Research Status and Prospects of Bio-based Materials for Grease Barrier Coatings on Paper Food Packaging

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Abstract: Increased environmental and health concerns over the use of plastic packaging or fluorine-containing coatings, in combination with increased market demand for products with a longer shelf life, make bio-based materials one of the most important research candidates for alternative paper packaging materials for oil resistance. These bio-based materials have excellent oxygen and oil barriers, which are critical for food packaging. Moreover, they are biodegradable, naturally renewable, and safe. In this artical, two main groups of bio-based oil repellents for paper food packaging, including polysaccharide-based biopolymers and protein-based biopolymers, are enumerated, and the advantages and weaknesses of bio-based oil repellents are discussed, and effective solutions are proposed. Finally, research status and prospects on the development of bio-based oil-resistant coatings for the food packaging industry are presented.

Keywords: bio-based materials; grease barrier properties; paper food packaging

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1 Introduction

The food-packaging revolution began when humans learned to cook food. Food packaging occupies a substantial part of the modern packaging industry, accounting for approximately 60%–70% of the industry. Among all types of food packaging materials, plastic occupies a large share of the food packaging market because of its light weight, low cost, flexibility, and good barrier properties. However, because of their non-degradability and potential toxicity, plastics in landfills and marine debris have a considerable impact on the environment and human health [1–2], which can be countered using paper-based packaging. Moreover, the promulgation and implement of the "Plastic Limit Order" also promote the partial replacement of paper packaging for plastic packaging.

However, the rapid development of the fast-food industry and the increased popularity of take-out food in recent years have promoted the continuous expansion of the paper food packaging market. There is a great, unsatisfied demand for paper packaging including wrapping paper, paper boxes and bags in the fast-food and take-out industries. Paper packaging is more environmentally friendly, biodegradable, and recyclable [3]. Further, for food packaging, especially bakery products, fast-food, and pet-food storage, products with grease barrier properties are in high demand and critically important because they require excellent resistance against soaking through fat migration from the product and penetration [4–5].

The most common method of improving the oil resistance of paper packaging is to use oil repellents in the pulp as additives or coatings on the paper surface. Perfluoroalkyl and polyfluoroalkyl substrates (PFAS) are the most widely used and effective oil repellents because of their low surface energies. However, PFAS pose a huge threat to human health because of their toxicity and degradation difficulty resulting from perfluorooctanesulfonyl compounds (PFOA/PFOS), which are produced in the production and utilization process [6–7]. Since the Stockholm Convention on Persistent Organic Pollutants (POPS) was fully implemented in 2014, restrictions on fluorine-containing oil repellents have become increasingly stringent. Thus, there is an urgent need to develop environment-friendly and fluorine-free oil repellents.

Bio-based packaging materials are derived from natural renewable sources. Paper coatings, polysaccharides, proteins, and lipids, or combinations of these components, can form a dense film on the surface of paper or paperboard to prevent oil from permeating the paper and paperboard [8]; they may also serve as gas and solute barriers to minimize food quality deterioration and extend the shelf life of foods [9]. Furthermore, bio-based packaging materials offer favorable environmental advantages, such as recyclability and reutilization, compared to conventional petroleum-based synthetic polymers.

As a result, many studies successfully used bio-based materials as coatings on paper or paperboard to provide an effective oil barrier for food packaging applications. This review aims to provide a synopsis on the current status of bio-based oil repellents for paper food packaging and a view of prospective developments in sustainable next-generation paper coatings.

Bio-based oil repellents, which are different from fluorine-containing oil repellents, can form a barrier layer on the paper surface to resist the permeation of oil because the higher critical surface tension makes it difficult to prevent paper from being wetted by oil drops. Therefore, materials with good film formation may perform better as oil barriers. Currently, research on bio-based oil repellents mainly focuses on two categories: polysaccharide-based biopolymers (chitosan, starch, and nanocellulose) and protein-based biopolymers (zein, whey protein, and soy protein). The status and prospects of these two kinds of biopolymers used for grease barrier coatings on food paper packaging were reviewed in this artical.

