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REVIEW

Recent Advances of Proteins, Polysaccharides and Lipids-Based Edible Films/Coatings for Food Packaging Applications: a Review

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Abstract
Most fruits and vegetables are susceptible to spoilage after harvest, the transportation and preservation conditions can further decrease the product quality. As one of the main food preservation technologies, edible films/coatings are made of eatable materials to form thin layers that can maintain food freshness during transport and storage. This review summarizes different films/coating materials, including proteins (soy, whey, wheat gluten, gelatin), polysaccharides (chitosan, starch, cellulose) and lipids. However, the films/coatings prepared by single material have many deficiencies which can be made up by the combination of composite films/coatings. Moreover, several prepared methods (electrospinning, casting, extrusion, dipping, fluidized-bed, spraying, panning) used for films/coatings are introduced. Finally, the application and future directions of films/coatings in the preservation of fruits, vegetables and other food products are also presented.

Keywords Edible films/coatings · Shelf life · Materials

Abbreviations

SPI	Soy protein isolate
WPI	Whey protein isolate
WGP	Wheat gluten protein
WVP	Water vapor permility
OP	Oxygen permility
EWP	Egg white protein
OEO	Oregano
TS	Tensile strength
EB	Elongation at break
CMC	Carboxymethyl cellulose

Introduction

It is well known that fruits and vegetables have a relatively short shelf life after harvest [1]. The abundance of nutrients and moisture in fruits and vegetables can cause microbial growth, sensory loss and decay [2]. A variety of physical and chemical methods have been used to maintain the quality and extend the storage life of fruit and vegetables [3]. Recently biodegradable edible films/coatings have attracted tremendous attention for food packages and storage. Edible films/coatings use edible natural polymeric materials (including proteins, polysaccharides, lipids, or complexes) as a film-forming material, with the incorporation of other functional compounds such as antimicrobials, antioxidants and anti-browning agents into their coating matrix to protect food from mechanical and microbial damages, reduces the water vapor permeability (WVP) and inhibits weight loss, then improve quality, safety and shelf-life of food products [4].

The coatings and films/coatings have the following functions (Fig. 1):

(1) Acts as a gas barrier [5] to control the gas exchange between fresh food and its surrounding atmosphere, reduce the rate of ethylene production and respiration, prevent the loss of natural volatile flavor compounds, and enhance the gloss and visual attractiveness of the food product.
(2) Acts as a moisture barrier to reduces food moisture loss, thereby maintaining food weight, flavor and appearance.
(3) The addition of active ingredients such as antioxidants and antibacterial agents can inhibit the occurrence of spoilage and browning caused by microorganisms.

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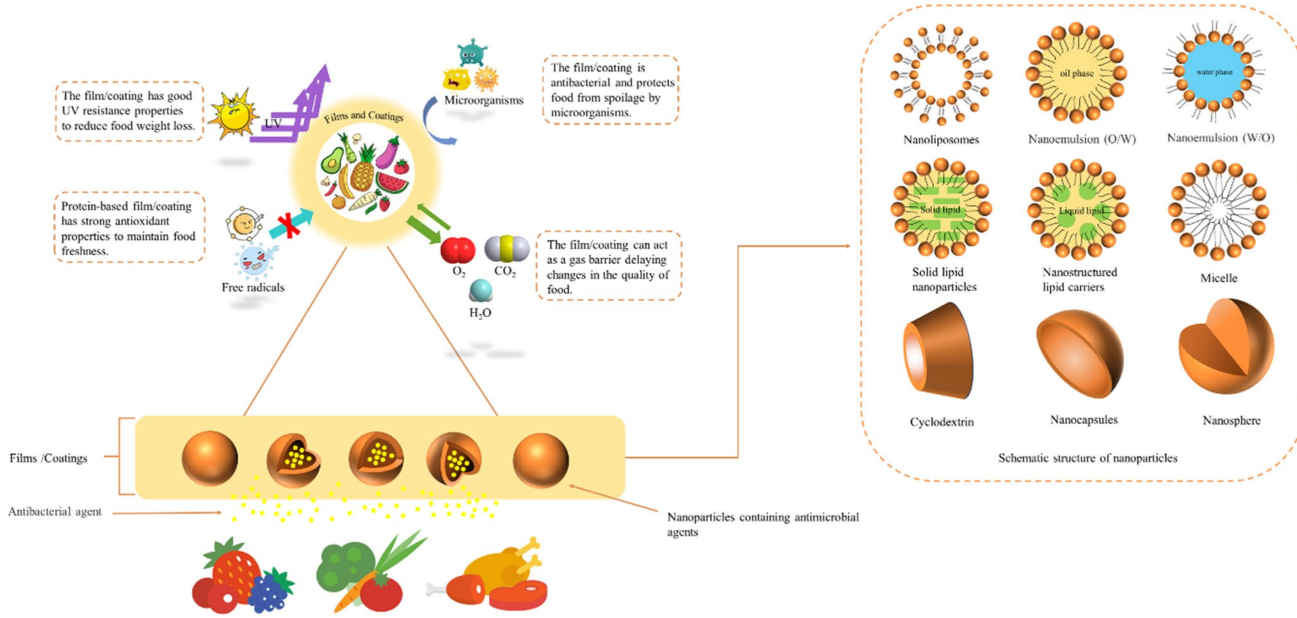

Fig. 1 Functional properties of edible films/coatings

(4) The addition of active ingredients such as antioxidants and antibacterial agents can inhibit spoilage and browning caused by microorganisms.
(5) These films/coatings materials can form nano-carriers for encapsulating additives and controlling the release of additives from the films/coatings.

In this review, the importance of edible films/coatings for food preservation are firstly summarized. Next, the biomaterials used and various preparation methods for edible films/coatings are discussed. The research progress of edible films/coatings to maintain freshness in fruits, vegetables, meats and other food products are also presented. Finally, the current challenges and future trends of edible films/coatings in the food processing industry are discussed.

