Proceedings of the 4th International Conference on Functional Materials and Civil Engineering
DOI: 10.54254/2755-2721/2026.36463

Research Progress on Matrix Materials, Preparation Technologies and Applications of Degradable Packaging Films

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Abstract. With the environmental problems caused by traditional plastic packaging becoming increasingly serious, degradable packaging films have attracted growing attention in the fields of food packaging and preservation. Compared with conventional petroleum-based plastics, bio-based films prepared from natural polymers show advantages such as renewability, biodegradability, environmental friendliness and potential food-preservation functions. Focusing on cellulose, starch, chitosan and carrageenan as well as their derivatives and composite films, this paper sorts out their basic material characteristics, film-forming properties and functional advantages. It further summarizes several common preparation methods such as solution casting, extrusion blown film, coating, electrospinning, multilayer compounding and hot pressing. In addition, the applications of degradable composite films in meat preservation, fruit preservation and paper-based food packaging are discussed. By integrating material selection, processing methods and practical application, this paper aims to provide useful references for the development of bio-based composite films in the fields of food preservation and green packaging.

Keywords: natural polymers, degradable packaging films, matrix materials, preparation technologies, preservation applications

1. Introduction

Driven by the rapid development of food consumption modes, takeaway and fresh retail industries, the consumption of packaging materials keeps rising, and environmental pollution derived from traditional plastic packaging has become increasingly severe. Against this backdrop, degradable packaging films have gradually turned into a key research field of green packaging due to their environmental friendliness, food contact suitability and fresh-keeping functions [1, 2]. Especially in meat, fruit and paper-based food packaging, composite films can not only serve as alternative materials for traditional plastics, but also extend the shelf life of food by blocking moisture, oxygen and microbial invasion [3], thus possessing high application value and practical significance.

At present, certain progress has been achieved in researches on degradable packaging films. Marinho [4] emphasized the renewability and structural designability of cellulose materials;

DOI: 10.54254/2755-2721/2026.36463

Paunonen [5] discussed the advantages of cellulose derivative films in mechanical strength and barrier properties; Li et al. [6] applied oxidized starch/Polybutylene Adipate Terephthalate (PBAT) composite films to chilled pork preservation, which reflected the application potential of degradable composite films. Nevertheless, existing literatures mostly focus on a single material, a single preparation method or a single category of applications, lacking comprehensive collation of the correlations among material systems, preparation technologies and practical application scenarios.

On this basis, this paper systematically reviews natural degradable composite packaging films. Firstly, it analyzes the functions of natural polymer materials including cellulose, starch, chitosan and carrageenan as well as their derivatives in degradable packaging films. Secondly, it summarizes the characteristics of mainstream composite film preparation methods such as solution casting, extrusion blow molding and coating. Finally, combined with the applications in meat preservation, fruit preservation and paper-based packaging, it discusses the practical value of composite films in food preservation. Through such systematic collation, this paper intends to offer references for material selection, process optimization and food packaging application of degradable composite films.

2. Matrix materials of degradable packaging films

2.1. Cellulose and its derivative-based degradable packaging films

As one of the most abundant natural polymers on the earth, cellulose boasts extensive sources, renewability and biodegradability, which has long made it a critical research object in polymer materials and sustainable chemistry. However, cellulose also has limitations including strong intermolecular hydrogen bonds, high crystallinity and poor solubility in common solvents, which motivates extensive researches and applications on cellulose modification and its derivatives.

Relevant studies point out that the value of cellulose lies not only in its natural abundance, but also in its structural designability and application prospects in green materials [4]. Randhawa et al. [7] regarded nanocellulose as an important extension of cellulose research, including cellulose nanocrystals, cellulose nanofibers and bacterial cellulose, which have attracted wide attention thanks to their excellent mechanical properties and functionalization potential. In addition, Paunonen's [5] research on cellophane and cellulose derivative films indicated that typical derivatives such as cellulose acetate, methyl cellulose and carboxymethyl cellulose possess obvious advantages in enhancing film strength and barrier performance. Overall, existing literatures generally agree that cellulose and its derivatives form a bio-based material system integrating structural designability, functional diversity and environmental friendliness, and their future development relies on the balance among structural designability, material functionality and sustainability.

2.2. Starch and modified starch-based degradable packaging films

Starch is a natural polysaccharide with abundant sources, featuring renewability, biodegradability, low cost and favorable film-forming performance. Therefore, it is recognized as an important raw material for preparing degradable packaging films. However, native starch films usually suffer from strong hydrophilicity, insufficient mechanical properties and poor stability. Accordingly, chemical or physical modification is commonly adopted to improve their properties in practical applications [8].

