity. This was achieved by employing a combination of SA, sodium carboxymethyl cellulose (CMC), and propylene glycol alginate ester. This grease-resistant paper achieves an oil resistance rating of 9. Moreover, the coating effectively forms a complete layer on the paper's surface while partially infiltrating the fiber network, thereby substantially improving the mechanical strength of the grease-resistant paper.

Kopacic et al. [45] coated primary fiber paper (PF) and secondary fiber paper (SF) with sodium alginate solution. The results showed that SF paper and PF paper had oil resistance ratings of 7 and 12, respectively. In contrast, PF and SF paper coated with chitosan solution had oil resistance grades of 6 and 5, respectively. Although chitosan can form a dense film on the surface of the paper, its oil resistance is not as good as sodium alginate, which may be because sodium alginate has a better hydrophilicity.

Shankar et al. [46] utilized alginate, CMC, carrageenan and grapefruit seed extract as base materials to develop antimicrobial wrapping paper. Their findings indicated that this innovative paper not only enhanced water and oil resistance but also exhibited significant antimicrobial performance.

3. Classification of Common Polysaccharide Polymer Coatings

Polysaccharide polymers are complex carbohydrates formed by the linkage of glycosidic bonds, and a variety of polysaccharides and their derivatives are currently being extensively studied to develop biodegradable paper-based composites (such as films and coatings). Common polysaccharide polymers include natural cellulose, chitosan, starch and seaweed derivatives (such as alginate). The molecular chains of these polysaccharides contain rich polar functional groups, such as hydroxyl, amino and carboxylic groups, which give their coatings excellent oxygen blocking properties. Studies have shown [47] that this excellent oxygen blocking property is mainly due to the interaction between polar molecules, such as the formation of hydrogen bonds. Figure 5 shows the application of four polysaccharide polymers and polymer coating on paper-based materials.

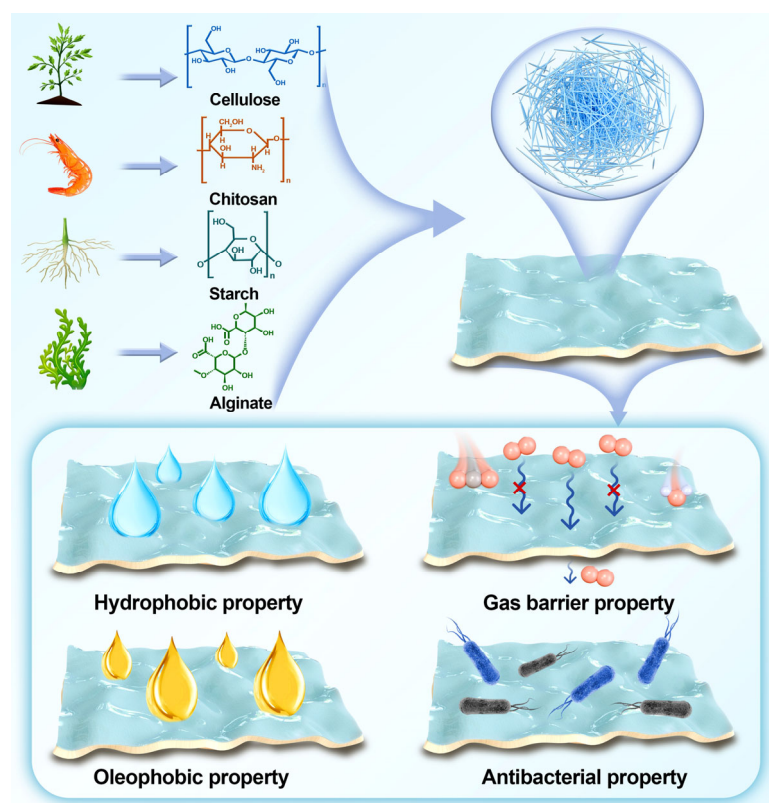


Figure 5. Four common polysaccharide polymers and their application as coatings in paper-based materials.

3.1. Natural Cellulose

Cellulose and its derivatives represent the most plentiful natural polymers on Earth, commonly occurring in the cell walls of plants. Figure 6a shows the structural formula of cellulose. These materials are produced in enormous quantities, with an estimated annual output of approximately 700 billion tons [48]. Cellulose is formed from β -D-glucopyranose rings connected by (1 $\rightarrow$ 4) glucoside bonds, and its molecular chain aggregation structure and hydrogen bonding give it high mechanical strength. However, because cellulose contains a large number of hydroxyl groups in the molecular chain, it is easy to adsorb water molecules, so it shows strong hydrophilicity. Cellulose is widely distributed in nature, for example, the cellulose content in wood is about 40%–50%, while the content in cotton is as high as 90%. In addition to plants, cellulose is found in a wide variety of organisms, such as algae, bacteria, fungi and even certain Marine animals such as tunicates.