Classification of Edible Films/Coatings

Several researchers developed numerous edible films/coatings from different materials (Fig. 2). According to the different materials selected, edible films/coatings are divided

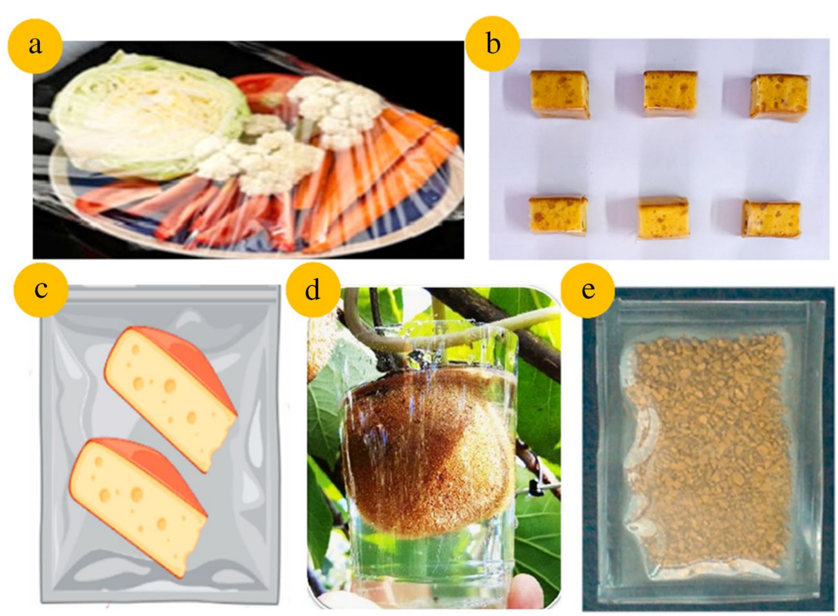

Fig. 2 Application of different types of edible films/coatings in various foods. **a** antimicrobial edible film is applied to vegetables [6]; **b** soy protein film is applied to muffin wrappers [7]; **c** bio-nanocomposite film is used in cheese [8]; **d** chitosan as an edible coating for kiwifruit [9]; **e** whey protein film is used in instant coffee [10]

into protein-, polysaccharide-, lipid-, and composite-based edible films/coatings. The commonly used types of these materials are shown in Table 1.

Protein-Based Edible Films/Coatings

Proteins are polymers made up of several amino acids linked to each other by peptide bonds. Amino acids are composed of central carbon bonded to amino, carboxyl, hydrogen and R groups. Proteins are compounds that can naturally exist in the form of fibrous or globular proteins. These protein-based materials are biodegradable used to prepare edible films/coatings [24]. It has been observed that the proteins have better mechanical properties than polysaccharides, owing to their distinctive structure, providing excellent barrier properties for aroma and oil. But, when compared to composite polymers, it has poor mechanical strength and greater water vapor permeability [25]. Soy isolates, whey proteins, wheat proteins and gelatin are often used to prepare films/coatings.

Soy Protein Isolate (SPI)

SPI is a globular protein isolated from soybeans, which is inexpensive and biodegradable with good oil and oxygen barrier properties and excellent film-forming properties. Its films are flexible and have smooth texture [26, 27]. It can be used on meat to prevent lipid oxidation and moisture loss, and is also applied as a coating to fruits and vegetables as it is highly permeable to water.

Zareie et al. [28] found that the tensile strength (TS) and elongation at break (EB) of the SPI-based films could be significantly increased with the addition of 2.5% chitosan, the edible film surface was denser and smoother with fewer cracks and pores.

Whey Protein Isolate (WPI)

WPI is a protein derived from cheese whey with high nutritional value and good digestion and absorption [29], which contains -OH and -SH and, can form adhesive films/coatings through disulfide bonds with hydrophobic interactions [30]. WPI-based films/coatings are colorless, odorless and transparent, with oxygen and oil resistance, but WPI is hydrophilic with a poor moisture barrier and plasticizers need to be added to overcome the brittleness of WPI-based films/coatings [31].

Vanden Braber et al. [31] prepared edible films/coatings using glycerol as a plasticizer, sodium tripolyphosphate as a neutralizer, WPI and a low concentration of water-soluble chitosan (WSCH) as the main ingredients. The results showed that the addition of (WSCH) reduced the solubility and elongation and greatly increased the surface hydrophobicity of the films. Moreover, the WPI/WSCH films showed good inhibition and barrier effect on the growth of fungal strains. Seiwert et al. [29] added xylan and transglutaminase to WPI to prepare composite films. The results showed that the composite film had a significant decrease in WVP,

Table 1 Applicable foods and functions of different materials films/coatings

Types of films/coatings	Substrate of the films/coatings	Food Products	Advantages	References
Proteins	Soy isolate protein	Eggshells, apples	Delays changes in color, hardness and acidity, reduction of microbial infection	[11]
	Whey isolate protein	Bananas, pears, cheese strawberries, apples, chicken, asparagus	Act as a gas barrier to reduce maturity, color change, hardness losses, mechanical losses and rancidity	[12–14]
	Wheat protein	Eggshell	good oxygen barrier and mechanical properties	[15]
	Gelatin protein	Strawberry, pineapple, blueberry, banana, pepper, eggplant, tomato	Decrease microbial growth and prolong the storage shelf life	[16, 17]
Polysaccharides	Chitosan	Fish, beef, tomatoes, carrots, asparagus, cabbage, blueberries, oranges	Biodegradable, non-toxic, antibacterial, good mechanical properties	[18, 19]
	Starch	Mango, green pepper	biodegradable, low cost, tasteless, good physical characteristics	[19]
	Cellulose	Strawberries, oranges, tomatoes, green peppers	Biodegradable, transparent	[19–21]
Lipids	Waxes	Lemon, orange, guava	Good water resistance	[22]
	Resin	Tangerine	Good water resistance	[5]
	Fats and oils	Kiwi, pork	Good water resistance, high tensile properties	[23]

oxygen permility (OP) and elongation, and significant increase in tensile stress and film strength.

Wheat Gluten Protein (WGP)

WGP is a hydrophobic protein composed of a combination of polypeptide molecules in the form of globular proteins [32]. The films formed by WGP can be used as a gas barrier on the surface of foods to reduce the permeability of oxygen and moisture and extend the shelf life of foodstuffs because of their extensibility, viscoelasticity and anti-oxidation properties. However, its drawbacks such as poor water resistance, fragility and poor mechanical properties [33] limit its application in food packaging. To overcome these weaknesses, WGP is often combined with other ingredients to prepare edible films. Dong et al. [34] modified WGP films with apple pectin (AP), carboxymethyl cellulose (CMC), egg white protein (EWP) and tartaric acid (TA) respectively. The results showed that WGP/AP-based films had better mechanical properties and improved film uniformity, WGP/CMC-based films had significantly improved thermal stability, WGP/EWP-based films had excellent physical strength and significantly improved opacity and light-shielding properties, WGP/TA-based films had excellent water barrier and UV resistance. Moreover, WGP and birch powder were designed into films/coatings for fruit preservation [35] (Fig. 3). The multifunctional films/coatings displayed excellent barrier properties to oxygen and water vapour and superior water-resistant, UV blocking, antimicrobial, degradation.