In the field of degradable packaging films, modified starch mainly functions to improve the comprehensive properties of films to meet practical packaging demands. Mehboob et al. [9] performed crosslinking, acetylation and dual modification on sorghum starch, and found that the

DOI: 10.54254/2755-2721/2026.36463

transparency, flexibility and mechanical properties of the resulting films were all improved. Among them, acetylated films exhibited better flexibility, while dual-modified films outperformed others in mechanical strength and water resistance, demonstrating that different modification methods can adjust the properties of packaging films in a targeted manner. Apart from single-component starch films, modified starch is also frequently blended with other degradable polymers to boost material practicability. Wongphan et al. [10] revealed that octenyl succinic anhydride modified cassava starch could enhance the compatibility with PBAT in PBAT/Thermoplastic Starch (TPS) blend films, thereby optimizing the morphology, thermomechanical properties and barrier performance of films, and meanwhile affecting material degradation. This proves that modified starch acts not only as a film-forming raw material, but also plays a key role in regulating the internal structure of blend films. Furthermore, Zhao et al. [11] discovered that oil-phase modification of crosslinked and acetylated cassava starch further improved the material's shear resistance and emulsion stability, which facilitated stable dispersion during film processing and subsequent functional design. Kim et al. [12] put forward pressure-moisture treatment, a physical modification method, and verified that it can improve the physicochemical properties of starch without introducing new chemical groups, providing a more environmentally friendly strategy for the development of green packaging films. In general, the value of modified starch consists in making up for the defects of native starch, as well as promoting its practical application in food packaging and sustainable packaging materials by improving mechanical properties, water resistance, barrier performance and compatibility [10-12].

2.3. Chitosan and its derivative-based degradable packaging films

Chitosan is a natural cationic polysaccharide obtained from deacetylation of chitin, possessing biodegradability, film-forming capacity, biocompatibility and natural antibacterial activity. Compared with many natural polymers, chitosan has unique advantages in antibacterial and fresh-keeping performance for food packaging. Nevertheless, pure chitosan films are plagued by insufficient water resistance, limited flexibility and unsatisfactory processing stability. Hence, modification approaches including quaternization, carboxymethylation, crosslinking or blending with other polymers are commonly adopted in practical applications.

In the field of degradable packaging films, the core value of chitosan and its derivatives lies in improving antibacterial activity, barrier performance and interfacial stability to make films more suitable for actual fresh-keeping requirements. Lai et al. [13] prepared composite films by compounding quaternary phosphonium chitosan with polyvinyl alcohol and crosslinking with citric acid. The results showed that this method not only elevated the mechanical properties and antibacterial activity of films, but also exhibited promising application potential in strawberry preservation. Wang et al. [14] further developed composite films composed of chitosan/esterified chitin nanofibers/eggplant peel anthocyanins. They found that the introduction of nanofibers densified the film structure and significantly improved its mechanical properties, water vapor barrier and oxygen barrier performance, while anthocyanins endowed the films with antioxidant, antibacterial and pH-responsive capabilities, enabling their application in pork freshness monitoring. This reflects the trend of chitosan-based films evolving from "protective packaging" to "intelligent active packaging". Besides, chitosan is often blended with other hydrophilic polysaccharides. Karim et al. [15] proved that chitosan/carboxymethyl cellulose composite films can serve as eco-friendly packaging materials for chilled shrimp preservation, which indicates that chitosan-based composite films have gradually stepped from simple performance optimization to specific food system applications. Overall, the significance of chitosan and its derivatives rests not only on their biodegradability and film-forming characteristics, but also on their capacity to lift the practical value of packaging films through antibacterial, barrier, antioxidant and intelligent responsive functions, so as to promote their further application in food preservation and sustainable packaging.

2.4. Carrageenan-based degradable packaging films

Carrageenan is a class of sulfated polysaccharides extracted from red algae, which has attracted extensive attention in bio-based material research due to its biodegradability, biocompatibility, water solubility and favorable film-forming ability. However, existing studies show that the physicochemical properties of carrageenan are affected by raw material sources and extraction processes. Chan et al. [16] compared carrageenan extracted from Kappaphycus alvarezii with various commercial carrageenans, and found remarkable differences in color, particle size, texture, functional characteristics and chemical composition. Martín et al. [17] further pointed out that ultrasound-assisted extraction can not only raise carrageenan yield, but also alter its color, water retention capacity and functional group characteristics. This demonstrates that the final performance of carrageenan materials depends on both their categories and raw materials as well as preparation methods.