2 Polysaccharide-based biopolymers

Polysaccharide-based biopolymers primarily include chitosan, starch, and nanocellulose. They have been used as adhesives in papermaking and paper coatings, and have received considerable attention for application in oil-barrier coatings for food packaging in recent years. As environmentally friendly materials, polysaccharide-based biopolymers can form a smooth and dense film on the surface to improve paper barrier performance owing to strong hydrogen bonding interactions. However, their natural hydrophilic characteristics limit their applications in the food packaging industry [10]. In addition to oil-resistant properties, water- or water vapor-resistant properties are also required for packaging materials, especially for oil-water mixtures in fast food packaging.

2.1 Chitosan

Chitosan is an edible and biodegradable material that exhibits excellent oxygen and oil barrier properties owing to its semi-crystalline nature, hydrogen bonds between molecular chains, and the positive charge on the amino group [11−14]. Chitosan also possesses good mechanical properties comparable to those of many synthetic polymers owing to its good film-forming ability. These properties make chitosan one of the most attractive polymers for coating paper or paperboard in food packaging applications.

Ham-Pichavant et al [15] compared the fat-barrier properties of uncoated paper and chitosan-coated paper using application tests. Evidently, compared with fluorinated resins, a similar efficiency could be obtained by coating with chitosan at a 2.2% coating level. However, the treatment costs remain high, although other natural molecules such as cellulose ethers and alginates have been blended into the coating formulations in an attempt to decrease the cost. Hamdani et al [16] indicated that the kit rating value (oil/grease) of chitosan-coated paper reached 12/12 when the coating weight was 6.1 g/m^2, which is sufficient to satisfy the requirements of food packaging applications. Jiang [17] studied the effects of carboxymethyl chitosan coating on paper barrier properties and showed that the grease barrier properties of chitosan-coated papers were significantly improved compared with those of uncoated paper, in which the highest grease rating number of 12 was achieved, and effective protection of hot oil was achieved when transparent paper was coated with 3.3 g/m^2 chitosan. Kjellgren et al [18] reported that grease resistance was excellent within the coating weight range of 2.4−5.2 g/m^2 for chitosan-coated paper.

Despite the good barrier properties that could be achieved with chitosan-coated paper or paperboard, the combination of paper and chitosan is not yet fully suitable for all food packaging applications; even setting aside the cost, water sensitivity is inherent to a majority of polysaccharides, which contain a large number of hydrophilic groups [19−20]. Several studies have been conducted in recent years to address this problem. Bordenave [21] found that compared to chitosan-coated papers, papers coated with either chitosan-palmitic acid emulsions or with a blend of chitosan and O,O’-dipalmitoylchitosan (DPCT) both improved the liquid-water resistance of the materials, and maintained good grease barrier properties (degree of resistance 6−8/12) at same time. Wang et al [22] reported that by coating paper with a mixture of chitosan and montmorillonite (MMT) instead of a chitosan solution alone, the coated paper exhibited lower air and water vapor permeability and enhanced oil resistance at a lower coating weight.

Chitosan has the potential to replace plastics in food packaging, although many problems need to be resolved before it can be commercially used; these concerns include the difficult process of chitosan coating during coating process, high viscosity and high cost. Optimizing both the cost and performance to realize the multiple functions (waterproofing, antibacterial, and water vapor barrier) of chitosan by chemical modification or compounding with other materials may be a viable alternative.

2.2 Starch

Starch and its derivatives are naturally available, inexpensive, and biodegradable polymers commonly used in paper coating. In its native form, it can be utilized as a sizing agent, whereas after modification, it can be used as a coating agent to enhance paper properties owing to its excellent film-forming ability. Starch and its derivatives were early biomaterials used in oil-resistant food packaging. Recently, some modified products have entered the market, such as Filmkote 370, a modified oil-proof starch produced in the USA; however, these products are not as effective as expected.

Many attempts have been made to improve the gas, oil, and water vapor barrier properties of starch, including plasticization, blending with other materials, modification (chemical, physical, or enzymatic method), and using different combinations of such interventions. Song et al [23] alkalized potato starch at a low concentration and used it to increase the oil resistance properties of paper via a one-sided coating on supercalendered papers. The results showed that excellent oil barrier properties could be achieved at a coating weight of 3 g/m^2 at room temperature and 12 g/m^2 at 60 ℃ for alkali-treated starch-coated paper. Zhang [24] reported that after coating using starch crosslinked with organic chromium, the oil resistance of the coated paper increased with an increase in the dosages of organic chromium and coating weight. In addition, starch was mixed with chitosan, PVA, and gum as coatings for paper, and the results showed that all these combinations could improve the grease and oxygen barrier properties of paper, in which the starch/chitosan composite-coated paper was the best. When the chitosan dosage was 20%, the oil resistance grade increased by 4 levels and the oxygen transmission rate was reduced by 89.7%. Menzel et al [25] reported that the crosslinking of starch with citric acid enhanced the barrier and mechanical properties of coated papers, depending on the pH value and coating process used. Buutkinaree et al [26] investigated water resistance and grease barrier properties by coating a combination of stearic acid and hydrophobic starch on a paperboard surface, and found that the water and grease resistance properties of the coated paperboard were significantly improved.