Gelatin

Gelatin is a colorless, odorless, water-soluble protein [35] due to its low cost, good biocompatibility and high safety, it commonly used in edible films [36]. Natural gelatin has the disadvantage of low antioxidant and antibacterial properties [37]. The addition of some active ingredients can improve its antioxidant and antibacterial properties. Roy & Rhim [38] prepared a gelatin-curcumin edible film, and the results showed that compared with the single gelatin film, the UV resistance of the composite film was increased, the antioxidant activity was improved and the antibacterial activity of escherichia coli and food-borne pathogens was significantly enhanced.

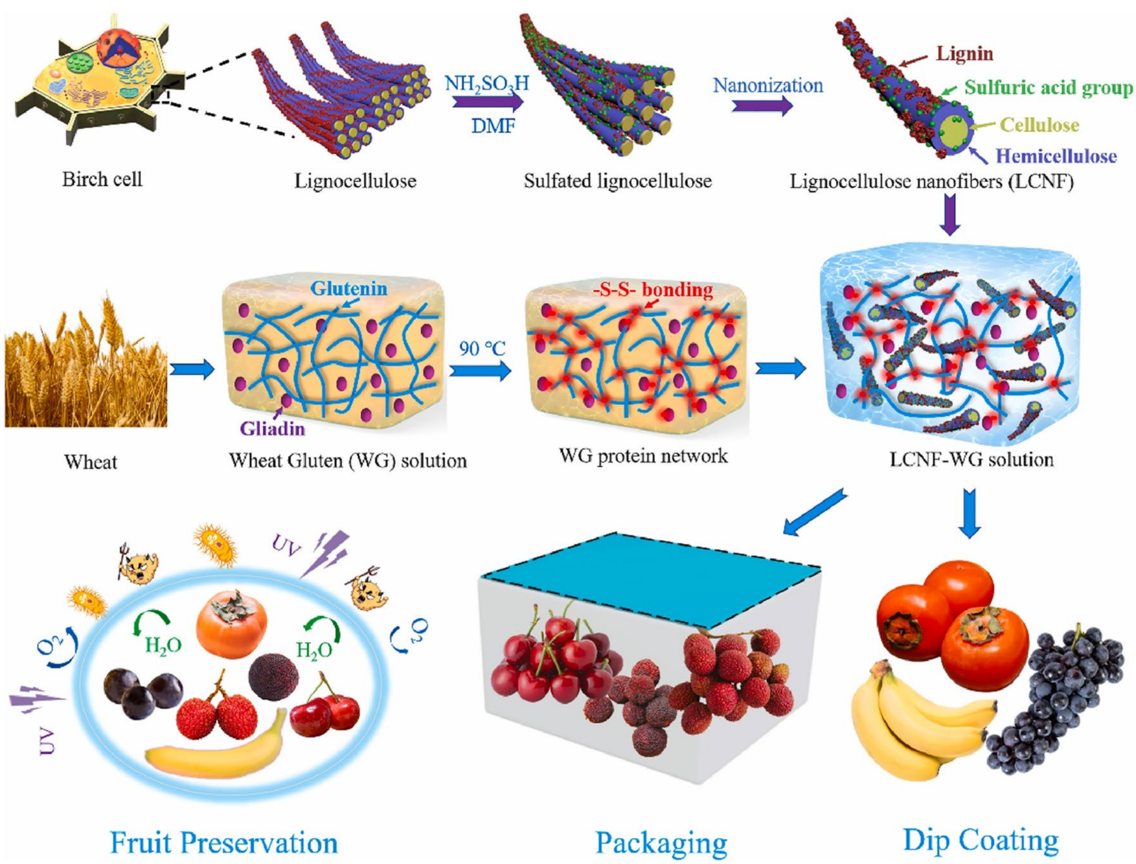

Fig. 3 Schematic illustration of preparation for lignocellulose nanofibers (LCNF)-Wheat gluten (WG) protein films/coatings for fruit preservation [34]

Polysaccharide-Based Edible Films/Coatings

Polysaccharide-based edible films/coatings are made of high molecular weight polysaccharides as the main materials, which form a dense networked film through interactions of intra- and intermolecular hydrogen bonding with good mechanical properties [39]. They have good barrier properties against O_2 and CO_2, but they have the disadvantage of being hydrophilic, resulting in a poor barrier against moisture loss in fruits. Polysaccharides such as chitosan, starch, and cellulose are utilized as biopolymers for the formation of edible films/coatings so that the typically used plastic packaging can be decreased.

Chitosan

Chitin is partially N-deacetylated to form biodegradable chitosan (Fig. 4) [40]. Due to its safe, biocompatible, antibacterial and film-forming properties, chitosan has been applied to food coating and packaging to prevent moisture and oxygen permeation. Chitosan films/coatings reduce the partial pressure of oxygen in the package and have excellent antimicrobial characteristics. It reduces the fruit browning due to enzymatic reactions and suspends losses due to dehydration. The chitosan also enhances the emulsifying effect which resulted in the improvement of the flavor, texture, and color stabilization [41]. Pavinatto et al. [42] successfully prepared chitosan-based