Cellulose and its derivatives have received a lot of attention in the field of food packaging due to their excellent mechanical properties, low price and easy access, and being tasteless, odorless and biodegradable [49]. Cellulose-based derivatives, such as CMC, methyl cellulose, hydroxypropyl methyl cellulose and hydroxypropyl cellulose [50] exhibit properties like reduced density, enhanced mechanical performance, economic viability, non-toxic characteristics, biocompatibility, biodegradability [51] and excellent film-forming abilities [52]. These materials can create a tightly packed film on the surface of paper-based substrates, effectively preventing the penetration of non-polar gases, including oxygen (O_2) and carbon dioxide (CO_2). Marta et al. [53] found that when using BC as a coating on paper, air penetration can be completely blocked because the microbial fibers can fill the space between the plant cellulose fibers, creating a perfect barrier.

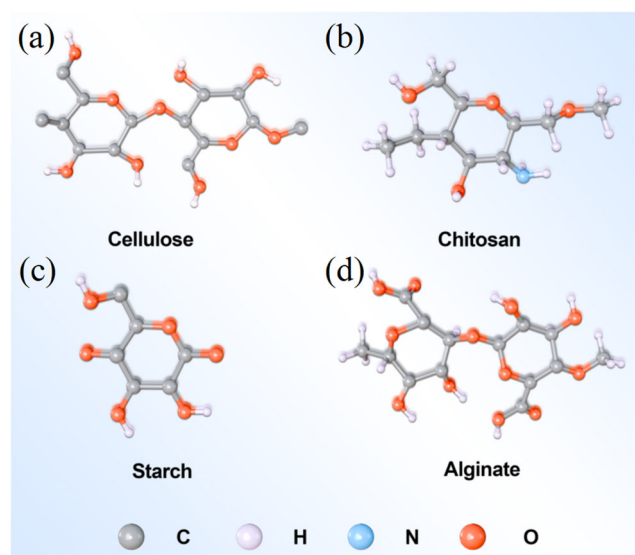


Figure 6. Structural formula: (a) cellulose; (b) chitosan; (c) starch; (d) alginate.

Among the derivatives of cellulose, CMC stands out as a linear, long-chain, water-soluble and anionic polysaccharide. Given its affordability, abundant sources, ease of pulping and superior film-forming properties, it has garnered significant attention for use in coatings applied to food wrapping paper. However, it lacks antimicrobial properties itself, so it is often used in combination with other antimicrobial agents. CMC itself is rich in hydroxyl and carboxylic groups, which can be crosslinked with metal ions with antimicrobial activity, thus acting as an effective carrier of antimicrobial substances.

Because cellulose is rich in hydroxyl, natural cellulose molecules without any treatment have certain hygroscopic properties, so the water vapor barrier performance is poor. Therefore, cellulose is usually modified and applied to the surface of paper-based materials as a coating. Yun et al. [54] introduced an innovative approach that utilizes cellulose-based paper as a substrate, involving the thermal fusion of ethylene-propylene side-by-side fibers and the application of a hydrophobic coating on the cellulose surface. Their findings demonstrated that, relative to pure cellulose paper, this method increased the water contact angle of the cellulose substrate from 25° to 153° and enhanced the wet tensile strength of the resulting composite by approximately 6.7 times. Notably, when compared with other related studies, the developed cellulose matrix composite exhibited superior hydrophobic properties and mechanical performance. This technique offers a novel chemical engineering solution for producing hydrophobic functional materials based on cellulose.

3.2. Chitosan

Chitin is mainly extracted from the shells of crustaceans (such as shrimp and crabs, etc.) and the exoskeletons of arthropods (such as insects), and can also be synthesized through the abiotic synthesis pathway catalyzed by chitinase. It is the second most abundant semi-crystalline polysaccharide in nature, second only to natural cellulose. Derived through the deacetylation of chitin, chitosan comprises 2-amino-2-deoxy- β -D-polyglucose monomers connected by (1 $\rightarrow$ 4) glycosidic linkages, as shown in Figure 6b. The presence of three nucleophilic groups within its structural units allows chitosan to undergo crosslinking, either chemically or physically, to form a gel and establish a network structure [55]. Soluble in acidic aqueous solution (amino group protonated reaction occurs in the medium with pH < 6.2), it is the only natural cationic polysaccharide at present [56], so it can inhibit the reproduction and metabolism of microorganisms and the integrity of the cell membrane, thus extending the freshness of food. Chitosan is a non-toxic, bio-

compatible and biodegradable material with good film-forming properties [57,58] and is widely used in medicine, biology, packaging and antimicrobial fields. Chitosan film or coating in low-humidity environmental conditions of gases (CO₂ and O₂) has good barrier properties and can maintain good mechanical properties; in conjunction with its inherent antimicrobial properties, it can extend the shelf life of food [59,60]. In addition, due to its excellent biocompatibility, biodegradability and film-forming performance, it has been included in the national standard for food additives and has become the main substrate for domestic and foreign scholars to study biodegradable food packaging [61,62]. Zhao et al. [63] used chitosan to make a plastic wrap with anticorrosive properties to extend the storage life of blueberries. Abdel-Naeem et al. [64] investigated the effectiveness of chitosan coatings on food preservation. They stored smoked herring with chitosan coating at -18°C for three months, performed microbiological, physicochemical and sensory analyses, and concluded that chitosan coating could be a good option for the fish industry to overcome problems in smoked fish. Lin et al. [65] developed a coating utilizing a composite aerogel made from chitosan, gelatin and microbeads, which they applied to E-flute corrugated cardboard. Their findings indicate that this coated cardboard exhibits remarkable antimicrobial and anticorrosive capabilities. Additionally, improvements were observed in the cardboard's mechanical strength, water-repellent properties and thermal insulation performance. Specifically, when the coating thickness reaches 30 μm , the tensile strength of the cardboard increases by 0.65 kN/m, the contact angle rises by 19.3° and the thermal conductivity decreases by 0.024 W/mK.