Starch-based oil repellents are a hot topic in the field of oil repellent research for food packaging. It not only has excellent performance, safety, and environmental protection property, but can also be combined with a variety of other materials to optimize its waterproofing and oil repellent effects, mechanical properties, antibacterial properties, thermal stability, and food preservation properties [27]. Considering that, these combinations may weaken the cost advantage, while the ultimate goal is to find a middle ground between performance and price.

2.3 Nanocellulose

The nanocellulose mentioned in this paper includes cellulose nanofibers (CNF), microfibrillated cellulose (MFC), and cellulose nanocrystals (CNC). Owing to its good barrier properties, the application of nanocellulose in food packaging has emerged as a promising alternative to plastic, attracting an increasing number of researchers in recent years. Their nanometer scale, high surface energy, and ability to form a nanoporous network make MFC an ideal material in paper coatings [28]. Several studies have employed MFC as composite coatings on paper substrates to obtain better barrier properties for gas, water, and oil.

To evaluate the potential application in packaging materials, Aulin et al [29] investigated the oil barrier properties of MFC films by surface coating them on base papers. It was evident that the paper with low air permeability showed superior oil resistance (1800 s for castor oil), which was possibly due to the dense surface porous structure formed by the nanofibers, as studied by SEM. Chen et al [30] coated a nanocellulose suspension prepared by TEMPO oxidation onto A4 paper to analyze its oil-resistant properties. The experimental results showed that the oil-proofing grade of the coated paper reached level 6 when the CNF coating weight was 2.8 g/m^2. As the nanocellulose coating weight increased to 4.9 g/m^2, the air permeability decreased continuously, while the grease-proof level of the paper increased to 12. Moreover, it was found that the paper acted to be heat oil and solvent-proof. Gicquel et al [31] only used CNC to be oil-proof coating to investigate the surface and barrier properties of coated paper. Concerning Tappi-454, paper coated with CNC yielded the first grease pinhole at 5 s, whereas the red oil immediately penetrated the uncoated paper. This demonstrates that the CNC-coated paper is more resistant to oil because the CNC coating increases the tortuosity of the network of paper fibers.

Nanocellulose-based coatings exhibit excellent gas and oil barriers. However, the difficulty in the application progress is due to the very high viscosity at low solids and susceptibility to moisture. These two limitations limit their commercialized prospects in application such as barrier coatings for food packaging. The solution, in this respect, is to chemically modify nanocellulose or develop nanocellulose-based barrier composite coatings with other materials such as nanofillers and lipids. Tyagi et al [32] developed a composite coating containing a mixture of CNC, a high-aspect-ratio nanofiller montmorillonite clay, an amphiphilic binder soy protein, and a surface-active agent, alkyl ketene dimer. CNC composite coatings exhibited a significant reduction in water absorption (up to 71% compared to surfaces with no coating, and up to 27% for surfaces with a CNC single coating), water vapor transmission rate (up to 27% compared to surfaces with no coating, and up to 6% for surfaces with a CNC single coating), and resistance to air permeation (up to 88% compared to surfaces with no coating, and up to 44% for surfaces with a CNC single coating). Furthermore, rheological analysis revealed a decrease in viscosity with the addition of sodium montmorillonite (MMT) and other additives, indicating the possibility of fabricating coatings with higher solid contents.