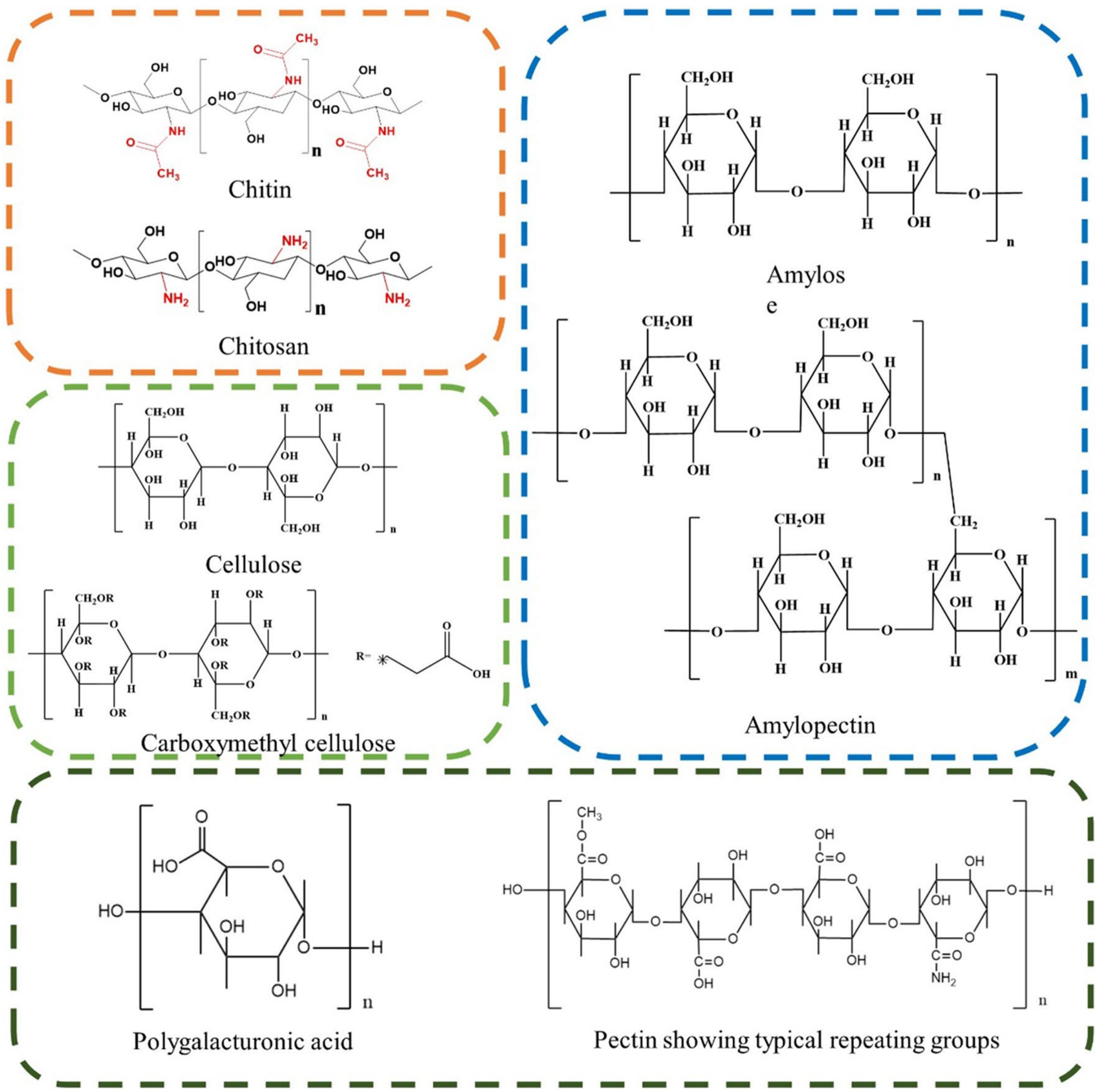

Fig. 4 Chemical structural of some polysaccharides

films containing glycerol for strawberry protection, the results showed that the films were highly hydrophobic and were able to protect strawberries from fungal attacks without altering their appearance, texture and flavor. Duran and Kahve [43] investigated the effect of vacuum packaging and chitosan-based film on beef storage. The results showed that the combination of vacuum packaging and chitosan-based films effectively reduced thiobarbituric acid, thermophilic aerobic bacteria, lactic acid bacteria and total volatile basic nitrogen values and completely inhibited S. aureus.

Starch

Starch is the main polysaccharide in plants and consists of amylose (relative molecular mass of $3.2\times10^{4}\sim1.6\times10^{5}$) and amylopectin (average relative molecular mass of 10,000-200 million) (Fig. 4). Amylose is formed by a combination of α-1,4-glycosidic bonds linked to D-glucopyranose units, and amylopectin consists of a straight-chain part (α-1,4-glycosidic bonds linked to D-glucopyranose) and a branching part (α-1,6-glycosidic bonds linked to D-glucopyranose) together. Among them, straight-chain starch plays an important role in film formation [44]. Starch-based films/coatings are odorless, transparent and bioabsorbable and the most commonly used starch for film formation is obtained from potatoes, wheat, corn, etc. [45, 46]. However, owing to the hydrophilicity of starch, starch-based films present water solubility and poor water vapor barrier.

Cellulose

Cellulose is the main component of plant cell wall and it is a macromolecular polysaccharide formed by β-1,4 glycosidic bond connecting D-glucopyranose, which is non-toxic, moisture-proof and biodegradable with good film-forming properties (Fig. 4). Due to the poor solubility of natural cellulose, modified cellulose such as CMC and hydroxypropyl cellulose is generally used to prepare coatings [1]. CMC edible films/coatings are usually tasteless, bendy, and are of low dietary energy, transparent, resistance to oil and fat, moderate to oxygen diffusion and moisture. Forato et al. [47] prepared edible coatings based on CMC and cashew gum (CG) with glycerol as a plasticizer for fresh-cut guava. Compared to uncoated guava, the CMC/CG/glycerol coating was able to reduce mass loss, delay color change and tissue decay and extended the shelf life of guava.

Pectin

Pectin is one of the main components of the plant cell wall chemically constituted by poly α-1,4- galacturonic acids, with varying degree of methylation of carboxylic acid residues and/or amidated polygalacturonic acids [48] (Fig. 4). Pectin is an appropriate polymer material for preparing edible films/coatings due to its biodegradability, biocompatibility, edibility and various chemical and physical properties (such as gelation and selective gas permeability), which could diminish respiration rate and food oxidation to a great extent, eventually leading to the protracted shelf life of food products [49]. These unique properties could retain the nutritional properties, prevent unwanted alterations (like enzymatic browning, off-flavor formation, aroma loss, retardation of lipid migration) of unpreserved food, and promptly, prevent pathogen attack while storage [50]. However, pectin films/coatings have higher WVP and brittleness due to hydrophilic nature. Therefore, pectin films/coatings often blend with other substances to weaken the hydrophilicity in high moisture environments [51].