In addition, the chitosan coating also has good air insulation properties. Henrik et al. [66] coated greaseproof paper with different air permeability with chitosan to study the conditions needed to obtain packaging materials with good gas barriers on an experimental scale. They chose sand-proof paper as the base for the coating, which showed a good oxygen barrier and good lipid resistance, but slightly lower water resistance, when the coating weighed more than 5 g/m². Wang et al. [67] coated the paper-based material with a mixture of chitosan and montmorillonite, using montmorillonite to improve the oil resistance of the chitosan and reduce the cost. The study showed that montmorillonite improved the thermal stability and mechanical properties of the paper while filling the space between the paper fibers.

3.3. Starch

Starch, a biodegradable, renewable and relatively inexpensive material, is another ubiquitous bio-based polysaccharide [68]. Figure 6c shows the starch structural formula. This substance is commonly present in the roots, stems and seeds of crops. It has the advantages of easy access, high yield, rich nutritional value, low cost, good biodegradability, good biocompatibility and edibility. Globally, the main sources of starch are maize (82%), wheat (8%), potato (5%) and cassava (5%) [69]. Starch is a polysaccharide substance formed by the glucoside linkage of glucose units, and its main components include amylose and amylopectin.

Amylose has a linear structure, and glucose units are connected by α -1,4 glucoside bonds to form linear molecular chains, which makes it easy to form coatings, and it has good toughness film properties. Amylopectin, on the other hand, consists of glucose units connected by α -1,4 and α -1,6 glucoside bonds to form branched molecular chains [70], which show better stability in water, but whose mechanical properties are relatively weak when forming films or coatings. Therefore, the difference in the ratio of amylose to amylopectin will significantly affect the mechanical properties of the corresponding films and coatings [71]. In addition, the properties of starch are also affected by plant species, particle size and shape, and other factors [72].

Due to the composition and structure of starch, there are a large number of hydroxyl groups in starch molecular chains and a large number of hydrogen bonds between molecules, which will increase the interaction force between starch molecules. At the same time, the existence of a large number of hydroxyl groups on the starch molecular chain makes it highly hydrophilic, resulting in the starch being difficult to blend well with other hydrophobic polymers, so that the performance of the blend material is not ideal. Therefore, natural starch is usually modified to improve its hydrophobic properties and thermal processing properties. Starch modification methods mainly include physical modification, chemical modification, enzyme modification and compound modification [73].

Ni et al. [74] utilized CMC as a sealing agent for SiO₂. Owing to its superior flexibility and film-forming properties, CMC enhanced the compatibility between starch and SiO₂, while also improving the flexibility of the starch coating. Consequently, a highly flexible starch-based coating was successfully developed for food wrapping paper, exhibiting unique hydrophobic characteristics and antimicrobial properties. In the synthesized films and coated papers, there was a notable increase in both the value and stability of the dynamic contact angle, along with improved thermal stability of the composite films. Additionally, the water vapor transmission (WVP) rate was reduced.

The paper coated with dialdehyde starch (DAS) solution grafted with guanidine hydrochloride (GH) prepared by Liu et al. [75] showed that it had a good antimicrobial effect on *Escherichia coli* and *Staphylococcus aureus*. Ziaee et al. [76] grafted guanidine polymer (PHGH) onto potato starch, and by grafting reaction obtained guanidine-modified starch and coated it on paper. It was found that the PHGH starch-coated paper had good antimicrobial activity. Quilez-Molina et al. [77] tested active starch foams coated with anthocyanins and carboxymethyl cellulose, and citrate starch foams improved the water resistance and foaming properties of pure starch foams. At the same time, they provided efficient colorimetric pH sensing characteristics. With an excellent colorimetric pH sensing response, the coating foam can accurately detect shrimp deterioration. However, a single starch film has the disadvantages of poor mechanical properties and high water permeability; in order to improve its performance, researchers tried to combine other biopolymers such as chitosan, which improved the biodegradability, barrier, thermal and mechanical properties of the film, and promoted antioxidant and antimicrobial properties [78].

In addition, in order to improve the waterproof performance of the starch film, the starch can also be physically blended with the waterproof material. Ni et al. [79] mixed nanometer zinc oxide with starch solution and then applied coating treatment. Among them, the contact angle of the coating paper containing 2 wt% nano-zinc oxide increased from 64.47° to 103.94°, indicating that the nano-zinc oxide and starch composite can enhance the waterproof performance of the paper. In addition to the physical modification of starch, starch can also be chemically modified to improve the water resistance of paper. However, the chemicals used may have negative effects on the human body and the environment. In contrast, the enzymatic modification of starch shows a greater potential.