Another approach to protect nanocellulose-based coatings from moisture is to have a multilayered structure consisting of alternating layers of nanocellulose and hydrophobic polymers such as guar gum, polyvinyl alcohol, polyhydroxyalkanoates (PHAs), shellac, and poly lactic acid (PLA) [33–35]. Rajesh Koppolu et al [33] combined nanocellulose and PLA to form thin multilayer coatings via a continuous process. Their research showed a significant reduction in the water vapor transmission rate of multilayer coatings compared to that of single coating with nanocellulose or PLA, even at a high relative humidity of 90%. In addition, the oxygen transmission rate of the multilayer coatings was 98% lower than that of the PLA-coated paperboard, and the grease barrier for the nanocellulose-PLA composite coatings increased 5-fold compared to that of single coating with nanocellulose, and 2-fold compared to that of single coating with PLA.

Although there are still significant gaps in the performance and cost between nanocellulose and potential replacements for petroleum-based materials, nanocellulose-based materials with competitive performance for food packaging seems attainable.

3 Protein-based biopolymers

The excellent barrier properties and satisfactory mechanical performance of protein-based biopolymers make them promising materials for food packaging application. Compared with polysaccharide-based biopolymers, the viscosity of the protein solution is lower at the same concentration, which is beneficial for the coating process [36]. In recent years, protein-based biopolymer films have been studied as coating materials for food containing oil to extend the shelf life and improve the safety of processed food [37–38]. Currently, most studies focusing on the oil resistance of protein-based biopolymers have mainly concentrated on zein, whey protein, soy protein, and casein.

3.1 Zein

In particular, zein is hydrophobic because of its high content of nonpolar amino acids [39], which makes the water resistance of zein-based films or coatings superior to that of other protein-based films or coatings [40]. Because of their good film-forming characteristics, zein-based coatings can be used as barrier layers for food packaging, providing resistance properties for oxygen, grease, and moisture.

Research on the application of zein-based polymers in food packaging began early. Trezza et al [41] measured the grease resistance of zein-coated paper with different coating weights to determine its grease-proofing ability and potential application in fast-food packaging. Zein-coated paper is as effective as polyethylene laminates for oil resistance in fast-food packaging. Furthermore, zein-coated paper would be adequate as grease barriers for 12 hours in fast-food packaging when the coating weight reaches to 4.4−6.6 g/m^2, and their oil-resistance becomes better with improved coating uniformity and a higher coating weight. Parris et al [42] examined the barrier properties of paper prepared from recycled kraft paper and linerboard coated with fibers. According to the results, the zein-based coating reduced the water vapor transmission rate (WVTR) and grease permeation by 16% and 100%, respectively, on paper prepared from reslushed kraft paper, and 8% and 97%, respectively, on paper prepared from kraft linerboard.

These results demonstrate the potential of zein in paper-based food packaging. Although zein-based coatings have relatively good water and water vapor resistances, they are insufficient for food packaging. Therefore, lipids or crosslinking agents have been used to improve the water and water vapor barrier characteristics of zein-based coatings [43]. Parris et al [44] measured the water and grease barrier properties of kraft paper coated with a combination of zein and paraffin wax. The results showed that the zein layer of the bilayer coating contributed to grease proofing, and the wax layer contributed to water resistance. Weller et al [45] determined the properties of single-layer and bilayer zein/lipid films prepared by coating dried zein films with medium-chain triglyceride (MCT) oil, laboratory-extracted sorghum wax (SW)/MCT oil, or commercially refined carnauba wax (CW)/MCT oil. The results indicated that the addition of a lipid layer significantly decreased water vapor permeability (WVP) of zein films from 9.07 $g \cdot mm/(m^2 \cdot h \cdot kPa)$ to as low as 0.115 $g \cdot mm/(m^2 \cdot h \cdot kPa)$.

The potential barrier ability of zein has spurred considerable research on its application in food packaging. However, to commercialize these applications, intensive researches on improving their performances of mechanical properties and flexibility are necessary. Current methods for increasing the tensile strength of zein-based coatings often result in reduced flexibility [46]. Therefore, maintaining balance within the desired performance is important.

3.2 Whey protein

Whey protein has excellent film-forming properties along with barrier properties against oxygen, aroma, and oils, which can be further enhanced by crosslinking the polymeric chains, making the whey protein-based coating material water-insoluble and more suitable for paper-based food packaging [11]. Whey proteins can be divided into two types based on quality and purity: whey protein concentrate (WPC) and whey protein isolate (WPI).