Sun et al. [52] prepared composite films with carboxymethyl chitosan (CMCS), pectin, blueberry anthocyanins (ACNs) and clove oil as raw materials. The hydrogen bonding and electrostatic interactions existed and maintained the network structures of films, which reached the most stable when the ratio of pectin and CMCS was 1:4. The addition of ACNs and clove oil could significantly enhance the water resistance, UV barrier properties, antioxidant activities and antibacterial properties, thus achieving the multifunctional activity of the films. Moreover, the edible films had also been proven to have a certain effect on delaying fruit senescence and ensuring postharvest quality, which was mainly achieved by the edible films forming microchambers with food products to avoid the rapid evaporation of water. For example, packaged strawberries stored in this condition still had good morphological structure after 7 days of storage. Danila et al. [53] prepared chitosan-pectin edible films/coatings. The formulation that showed the best properties for packaging dry foods was the one with 70 wt% of chitosan and 30 wt% of pectin. Because chitosan and pectin interact through electrostatic force, it is helpful to improve the elastic modulus of the films/coatings, reduce the WVP and OP, increase its thermal stability and reduce its water solubility. In addition, the composite films/coatings showed high antioxidant activity, good transparency. In another study, composite films were prepared with pectin and polyphenols obtained from watermelon peel as raw materials to delay the qualitative degradation of chilled mutton during super-chilled storage. The barrier properties, mechanical properties, light transmittance, and thermal stability of pectin-polyphenols films significantly improved at a polyphenol concentration of not more than 1.5% due to the formation of hydrogen bonds between pectin and polyphenols. With an increase in the storage time, the pH, total volatile basic nitrogen and total bacterial count values of mutton in the super-chilled film group displayed the lowest increase compared to the mutton in the refrigerate and super-chilled groups [54].

Lipid Edible Films/Coatings

Various waxes (beeswax, carnauba wax, candle wax, etc.) and resins (shellac resin, coumarin resin, turpentine, etc.) are often used as lipid coating components [39]. Lipids possess good moisture barrier properties and prevent food products from deterioration due to their hydrophobic nature, the formed films/coatings have good water resistance and are mainly used for surface coatings of fruits and vegetables to maintain gloss and brightness. Due to the flexibility of lipid films/coatings being poor, lipids are usually used in combination with starch, chitosan and protein to prepare films/coatings [55].

Khanzadi et al. [55] prepared films using different ratios (70:30, 50:50 and 30:70 (% w/w)) of whey protein concentrate (WPC): prululam (PUL) and glycerol at 20% (w/$w_{drymatter}$) as a plasticiser, in addition to 0, 10, 20 and 30% (w/$w_{glycerol}$) of beeswax. By measuring their thickness, water content, water solubility and water vapor permility, it was found that 30% beeswax and WPC : PUL (70:30) were the best films, with good water solubility, water vapor permility and elongation. Ma et al. [56] prepared composite coatings made from shellac and tannic acid in comparison with the coatings prepared from shellac alone. The results showed that the composite coatings improved its antifungal activity, reduced lipid oxidation, improved the overall quality of mangoes and extended their post-harvest storage life.

Composite Edible Films/Coatings

The composite edible films/coatings consist of two or more materials of proteins, polysaccharides and lipids. The composite films/coatings combine the advantages of each component, overcome the limitations of single-component films/coatings with excellent mechanical properties and barrier properties [28]. Composite films/coatings mainly include protein-protein composite films/coatings, protein-polysaccharide composite films/coatings, polysaccharide-polysaccharide composite films/coatings, and protein-polysaccharide-lipid composite films/coatings.

Protein-Protein Composite Films/Coatings

The film-forming ability of proteins is affected by ionic bonds and disulfide bonds. The interaction strength between protein chains determines the mechanical strength and barrier properties of edible films/coatings, different sources of protein have different mechanical properties and barrier properties [57]. Zein has good water barrier properties and heat sealability, but is fragile, whey protein-based films are colorless, highly transparent and have barrier properties under low humidity conditions [58]. Therefore, Tsai and Weng [59] prepared composite films of WPI and zein in three ratios (2:1, 1:1, 1:2). The results showed that the composite films had good heat sealability and water solubility. It overcame the shortcomings of poor heat sealability of WPI-based films and water insolubility of zein.

Protein-Polysaccharide Edible Films/Coatings

Protein-based films/coatings have good flexibility and good gas barrier properties, but their mechanical properties are poor. Polysaccharides have stable film-forming properties and good biocompatibility, but their water barrier properties are poor [59]. The composite edible films/coatings prepared by protein and polysaccharide can overcome the shortcomings of one material alone and improve the mechanical properties and barrier properties of the films/coatings. Zhou and Wang [60] prepared zein and methylcellulose composite films with oleic acid and polyethylene glycol as plasticizers, and the mechanical properties of the films were measured. The elongation and TS of the composite films were higher than films prepared from a single material. In the composite films, methylcellulose improved the TS and elasticity of the composite films and zein reduced water vapor permility and solubility. Huang et al. [59] found that with the increase of EWP content, and light transmittance of the EWP and κ-carrageenan composite films increased to 10.85% and 53.3%, respectively.

Polysaccharide-Polysaccharide Edible Films/Coatings

Polysaccharide edible films/coatings have excellent stability due to the formation of hydrogen bonds in the process of films/coatings formation, but its water resistance is poor. Therefore, two or more different polysaccharides can be used to make composite films/coatings to make up for the shortcomings of single polysaccharide-based films/coatings. Ma et al. [61] prepared four chitosan-based composite films (konjac glucomannan/chitosan, tapioca starch/chitosan, maltodextrin/chitosan, gelatin/chitosan). The results showed that the konjac glucomannan/chitosan-based composite films showed the strongest mechanical properties and the highest transparency, and the tapioca starch/chitosan-based composite films showed the highest water resistance and better protective properties. Lan et al. [62] prepared corn starch/CMC/Nisin-based composite films. The addition of CMC improved the mechanical properties of the corn starch films and the EB increased from 49.37 to 86.75%. Since nisin has a high inhibitory effect on Gram-positive bacteria, the film showed excellent antibacterial activity with an inhibitory rate of 60.75% against staphylococcus aureus.