3.4. Seaweed Derivatives

Alginate is a natural polysaccharide mainly extracted and purified from different species of brown algae, and it can be used as the base component of polysaccharide-based composites [80]. It is a biodegradable, linear anionic water-soluble polysaccharide composed of β -D-mannuronic acid (M) and α -L-gulonuronic acid (G) linked by (1→4) glycosidic bonds to form a binary copolymer [81]. It has a linear chain structure similar to cellulose and is abundant in brown algae [82]. Figure 6d shows the alginate structural formula. It has a unique colloidal state when dissolved in water, and it has excellent properties, such as good film formation, high stability, biocompatibility, non-toxicity and

biodegradability, making sodium alginate [83] and its derivatives widely used in food additives and packaging fields.

Domestic and foreign studies have confirmed that sodium alginate can improve the oil-resistance [84,85] of paper and reduce the migration rate of organic volatile compounds [86] by filling the pores between cellulose paper-based materials. However, alginate has some problems, such as poor mechanical properties, a high swelling rate and poor water stability. Alginate reacts with bivalent and trivalent metal cations to form thin films, such as calcium, magnesium, manganese and aluminum; ferrous and iron ions can be cross-linked with alginate.

Li et al. [87] prepared a high-performance paper-based barrier material by using sodium alginate, chitosan and silica gel emulsion to form a barrier coating. Through the double coating, the waterproof and oil-proof functions are superimposed. The test results show that there is no chemical reaction between the paper-based coatings, and the surface of the coated paper is well formed. The double-layer method improves the thermal decomposition temperature of the overall barrier layer, effectively improves the thermal stability of the coating and achieves high barrier performance at a lower cost and under extreme working conditions.

4. Application Classification of Polysaccharide Polymer Coating for Improving the Performance of Paper-Based Packaging

4.1. Hydrophobic Properties

Paper is made up of plant fibers, and the hydrophilic nature of cellulose along with the porous surface of paper means that it inherently lacks hydrophobic and oil-repellent properties [88]. In order for paper to be used in food packaging applications, it needs to be modified [89]. The polar groups in the polysaccharide polymer give it good hydrophilicity, so it is usually necessary to carry out hydrophobic modification on the polysaccharide polymer (such as esterification, etherification, graft copolymerization and crosslinking, etc.), seal the polar groups, reduce the group hydrophilicity, and then further combine with the paper-based material, so that the hydrophobicity of the paper-based material can be improved. The hydrophobic oil-repellent coating developed based on the surface modification of paper is easy to use and has a wide application range, which has attracted more attention in the area of paper modification. Table 1 shows the application of polysaccharide polymer-coated paper-based materials to improve hydrophobicity.

Huang et al. [90] developed a polyvinyl alcohol (PVA)/CNF coating that can be used for paper surface modification through a composite method, and the coating has certain hydrophobic and oil-proof properties. The hydrophilic properties of PVA and CNF limit the further improvement of the hydrophobic properties of PVA/CNF coatings [47]. In general, a single polysaccharide is used in paper coating, and it often does not achieve the desired effect due to its structural limitations; mixing different polysaccharide components by physical or chemical methods can often achieve better results. Peng et al. [91] developed a single-step coating process for preparing paper with hydrophobic and oil-repellent properties. The process uses a templated methylene chloride aqueous solution (O/W type) to stabilize Pickering emulsions composed of CNC. Chitosan and glutinous rice starch (GRS) were added to the water phase, while polylactic acid (PLA) was added to the oil phase. Among them, chitosan and GRS are used as the main substrate materials of the coating, while hydrophobic PLA is embedded in the form of microspheres [92]. The optimized surface structure endows the coated paper with better hydrophobic properties. This innovative strategy offers a new sustainable path for the mass production of all-bio-based, recyclable, hydrophobic, oil-resistant, self-cleaning and biodegradable paper materials. Through the bionic structural design and the application of intelligent response

technology, the prepared coatings significantly improve the properties of the paper. For example, nanoscale silica particles are coated on the surface of the paper to form a micro-nano pore structure, which mimics the micro-nano papillae hierarchical structure in the lotus leaf effect, making the waterproof performance better than that of paper produced by the traditional direct coating method [93].

Tudor et al. [94] investigated the difference in waterproofing performance between chitosan derivatives and micro-fibrotic cellulose as a single-component coating. The results showed that alkyl chitosan increased hydrophobicity, while micro-fibrotic cellulose, carboxymethyl chitosan and quaternary chitosan coating increased the hydrophilicity of the surface of the paper. The water vapor transfer rate of carboxymethyl chitosan and alkyl chitosan coating decreased by 30% due to the presence of hydrophobic groups of alkyl chitosan. And the film structure of carboxymethyl chitosan is compact and homogeneous.

Table 1. Research on the application of polysaccharide polymer-coated paper-based materials to improve hydrophobicity.