Han et al [47] tested the contact angle changes of oil drops on WPI-coated paper to examine the potential practical uses of WPI-coated paper for food packaging. The experimental results showed that the reduction rate of the corn-oil contact angle of the WPI-coated paper decreased with increased coating weight. Especially, when the coating weight reached to 18 g/m^2, the reduction rate was 0.0482 °/min, which is statistically nearly 0. Therefore, the WPI coating could be used as an oil barrier on paper packaging materials for the hamburgers or fried foods at a fast-food service restaurant. Additionally, the excellent oil resistance of WPI-coated paper is comparable to that of commercial flexible plastic laminated paper (low-density polyethylene at 9.5 g/m^2). The study conducted by Chan et al [48] also demonstrated the excellent oil barrier properties of paper coated with whey protein.

The incorporation of plasticizing agents is necessary to overcome the intrinsic brittleness of whey protein coatings [49–50]. Chan et al [51] found that paperboard coated with WPI and glycerol as plasticizers exhibited a good oil barrier, along with the migration of the glycerol plasticizer into the paperboard during storage. Lin et al [52] discussed the effect of sucrose (Suc) as an alternative to glycerol plasticizers. They found that Suc imparted excellent grease resistance as a plasticizer, which was similar to that of glycerol, and Suc performed better than glycerol in preventing the cracking of WPC coatings during storage. In addition, they reported that WPC with approximately 80% protein coating on paperboard provided a grease barrier which was comparable to that of WPI coating at a substantially lower cost. To explore the potential of this alternative in practical application, Yoo et al [53] used WPC combined with Suc plasticizer for paperboard coating in food packaging to determine the differences in grease barrier properties. The results showed that the paperboards coated by 10% WPC showed excellent oil barriers comparable to commercial fluorinated hydrocarbon, while the paperboards with 10% WPC: 20% Suc solution coating had oil barrier comparable to polyethylene-coated paperboards.

As mentioned above, whey protein coatings containing plasticizers are flexible, safe, biodegradable, and have excellent barrier characteristics for oxygen, oil, and aroma [54]. Similar to other hydrophilic materials, whey proteins have limitations in terms of moisture content [43]. Therefore, the application of lipid materials (fats and oils) may be indispensable for this system to improve water and water vapor barrier properties. Overall, whey proteins are promising ecofriendly alternatives to synthetic polymers.

3.3 Soy protein

Soy protein isolate (SPI) also has a remarkable film-forming capacity, resulting in better oxygen barrier performance than zein and wheat gluten [54–55]. Different from zein, SPI is an inexpensive biopolymer that competes with polyethylene in terms of cost.

Park et al [56] measured the grease barrier properties of SPI-coated paper and compared with those of two commercial polyethylene-laminated papers. The results showed that the SPI-coated paper was impermeable to grease penetration for the first 2 h. When the paper was coated with SPI over 2.0 g/m^2, there was little grease migration, which was slightly superior to that of the polyethylene-laminated papers.

As reported, soy protein-coated paper was found to impart gas and oil barriers as well as adequate mechanical properties [56], while providing poor water barrier properties owing to the hydrophilic nature of soy protein. Researchers have attempted to address this problem. Han et al [57] incorporated SiO_2 nanoparticles with SPI films. The results indicated that compared with pure SPI films, using nanoparticles with the minimum size not only induced the greatest decrease in the WVTR and oxygen transmission rate (OTR) values by 11.79% and 9.66%, respectively, but also improved the contact angle and tensile strength by 21.12% and 17.65%, respectively. Wang et al [58] prepared SPI films with oleic and stearic acids, and characterized their water vapor barrier properties and contact angles on a hydrophobic surface. The data showed that the addition of oleic acid and stearic acid was effective in improving the water vapor barrier abilities, which was embodied in the decrease in the WVP by 70% and the increase in the contact angle by approximately 65° (the highest was 135°) compared with those of the control group.

Given the above, we may reasonably arrive at the conclusion that adding nanoparticles, fatty acids, and lipids to overcome the defects that have limited the application of SPI films or coatings in food packaging seems to be a promising approach.

3.4 Casein

Casein, especially sodium caseinate (NaCAS), is usually used as caseinate because of its poor solubility in water. NaCAS appears to provide better gas barrier properties than nonionic polysaccharides [59], which may be related to its more polar nature and more linear (non-ring) structure, leading to a higher cohesive energy density and lower free volume [60]. In addition, NaCAS also possesses good mechanical properties, as demonstrated by Khwaldia et al [61]. These properties make caseinate an attractive paper coating alternative to plastics for food packaging.