Protein-Polysaccharide-Lipid Composite Films/Coatings

Protein-based films/coatings and polysaccharide-based films/coatings have good mechanical properties and hydrophilicity, but the water resistance is poor. The lipid films/coatings have good water resistance, but their mechanical properties and film-forming properties are poor [41]. Therefore, the protein-polysaccharide-lipid-based composite films/coatings can be prepared to improve the comprehensive properties of the films/coatings. Khanzadi et al. [56] produced whey protein, tapioca starch and beeswax composite films, the addition of hydrophobic beeswax increased reduced WVP. The mechanical properties were affected by the content of different components, the composite films with 3.0% chitosan, low content of beeswax and high content of whey protein exhibited the highest EB.

Additives

In addition to the main preparation materials of edible films/coatings, some additives can also be added to enhance the functionality of the films/coatings. Plasticizer is a low molecular substance, which can interact with polymer matrix in films/coatings to form hydrogen bond (Fig. 5a), reduce TS of films/coatings and increase flexibility of films/coatings (Fig. 5c) [39]. Commonly used plasticizers are glycerol, sorbitol, polyethylene glycol, etc. Huntrakul and Harnkarnsujarit [63] studied the effect of different plasticizers on the water absorption of WPC-CMC blend films, and the results showed that sorbitol greatly improved the stability of blend films resistance and water resistance.

Emulsifiers are a class of macromolecular stabilizers that reduce the surface tension at the interface of the two phases to make the mixed liquid of two immiscible components form a stable emulsion (Fig. 5b), which can maintain the balance of hydrophilic and lipophilic to prevent phase separation [41]. Span 80, Tween 80, lecithin and monoglyceride fatty acid glycerides are commonly used emulsifiers in the food processing industry. Rashid et al. [67] used Tween80 as an emulsifier to configure a polysaccharide-based edible coating for postharvest storage of apples. The results showed that the coating-based apples had less weight loss, strong antibacterial properties, improved quality and safety, and increased shelf life and stability. Mendes et al. [65] also used Tween80 as an emulsifier to prepare an edible film that improved the mechanical properties of the film and increased the TS of the film.

In order to enhance the antibacterial properties of edible films, the use of antibacterial agents is essential. Among them, EOs, as a natural food preservative, is applied to edible films to prolong the shelf life of food due to its antibacterial property. Phenolic components of EOs can destroy plasma membrane, change microbial cell permeability, inhibited proton power and interfere with cell energy (ATP) production system. As a result, the permeability of the plasma membrane is damaged, which eventually leads to cell death. The antibacterial mechanism of EOs is shown in the Fig. 5e. Aitboulahsen et al. [67] studied the effect of mentha pulegium essential oil (MEO) on the physical and chemical properties of gelatin-based coating. Figure 5d showed that compared with the control, the gelatin coating of MEO significantly inhibited total aerobic mesophilic flora (TAMF) and yeast and mold (YM).

Preparation Method

Electrospinning

Electrospinning is a special technology for fiber films preparation, in which biopolymer-based film-forming solution is jet spun in strong electric field [68]. Electrospinning device is equipped with a high voltage power supply, syringe pump and collector [69] (Fig. 6a). In the process of electrospinning, the solution is added into the syringe and the droplet at the tip of the needle will change from a sphere to a cone. With the help of an electric field, the filament is obtained from the tip of the cone. In this way, the fiber on the nanometer scale can be obtained. Nanofibers are collected on the collector to form nanofiber films. It can generate nanofibers with a more uniform diameter, higher surface area-to-volume ratio, richer composition and thinner film compared with other methods [70].

For example, the nanofiber film composed with Zanthoxylum bungeanum essential oil (10%) and polyvinyl alcohol/β-Cyclodextrin (6:1) complex was innovatively fabricated by electrospinning technology, providing much longer-term protection for both strawberry and cherries to avoid the microbial infection and prolong the shelf life of the berry fruit. The results showed that the average diameter of nanofibers was 185 nm, which showed sufficient mechanical properties. TS and EAB were 10.69 ± 1.97 MPa and 268.64 ± 64.28 MPa, respectively. In addition, this study proved that film prepared by electrospinning had good potential as antifungal fruit packaging material, with remarkable antifungal activity and antiseptic performance [71].

Nozzle-less electrospinning devices can produce food-grade nanofibers with high production yield compared with nozzle-based instruments on a large scale, which can make up for the shortcomings of the above methods. Altan A., & Çayir Ö`. [66] prepared chitosan/gelatin nanofibers films coated with thyme essence by nozzle-less electrospinning. The results showed that the nanofibers with chitosan/gelatin ratio of 1: 6 and 40% essence had suitable diameter (150.8 ± 50.5), high stability, antibacterial property and oxidation resistance.