Materials	Method	Features	Ref.
Cellulose nanofibers	Treat the bamboo fiber with biological enzymes, reduced fiber size and increased crystallinity.	The increase of cellulose nanofibers can effectively increase the hydrophobicity, hydrophobicity and tensile properties of paper-based materials.	[90]
Chitosan, glutinous rice starch	Chitosan and glutinous rice starch are aqueous phase, polylactic acid is oil phase and hydrophobic polylactic acid is embedded in the form of microspheres, mimicking the lotus leaf effect.	Coated paper has a high hydrophobicity and water barrier. The water contact angle is greater than 130, and the water vapor penetration rate is $3.57 \times 10^{10} \text{ g m}^{-1} \text{ s}^{-1} \text{ Pa}^{-1}$.	[91]
Chitosan derivatives and micro-fibrotic cellulose	Three water-soluble chitosan derivatives with specific functions were tested separately (alkyl chitosan-ach, season chitosan-qch and carboxymethyl chitosan-ch).	Alkyl chitosan alone or in combination with microfiber cellulose improves the barrier of water and water vapor; carboxymethyl chitosan improves the tensile strength; and seasonal chitosan has a general effect on the water barrier and strength performance.	[94]
Chitosan	Hydrophobic compounds grafted to chitosan.	The gas-phase barrier performance of chitosan-palm emulsion-coated paper remained unchanged, while chitosan-O, O'-double palmitoyl-chitosan-coated paper decreased significantly.	[95]
Hemicellulose	Hemicellulose-grafted lauric acid micelles were used as a nano-stabilizer.	The coating density was 8.4 g/m^2 , the water contact angle was about 130 and the waterproof time was 6 h.	[96]
Chitosan	The chitosan-grafted sunflower seed oil paper coating was prepared by chitosan open-ring method.	A good thermal stability at 250°C .	[97]

Bordenave et al. [95] applied coatings to two different types of paper using either a chitosan–palmitate emulsion or a blend of chitosan and O, O-dipalmitoyl chitosan (DPCT). Their research demonstrated that attaching hydrophobic compounds to chitosan facilitated better integration into the chitosan matrix, allowing deeper penetration into the paper structure. The findings indicated that both approaches effectively increased the hydrophobic properties of the paper.

Wang et al. [96] used hemicellulose-grafting lauric acid micelles as a nano-stabilizer to prepare a uniformly dispersed water-coated beeswax Pickering emulsion and coated the paper. The coating had a density of 8.4 g/m^2 , excellent waterproof performance, a water contact angle of about 130° and a waterproof time of 6 h.

Kansal et al. [97] developed a chitosan-grafted sunflower oil paper coating through partial epoxidation of the ring-opened sunflower oil and chitosan, resulting in a bio-based coating that exhibits excellent water and oil resistance. Thermogravimetric analysis revealed that the coated paper material demonstrated notable thermal stability at temperatures up to 250°C .

4.2. Gas Barrier Property

Cellulose is rich in a large number of hydrophilic hydroxyl groups, and the three-dimensional network structure formed by these hydroxyl groups is easy to cause a capillary effect, resulting in high hygroscopicity [98]. In addition, water vapor and oxygen in the air can be bonded through a fibrous porous network of paper-based materials. Once inside, these gases accelerate the physical, chemical and biological reactions of food, thereby shortening the shelf life of food. Table 2 shows the application research of polysaccharide polymer-coated paper-based materials for lifting the gas barrier. Shi et al. [99] used the excellent film-forming ability of chitosan and CMC to successfully prepare film materials with excellent mechanical properties. Chitosan itself has a good antimicrobial effect, and the rich carboxyl group on the surface of CMC has a negative charge, which can effectively prevent bacteria from attaching to the surface of the film. The chitosan/CMC multilayer film was constructed by continuous deposition of chitosan and CMC on the surface of the paper, thus enhancing the antimicrobial properties of the film. The air permeability and WVP test of the paper modified by chitosan/CMC multi-layer film showed that with the increase of the amount of chitosan/CMC multilayer film, the air permeability and WVP decreased gradually, and the air resistance increased significantly.

Jiang et al. [100] have successfully developed a new type of wrapping paper using a mixture of PLA and cinnamaldehyde (CIN) as a barrier screen and nano-silica modified stearic acid (SA/SiO₂) as the superhydrophobic coating. The cellulose wrapping paper shows good thermal stability, as PLA not only has excellent film-forming properties but also promotes the dissolution of CIN, enabling the antimicrobial agent CIN to be evenly distributed in the film. The interaction between PLA and SA at the multi-layer interface gives the paper hydrophobic properties, and the distribution of particles similar to the lotus leaf structure formed by SiO₂ nanoparticles on the surface of the paper further enhances this hydrophobic effect, thus giving the paper super-hydrophobic and anti-pollution capabilities. In addition, the dual-function coating changes the adsorption characteristics and transport paths of small molecules on its surface, showing excellent barrier properties to oil and natural gas. With the strong antimicrobial action of CIN, the composite paper achieves 100% antimicrobial efficiency, showing broad application prospects in the field of degradable food packaging materials.

Table 2. Research on the application of polysaccharide polymer-coated paper-based materials to lift the gas barrier.