In recent years, researches on carrageenan have concentrated on degradable packaging films. Farhan and Hani [18] took semi-refined carrageenan as the matrix and incorporated fenugreek seed sprout extract to prepare active edible films. The results indicated that the extract increased the total phenolic content, antioxidant activity and color-related properties of films, and exerted positive effects on chicken breast preservation under refrigeration. Wang et al. [19] further fabricated corn starch/carrageenan composite films loaded with grape seed ethanol extract, and verified that the films possessed both antioxidant and antibacterial activities. In addition, Zarina and Ahmad [20] introduced kenaf fiber-derived cellulose nanocrystals into carrageenan films, and confirmed that composite films with appropriate loading exhibited better dispersion, superior mechanical properties and higher thermal stability. Wu et al. [21] synergistically incorporated peppermint essential oil and thymol into carrageenan-based films to produce degradable food packaging films with enhanced antioxidant and antibacterial capacities. In summary, existing researches prove that carrageenan has developed from traditional thickeners and gelling agents into designable bio-based packaging materials, and displays great application potential in mechanical properties, barrier performance as well as antioxidant and antibacterial functions.

3. Preparation methods of degradable packaging films

3.1. Solution casting method

Solution casting is one of the most mainstream and common laboratory film-forming methods for natural polymer degradable composite films, especially suitable for preliminary formula screening and performance research of hydrophilic film-forming materials such as starch, chitosan and carrageenan. It is widely adopted owing to simple operation, mild conditions and convenient formula regulation. Its basic procedures are as follows: first, dissolve or disperse film-forming polymers in appropriate solvents respectively, then add functional components including plasticizers, nanofillers or plant extracts, form homogeneous film liquid via heating, stirring or ultrasonic treatment, defoam the liquid, cast the liquid on flat substrates, and dry to form films under controlled temperature [22]. Choo et al. [23] adopted this method to prepare chitosan/acetylated starch-essential oil composite films. The results showed that the addition of essential oils improved the light, water vapor and oxygen barrier performance of films, yet reduced tensile strength and

DOI: 10.54254/2755-2721/2026.36463

elongation at break at high dosage. Wang et al. [22] compounded corn starch, carrageenan and onion skin ethanol extract, and the research demonstrated that the extract could inhibit starch recrystallization and enhance the antioxidant and antibacterial activity of films, making them potential active food packaging materials. In general, the advantages of solution casting lie in intuitive process, low equipment requirements, and suitability for formula design and optimization of composite films at laboratory stage.

3.2. Extrusion blow molding method

Extrusion blow molding is a preparation method for degradable composite films that is closer to industrial production. This method mainly melts and blends thermoplastic starch, PBAT and other polymers under high temperature and shear force, then extrudes the mixture through a die head and stretches it by blowing to form continuous films. Li et al. [6] adopted extrusion blow molding to prepare oxidized starch/PBAT degradable active films, which illustrated that modified starch can be compounded with PBAT during melt processing to form packaging films with certain mechanical properties and antibacterial functions. Katekhong et al. [24] blended nitrite-containing thermoplastic starch with PBAT and prepared active packaging films via extrusion blow molding. The research proved that this method can introduce functional additives during film formation, thus improving the oxygen barrier performance and meat preservation effect of films. Overall, extrusion blow molding features high continuity and production efficiency, and facilitates the transformation of degradable composite films from laboratory research to practical packaging applications.

3.3. Coating method

Coating is a surface modification method commonly used to prepare degradable composite packaging films. Its core idea is to firstly prepare a base film such as PLA (polylactic acid), PBAT, starch film or paper-based material, then coat chitosan, carrageenan or active extract solution on the surface, and form composite coatings after drying, so as to improve the antibacterial activity, barrier performance or fresh-keeping capacity of films. Stoleru et al. [25] took PLA films as substrates and covered the surface with chitosan coating liquid containing clove oil or argan oil to fabricate composite packaging films with active functions. This study revealed that coating can introduce natural active ingredients without altering the main structure of base films, and elevate their application performance in meat and cheese packaging. Kumbunleu et al. [26] adopted chitosan solution for single-side coating of PLA films and applied the coated films to vegetable and fruit preservation. In general, coating is relatively simple to operate, and can improve antibacterial activity, antioxidant capacity, barrier performance or fresh-keeping effect without completely changing the structure of base films, serving as an important method to enhance the practical application performance of packaging films.