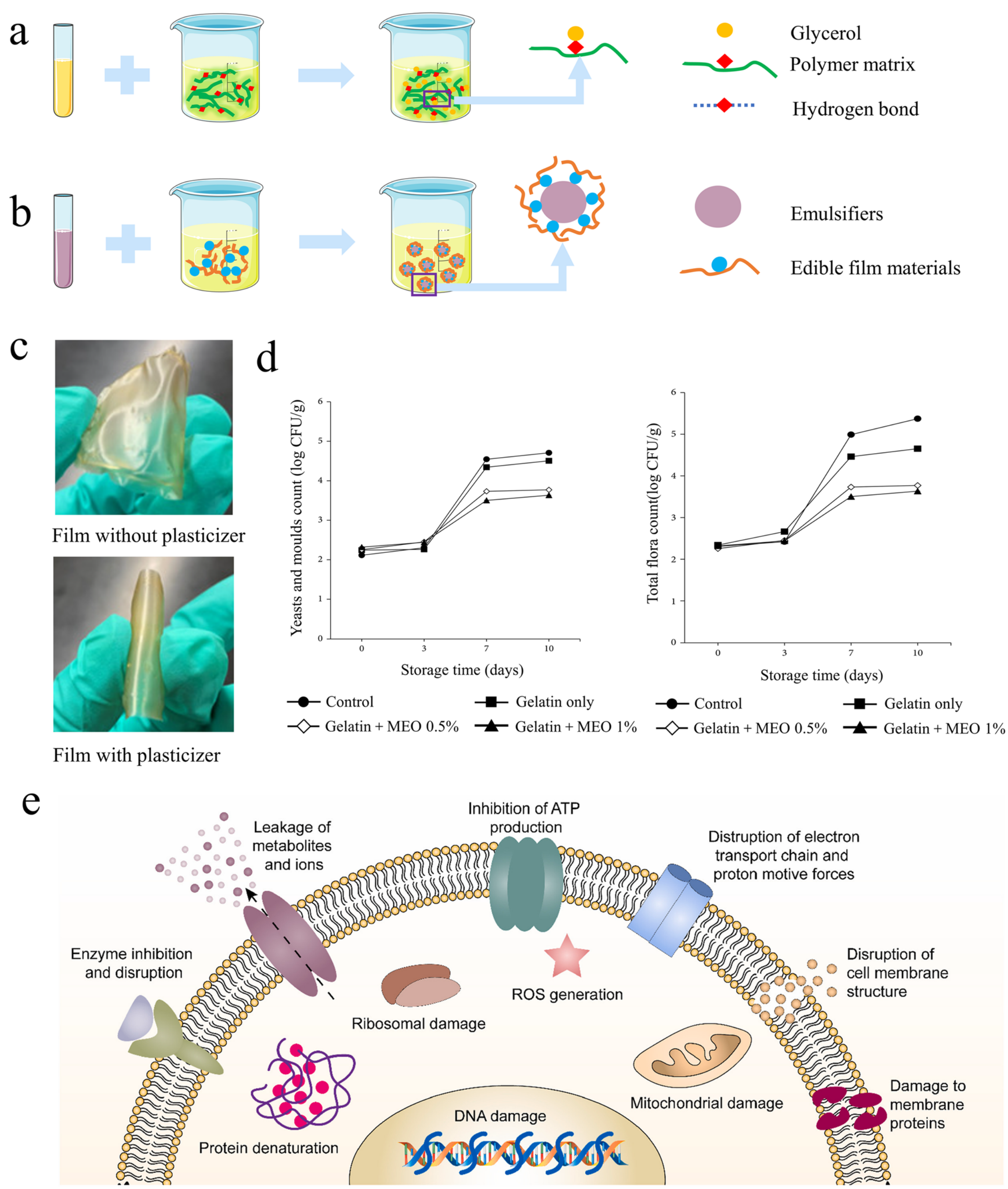

Fig. 5 **a** and **b** represents the formation mechanism of plasticizer and emulsifier respectively; **c** effect of plasticizer on film, copyright permission from [64] **d** antibacterial effect of mentha pulegium essential oil (MEO), copyright permission from [65]; **e** antibacterial mechanism essential oil, copyright permission from [66]

Casting Method

The casting method is one of the most commonly used methods for preparing edible films (Fig. 6b). The main steps of this method are: (i) dissolution, (ii) casting, (iii) drying. First, the film-forming material is dissolved in the solvent by stirring [72]. Then the prepared solution is poured into a specific mold to prepare a film [42], Since different temperatures during drying can affect the properties of the films, the selection of drying conditions is generally considered when prepared by casting method. Rodríguez et al. [73] prepared papaya edible films with antioxidant activity by casting method. The results showed that the films dried at room temperature showed

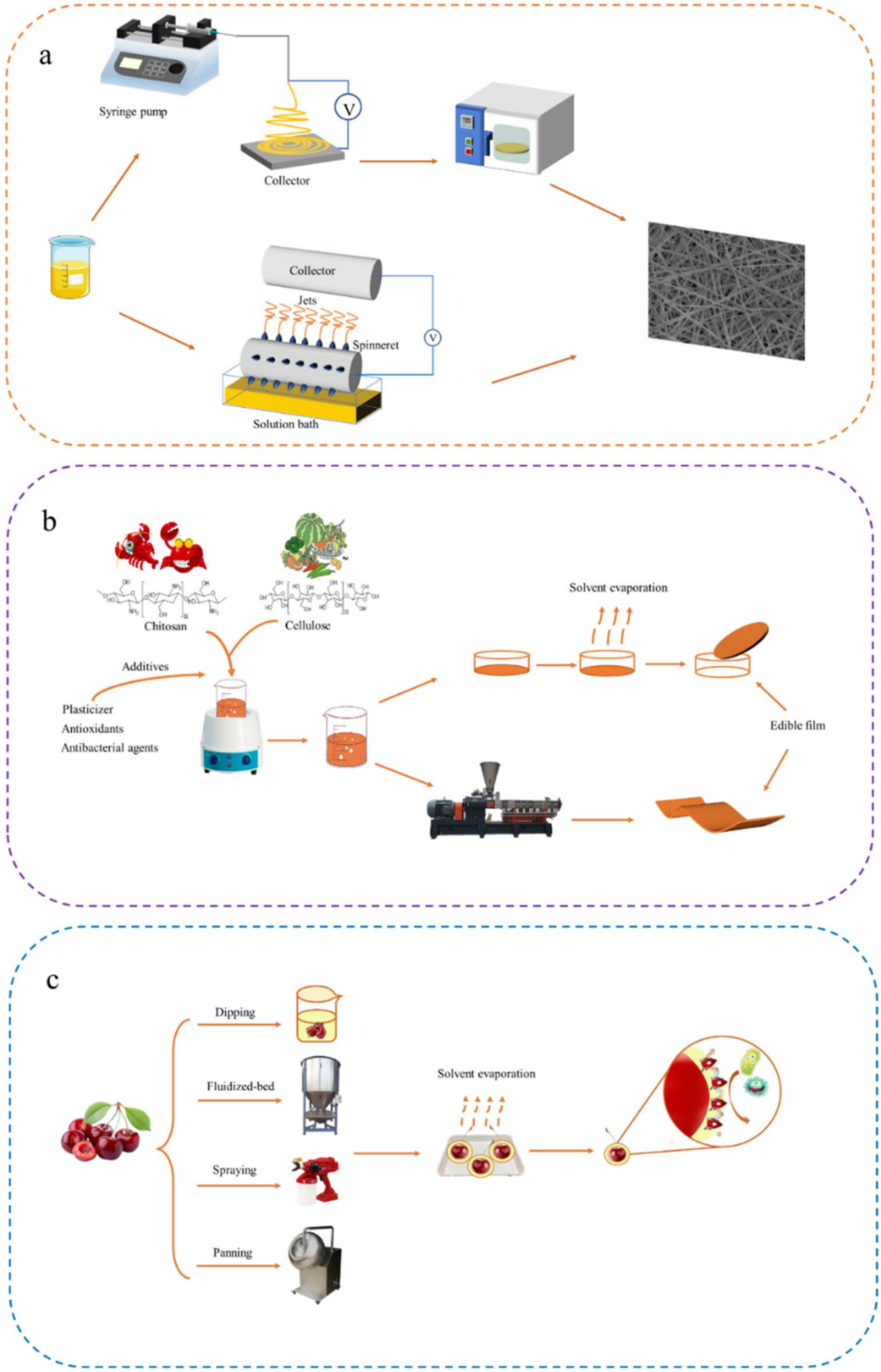

Fig. 6 Various preparation methods. **a** electrospinning, **b** edible films, **c** edible coatings

the highest stability. The color of dry film is dark yellow, which is due to non-enzymatic browning of polyphenols in papaya film at high temperature. The casting method did not require special instruments and equipment, and had a simple operation and low cost.