Materials	Method	Features	Ref.
Polylactic acid	Cellulose paper as paper-based material, polylactic acid and cassia bark aldehyde as barrier layer, and nano-silica modified stearic acid as superhydrophobic layer.	With thermal stability, can block oil and natural gas, with hydrophobic, pollution prevention and antimicrobial functions.	[100]
Long chain stearic acid	Long-chain stearic acid modified the microcrystalline cellulose to form hydrophobic microcrystalline cellulose esters, and it was mixed with stearic acid.	The coated paper forms a continuous hydrophobic film, which completely covers the pores of the original bagasse fiber paper, and shows good water resistance and oxygen resistance activity. The water also showed good dimensional stability and good wet tensile strength.	[101]
Bacterial cellulose	Cellulose was used as a scaffold, and the ethylene-propylene fibers were coated on the cellulose surface.	Plant-bacterial cellulose fibers were fully filled, and hot-coated ES fibers increased the water contact angle of the cellulose scaffold from 25° to 153°, while increasing the wet tensile strength of the composite by about 6.7 times compared to pure cellulose paper.	[53]
Cellulose nanofibers	Foam coating was prepared from a mixture of anionic surfactants and cellulose nanofibers.	Low coating weight and high air barrier efficiency.	[102]
Cellulose	Carboxyethyl-pretreated bleached bamboo pulp.	After carboxyethyl pretreatment, the cellulose coating of bleached bamboo pulp had the highest efficiency.	[103]

Huang et al. [101] developed a hydrophobic microcrystalline cellulose ester modified with long-chain stearic acid. This material was blended with stearic acid and applied as a coating on paper, resulting in a modified substrate (MSP) that exhibited remarkable superhydrophobicity and oxygen barrier properties. The study demonstrated significant reductions in WVP rate and oxygen transmission rate for the coated MSP samples. In further studies, polyhexamylguanidine was chemically grafted to the free carboxyl group on the MSP surface. Using the antimicrobial mechanism of positive amino charge, the resulting antimicrobial paper demonstrated excellent antimicrobial activity. By using raspberries for fresh-keeping tests, the results showed that the experimental film had reduced transpiration and respiration through the oxygen barrier compared to commercial polyethylene film, and the freshness of raspberries after 3–5 days was significantly higher than that of polyethylene film packaging. The film showed better fresh-keeping performance of fresh fruit. When Marta et al. [53] utilized BC obtained from *Lactobacillus plantarum* cultures and applied it to cork, hardwood cellulose pulp or other types of paper, the resulting material became entirely airtight. This effect occurred because the microscopic microbial fibers occupied the spaces between the plant cellulose fibers, thereby preventing air from

passing through the layers, creating a natural composite that is completely filled with plant-bacterial cellulose fibers.

Kinnunen et al. [102] used a mixture of anionic surfactants and CNF to prepare a foam coating, showing that at a lower coating weight, the paper surface did not achieve complete surface coverage, but there was a reduction in air permeability.

Zhang et al. [103] examined the barrier performance of various cellulose coatings by evaluating their permeability to air, oxygen and water vapor individually. The results showed that although the surface of cellulose-coated cardboard was more hydrophilic than that of uncoated cardboard, the cellulose coating of bleached bamboo pulp had the best oxygen and water vapor barrier properties after carboxyethyl pretreatment.

4.3. Antimicrobial Performance

In the food storage and processing links, if the hygiene conditions are not up to standard or there are defects in the operation process, it is easy to lead to microbial contamination. The moisture and nutrients in food will promote the growth of microorganisms. Therefore, there is an urgent need to take antimicrobial measures to extend the shelf life of food during transportation and storage. Packaging materials with antimicrobial properties came into being and are widely used in daily necessities, such as antimicrobial wipes and antimicrobial napkins [104]. Table 3 shows the application of polysaccharide polymer-coated paper-based materials to improve antimicrobial properties.

Todorova et al. [105] applied Bulgarian lavender essential oil to wrapping paper and discovered that the treated material exhibited notable antifungal properties while also enhancing its resistance to microbial contamination. However, due to the increased wettability, the mechanical properties of the paper decreased. Lu et al. [106] developed a silver ceramic antimicrobial food preservative paper, which not only maintained good mechanical properties, but also showed excellent antimicrobial effects. Experiments showed that using the paper to package spinach could effectively reduce the loss of vitamin C and chlorophyll.

Xia et al. [107] developed a biomass-based paper coating using three polysaccharide derivatives: carbamate starch (Sc), calcium Lignosulfonate (CL) and CNF. Through electrostatic interactions and hydrogen bonding, the coating enhances the compatibility between Sc, CL and CNF and silver nanoparticles, thereby improving the interface binding force between the paper and the bio-composite coating. The findings demonstrated that the coated paper exhibited substantial enhancements in mechanical strength, barrier performance and antimicrobial efficacy.

Irimia et al. [108] used cellulase as an activator, fixed vegetable oils with antimicrobial and antioxidant properties (such as clove oil and cold-pressed grapeseed oil) on the surface of bleached kraft paper and successfully designed and developed a new bioactive packaging material. This material has both antimicrobial and antioxidant properties.

Zhang et al. [109] first prepared a chitosan/carnauba wax emulsion by a one-step method and used it as an oil and water repellent coating for cellulose-based food wrapping paper. The coating not only endows the paper with excellent water, oil and heat resistance, but also makes it have good antimicrobial properties. In addition, the material is non-toxic, widely sourced and renewable, the preparation process is simple, no special emulsifier is required, and the desired antimicrobial effect can be achieved with a single coating.