Aloui et al [62] used NaCAs reinforced with halloysite nanotubes (HNTs) as a coating material on paper surfaces to investigate the effects of the coating weight and HNTs content on the mechanical, optical, and barrier properties of the coated papers. Compared with uncoated paper, coatings based on NaCAs, either alone or reinforced with HNTs, were able to reduce or prevent oil permeation through the paper. Papers coated with NaCAS at coating weight of 9.9 g/m^2 exhibited excellent grease resistance represented by the appearance of the first oil stains on the undersurface after 372 min; papers within the coating weights range of 9.9 g/m^2 and 17 g/m^2 would be regarded as fully greaseproof as no oil stains were observed on the undersurface after 24 h, while oil stains of the uncoated paper appeared after 7 s. Further, the addition of HNTs can improve grease resistance properties at a lower coating weight.

Although caseinates have the aforementioned advantages, some drawbacks still need to be addressed to adapt them to practical application, such as the lack of water resistance and flexibility. Currently, the use of functional additives, such as wax, fatty acids, plasticizers, and crosslinkers, is an effective solution [63–66]. For instance, films prepared from aqueous emulsions of calcium caseinate and beeswax exhibited a 90% reduction in WVP compared with those of films prepared from calcium caseinate alone [67]. Since additives influence other properties, further comprehensive studies are required to determine optimal performance.

4 Conclusions and future perspectives

As described above, problems that are similar with most biopolymers, such as hydrophilicity, crystallization behavior, lack of flexibility, and high cost, prevent their full commercial exploitations. We also discuss some corresponding solutions proposed in many studies as representative examples. Although bio-based materials, compared with fluorinated oil repellents, still have a great shortage in effect and cost, extensive researches are needed in the paper food packaging industry towards developing methods for new coating formations, conveying costs and improving the properties of paper coatings [68]. Bio-based materials are considered to be promising oil repellents with grease barrier for the future development of mainstream direction in paper food packaging.

In the future, numerous combinations can be used cooperatively on an industrial scale, depending on consumers demands, products characteristics, and operating conditions. In our view, the research directions of bio-based grease barrier will focus on the following three aspects in the years ahead.

4.1 Modification or compounding with other materials.

The barrier properties of bio-based materials may be improved by chemical and physical crosslinking or by the incorporation of additives and other bio-based materials. Chemical modifications have been specifically used to provide good water or water vapor barrier properties, including grafting, acetylation, and alkylation. The moisture sensitivity of bio-based coatings can be decreased by crosslinking, leading to improved barrier properties against water vapor and oxygen transmission at high humidity [69]. The combination of additives and other bio-based materials can improve certain properties. For example, the addition of plasticizers can make coatings flexible, whereas the addition of nanocellulose may enhance the oxygen barrier performance. However, all these improvement approaches may have a negative influence on the biodegradability of the coatings, which means that we should consider the balance of both sides.

4.2 Innovation of processing method

Paper coating technology has matured abroad and at home, including bar coating, knife coating, dip coating, and curtain coating. However, innovative processing methods need to be applied to contribute to the bio-based coatings properties as auxiliary means. Drawing lessons from successful practices in other related industrial areas is an accessible approach, such as in the fields of extrusion, enrolling, fluidization, spraying, and UV polymerization. Currently, extrusion and compression molding, which are well-known commercial methods applied to develop films and plastic granules, are considered the most likely to be successful in the process of preparing bio-based barrier coatings. However, many problems such as plasticization and process monitoring still need to be solved.

4.3 Multilayer barrier coatings

Given that most bio-based materials do not present a sufficient water vapor barrier for food packaging, the combination of individual layer with a multilayer coating structure is often necessary to improve their competitive edge against synthetic films [70–71]. The application of hydrophobic compounds as the top coating layer can provide the coated paper with water vapor barrier properties and minimize the unfavorable effects of moisture on the oil-resistant layers. In fact, some hydrophobic compounds such as polylactic acid and shellac can also contribute to oil barrier properties as top coating layer. Owing to the compatibility of the multilayer coating structure, an additional functional layer can provide certain properties, such as antimicrobial ability or printability, which may expand the application of related products.

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