3.4. Other methods

Electrospinning, multi-layer lamination and hot pressing are also frequently utilized for the preparation of degradable composite films. Electrospinning mainly stretches polymer solutions into nanofibers under high-voltage electric field, and deposits the nanofibers on base film surfaces or directly forms fiber films. This method is suitable for preparing functional layers with high specific surface area, which facilitates loading antibacterial agents, antioxidants or natural extracts. Gulzar et al. [27] electrospun gelatin/chitosan nanofibers loaded with tannic acid and chitooligosaccharides

DOI: 10.54254/2755-2721/2026.36463

onto PLA film surfaces, which indicated that electrospinning can be applied to construct surface functional layers of active packaging films.

Multi-layer lamination prepares films by compositing different materials layer by layer, enabling each layer to undertake mechanical support, barrier, antibacterial or fresh-keeping functions separately. Compared with single-layer films, multi-layer films can more easily realize functional complementarity among different materials, yet their preparation process is relatively complex. Fu et al. [28] prepared chitosan/pectin bilayer films via layer-by-layer casting, which reflected the advantages of multi-layer structures in regulating interfacial interactions and improving film properties.

Hot pressing generally compresses pre-mixed materials into films under certain temperature and pressure, which is suitable for thermoplastic degradable polymers and their composite systems. Yan et al. [29] crushed PLA/chitosan composites and prepared dense composite films via hot pressing, proving that hot pressing can fabricate compact composite films under solvent-free conditions. Overall, these three methods are mostly adopted for functionalization or structural optimization, acting as supplements to mainstream film-forming methods.

4. Applications of degradable packaging films in food preservation

The applications of composite films in food preservation are mainly reflected in inhibiting microbial growth, reducing water loss, slowing down oxidation reactions and maintaining food quality. For meat products, high protein and fat contents easily lead to microbial reproduction, lipid oxidation and color deterioration during storage. Therefore, composite films are widely used for packaging fresh meat, poultry and aquatic products. The oxidized starch/PBAT degradable active films studied by Li et al. [6] can be applied to chilled pork preservation, which demonstrates the application potential of starch-based composite films in meat packaging.

In terms of fruit preservation, composite films are mainly applicable to soft fruits such as strawberries, cherries and citrus, as well as fresh-cut fruits. Such fruits have thin epidermis and soft tissues, which are vulnerable to mechanical damage, microbial invasion and water loss after picking. Natural polymers including chitosan, starch and pectin can form a protective film on fruit surfaces via spraying, dipping or coating, so as to reduce water evaporation, lower external microbial contamination, and delay softening and spoilage to a certain extent [30]. Khan et al. [31] coated strawberries with chitosan and its derivative coatings, and found that the coatings reduced water loss, rot and microbial proliferation during strawberry storage, which proved the promising application prospect of composite film materials in fresh fruit preservation.

In addition, composite films can be used as sustainable packaging materials for paper-based tableware, inner coatings of takeaway boxes and functional layers of fresh food packaging films in supermarkets. Compared with traditional petroleum-based plastics, composite films made of natural polymers such as starch, chitosan and carrageenan feature biodegradability and renewable raw material sources. When used as food-contact packaging films or inner coatings of paper-based packaging, they can improve the water resistance, oil resistance and fresh-keeping performance of materials to a certain degree. Kopacic et al. [32] coated cardboard with alginate and chitosan, and verified that natural polymer coatings can enhance the barrier performance of paper-based packaging materials.

DOI: 10.54254/2755-2721/2026.36463

5. Conclusion

In summary, natural degradable composite films are based on natural polymers including cellulose, starch, chitosan and carrageenan. Among them, cellulose and carrageenan mainly exhibit favorable structural designability and film-forming performance; modified starch possesses advantages in cost reduction and processing performance improvement; chitosan stands out in food preservation due to its inherent antibacterial activity. Common preparation methods of natural degradable composite films include solution casting, extrusion blow molding, coating, electrospinning, multi-layer lamination and hot pressing. Solution casting fits formula screening at laboratory stage, extrusion blow molding is more suitable for industrial production, and coating facilitates the construction of functional layers on base film surfaces. In terms of application, composite films can be applied to meat, fruit and paper-based food packaging by blocking moisture and oxygen, inhibiting microbial growth and mitigating oxidation. Overall, natural degradable composite films can not only alleviate the environmental pressure brought by traditional plastic packaging, but also provide a new direction for the development of food preservation and green packaging materials.

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