Liu et al. [75] first used sodium periodate and concentrated sulfuric acid to oxidize starch into DAS, then modified DAS with GH to obtain GH-grafted DAS (DAS-GH), and finally coated paper with the DAS-GH solution and studied the antimicrobial properties of the coated paper. The findings demonstrated that the paper coated with DAS-GH exhibited effective antimicrobial properties against both *E. coli* and *Staphylococcus aureus*.

Hossen et al. [82] used nanocellulose, sodium alginate and biosynthetic silver nanoparticles to prepare bio-based nanocomposites as paper coating materials. Sodium alginate and nanocrystalline cellulose improved the water resistance of coated paper and made it have high tensile strength. The paper-based material has antimicrobial activity against *E. coli* due to the presence of silver ions.

Table 3. Research on the application of polysaccharide polymer-coated paper-based materials to improve antimicrobial properties.

Materials	Method	Features	Ref.
Carbamate starch, and cellulose nanofibers	The three polysaccharide derivatives enhance the compatibility between the polysaccharide derivatives and the nanoparticles through electrostatic interactions and hydrogen bonding interactions, thus improving the interfacial binding force between the paper and the bio-composite coating.	Hydrophobicity and excellent mechanical, air barrier and UV light blocking properties; inhibition of <i>E. coli</i> and <i>Staphylococcus aureus</i> .	[107]
Cellulase	Using cellulase-activated kraft paper and dipping kraft paper with clove essential oil and cold-pressed grape seed oil.	The hydrophobicity of the modified kraft paper was improved, and the water contact angle increased from 97° to above 110° and showed different antioxidant and antimicrobial properties.	[108]
Chitosan	Chitosan/Brazilian palm wax emulsion was prepared by using the one-step method as an oil-resistant and waterproof coating for cellulose-based food packaging paper.	When the concentration of chitosan is 3% and the amount of CW is 90% of the total solid content, the comprehensive performance of coated paper is the best in water resistance and oil resistance and has an excellent thermal stability and high antimicrobial rate of 99.1%.	[109]
Starch	The guanidine-modified starch was prepared by two-step reaction with guanidine hydrochloride as a modified agent.	Growth hormone was successfully grafted onto starch by Schiff base reaction, and the film was antimicrobial against both <i>E. coli</i> and <i>Staphylococcus aureus</i> .	[75]
Nanocellulose, sodium alginate	Nanocellulose was prepared by acid hydrolysis-fractionation.	Sodium alginate–nanocellulose composite coating improves the barrier and strength of paper, and the incorporation of AgNPs into the coating mixture also introduces the antimicrobial activity of paper.	[82]

4.4. Oil Resistance

Studies have shown that oils cannot be diffused by dissolving or moistening paper fibers as water can, because oils cannot interact directly with fibers. However, due to the large number of tiny pores inside the paper, the oil can permeate through these pores through capillary effects and gradually spread throughout the paper. Therefore, in order for the paper to have oil-resistant properties, it must be treated with a special treatment. Table 4 shows the application research of polysaccharide polymer-coated paper-based materials for improving oil resistance. Henrik Kjellgren [66] and his team conducted an

in-depth study of the oil-proof paper coated with chitosan at the laboratory and pilot scales and came to the following conclusions: (1) Regardless of the paper's air permeability, the oil-proof effect improved significantly with the increase of the amount of coating. (2) the tensile strength of the paper remained basically stable, unaffected by changes in the amount of coating, but when the amount of coating exceeded 5 g/m^2 , the breaking strain of the paper reached a peak. (3) When the coating amount of chitosan exceeds 5 g/m^2 , the oil resistance is greatly improved; however, due to the high viscosity of chitosan solution, this coating amount can only be achieved under laboratory conditions; under pilot conditions, the maximum coating amount is 0.2 g/m^2 .

Table 4. Research on the application of polysaccharide polymer-coated paper-based materials to improve oil resistance.

Materials	Method	Features	Ref.
Chitosan	Apply different breathable oil-proof papers with chitosan.	When the coating amount exceeds 5 g/m^2 , the paper fracture strain is the highest, and the paper obtains high oil prevention performance.	[66]
Chitosan	Food oil-resistant packaging materials were prepared with a chitosan/starch compound oil agent.	Adding cationic starch to the oil repellent can improve the oil repellent ability of the paper.	[110,111]
Chitosan and sodium alginate	Chitosan solution and sodium alginate solution as raw material, by ferulic acid crosslinking, coated on food packaging base paper.	When the quality ratio of chitosan and sodium alginate is 8:2 and the coating amount is 4 g/m^2 , the oil control paper can reach the highest oil control grade (grade 12).	[113]
Chitosan/cationic starch	The oil-proof properties of chitosan and chitosan/cationic starch-coated paper were compared.	Chitosan coating paper and chitosan/cationic starch coating paper both have good oil resistance, and their oil resistance increases with the increase of coating weight. The thermal stability and water resistance of the chitosan/cationic starch composites are better than the chitosan-coated paper.	[114]
Oxidized acetate starch	Organic chromium was crosslinked with oxidized acetate starch	The highest oil protection grade of the coated paper is grade 7.	[115]
Cationic starch, oxidized starch and cassava starch	Cationic starch, oxidized Starch and cassava starch with a certain amount of polyvinyl alcohol and oil prevention agent compound.	The adhesive agent made of cationic starch is better than the other two kinds of starch.	[116]

Peng, Long et al. [110,111] prepared the oil-proof agent and its coated oil-proof paper from chitosan and discussed the effects of the mass fraction, ratio, dosage, drying temperature, time and ambient temperature and humidity of the oil-proof agent on the oil-proof performance of the paper. Scanning electron microscopy observation showed that the surface of the paper changed significantly before and after coating. The results show that

the above factors can affect the oil-proof properties of the paper, and adding cationic starch to the oil-proof agent can further enhance the oil-proof effect.

Research by Jiang et al. [112] examined the effectiveness of carboxymethyl chitosan as an oil-repellent agent. Their findings indicated that applying carboxymethyl chitosan significantly enhanced the oil resistance of both transparent paper and kraft paper. More precisely, coating transparent paper with 4.8 g/m^2 of carboxymethyl chitosan achieved an oil resistance level of 12. For kraft paper, a coating of 6.7 g/m^2 of carboxymethyl chitosan resulted in an oil resistance level of 7.

Zhu et al. [113] utilized chitosan and sodium alginate as primary components, along with ferulic acid as a crosslinking agent, to develop a composite oil-resistant coating. This approach significantly enhanced and stabilized the oil-proof properties of the treated paper. Specifically, when the chitosan-to-SA mass ratio was set at 8:2 and the coating weight was 4 g/m^2 , the treated paper achieved its optimal oil-proof rating of level 12.

Long et al. [114] prepared a composite material consisting of chitosan and cationic starch and applied it as a coating on paper. Their findings indicated that the chitosan and cationic composites exhibited superior thermal stability and oil resistance compared to chitosan alone. Specifically, when the mass ratio of chitosan to chitosan was set at 1:2 and the coating weight was 1.5 g/m^2 , the paper achieved the necessary oil resistance for food packaging applications. Additionally, using organic chromium and biomass materials as raw materials in the preparation of composite coatings also enhanced the paper's oil resistance.

Zhang et al. [115] crosslinked the organic chromium and oxidized acetate starch and coated it on the surface of the paper, effectively improving the oil resistance of the coated paper. The oil-proof grade of the coated paper can reach up to 7, which meets the requirements of food wrapping paper. In addition, the biomass material can be mixed with the oil repellent to produce a composite oil repellent.

Wu et al. [116] mixed cationic starch, oxidized starch and cassava starch with PVA and oil repellent, respectively, to make an oil repellent sizing agent and then coated the paper. It was found that the oil-proof performance of the sizing agent made of cationic starch was better than that of the other two kinds of starch, and the oil-proof grade of the paper could reach 7.

5. Challenges and Future Perspectives

Due to the shortcomings of traditional plastic packaging, such as being non-degradable and non-renewable, resulting in increasingly prominent environmental pollution problems, it is urgent to find green renewable materials to replace plastic products. Because of its green environmental protection, functional development and other characteristics, the application field for paper-based materials continues to extend and expand. Polysaccharides are abundant in nature, biodegradable, non-toxic and harmless and have excellent film-forming properties, so they are excellent bio-based materials. Therefore, using natural plant fiber paper-based materials as the substrate and polysaccharide polymer as the coating material can improve the problems of high water absorption and poor mechanical and physical properties of paper-based materials, meeting the needs of paper-based packaging materials in various application fields.

This review reports the barrier mechanism and research progress of polysaccharide polymer coating on paper-based materials, but there are still some problems, such as high coating amount, high raw material cost, the demand of paper-based packaging contents on the properties of composite materials, the problem of interface bonding between polysaccharide polymer and paper-based materials and the improvement of the performance stability of paper-based composites in high humidity environments. How nanotechnology

or hybrid coatings can improve the performance of polysaccharides, recycling of coatings, etc., remains to be investigated. In addition, the chemical modification of polysaccharides still has the disadvantages of complex process, low efficiency and high cost, and whether excessive chemical modification affects its biodegradability and causes damage to the environment is also a problem we should pay attention to. In addition, the coating method is the main way to combine the coating with the paper base, and it essentially cannot solve the defects and problems of the paper base material; whether we can explore the combination of other composite materials to improve the interface binding degree or to improve the hydrophilic and oil-philic problems of the paper base material is the challenge we are currently facing.

Based on these problems, we need to further discuss the stability and durability of the coating and its compatibility with the substrate. Researchers can explore issues such as how to improve the binding and reuse of polysaccharides with paper-based materials, and how to further improve the performance of polysaccharide coatings. After solving these problems, researchers can continue to explore whether they can try to improve the barrier properties of paper-based materials while giving them other functional properties, such as both hydrophobic and electrical conductivity, according to temperature and humidity, pH changes and changes to the barrier efficiency of paper-based materials. With the deepening of research and technological progress, polysaccharide polymer-coated paper-based materials will make greater contributions to the development of human society in the field of packaging and other fields.

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