Extrusion Method

The extrusion method is often used for commercial scale film processing [74], (Fig. 6b). Firstly, all materials were fully mixed by a twin-screw extruder, and the temperature gradient was set from the feed area to the die area according to the experimental requirements, the mixture was extruded to prepare particles, then the particles were dried. Finally, the films were prepared by single screw extrusion mechanism [75]. During extrusion, plasticizer was added to improve the mechanical properties of films and the type of plasticizer has an impact on the film-forming ability.

Park et al. [76] studied the effects of three plasticizers (glycerol, sorbitol, glycerol and sorbitol mixture (1:1)) on the extrusion films. The results showed that the films added with glycerol showed the highest TS and transparency, but the films added with sorbitol were not stretchable. Therefore, sorbitol was not suitable for extrusion as a plasticizer. Colak et al. [77] used the extrusion method to obtain lysozyme films. It was very important to adjust the extrusion process conditions (barrel temperature, screw speed, moisture content others) to maintain the activity of the lysozyme.

Dipping Method

In the dipping method, food is immersed in the prepared coating solution at a constant speed to form the film on the food surface, and finally the coating is dried [71]. The method is widely used in fruits and vegetables (Fig. 6c). Ait-boulahsen et al. [65] prepared gelatin-based edible coatings containing peppermint essential oil to prolong the shelf life of strawberries by dipping method. Compared with uncoated strawberries, the coatings could significantly inhibit mold and yeast, the changes in pH, weight loss, hardness and color of strawberries were also slowed down. The gelatin coating containing 1% Mentha pulegium essential oil could protect at least 60% of strawberries from deterioration after 13 days of storage.

Fluidized-Bed Method

The fluidized-bed coating is a technology used to coat dry particles with a very small diameter and low density [78]. Fluidization occurs when the fluid flowing upward reaches a sufficient speed to support the particles without taking the particles away in the fluid. After nebulization by the nozzle, the droplets adhere and diffuse on the particle surface and finally, the solvent evaporates to form the film. In this way, the coating solution is completely covered on the surface of the particles in the fluidized bed for a certain time. Fluidized bed technology is divided into three types: top, tangential and bottom jet fluidized bed [71]. Compared with other fluidized bed technologies, the top spray fluidized bed has high versatility and is suitable for hot-melt coating in the food industry (Fig. 6c). It has been studied [79] that the surface of peanuts is coated by fluidized bed technology. Compared with pan coating, the surface of peanuts coated by fluidized bed technology is more uniform and consistent and the drying time is shorter.

Spraying Method

The spraying method (Fig. 6c) is to spray the coating solution onto the surface of the product to form a film with uniform thickness [16]. In the spraying process, the coating solution will evaporate from the product surface, reducing the drying time of the coatings. Spraying technology can be used alone, or spraying matrix solution with a nozzle is also involved in a pan and fluidized bed technology. Some studies [80] have applied xanthan gum-based coating on the surface of fresh-cut lotus root by spraying technology. The results showed that compared with the untreated lotus root, spraying technology could effectively reduce the enzymatic browning of lotus root during storage. In recent years, electrostatic spraying technology is widely used in food coating. Compared with traditional spraying technology, this technology can control the size of droplets. Peretto et al. [81] applied electrostatic spraying technology to fresh strawberries. Compared with traditional spraying, electrostatic spraying improved the quality of strawberries and prolongs the storage time. Jiang et al. [82] also used electrostatic spraying for strawberries coating. The results proved that electrostatic spraying could form a more continuous and uniform film and prolonged the shelf life of strawberries.

Panning Method

The panning method is to put the product into a large rotating pan, (Fig. 6c) then coating solution is sprayed onto the surface of the product to form a coating layer through the nozzle, and the surface of the product is fully coated with the solution during rolling [83] and dried at high temperature. The panning method is most suitable for the coating of candy and nuts, because the shape of the candy and nuts is not easy to be damaged in the process of rolling [71]. For example, pan coating can be applied to the surface of the chocolate to increase brightness and smoothness, the operation is simple and can repeatedly coat the products.

Characterization of Edible Films/Coatings Properties

Mechanical Properties

Edible films must have good mechanical properties and can resist physical damage. This physical damage (external force) may occur in food processing, handling and storage. The mechanical properties of edible films show the ability to resist external force and maintain good shape and quality of food [84]. The mechanical properties are generally evaluated by TS and EB. TS is the maximum force that the film can bear without breaking, and high TS results in high toughness of the films. EB is the extent to which the film stretches or elongates before the breaking point, representing the flexibility of films. The mechanical properties of edible films are also related to the interactions between the molecules, which may be enhanced by the incorporation of some active ingredients.

The effects of different concentrations of thyme glycerol (a glycerol extraction of thyme, TEO, 0%, 0.4%, 0.8%, 1.2%, 1.6%) on the mechanical properties of edible films were studied [85]. The results showed that with the increasing concentration of TEO, the TS decreased from 40.35 MPa to 23.86 MPa. The reason may be that with the addition of TEO, the interaction between polymer and TEO gradually replaced the interaction between polymer and polymer, which weakened the network structure of edible films and reduces TS. However, with the increasing concentration of TEO, the EB of the films increased from 6.98 to 12.41%. Akyuz et al. [86] evaluated the effects of different oils and fats on the mechanical properties of edible films. The results showed that the addition of olive oil and corn oil increased the TS of the films compared to the chitosan films without oils, the TS and EB of the olive oil/chitosan films were both increased. Olive oil contains the long chain with the highest monounsaturated oil ratio, which improves the flexibility and ductility of the film. The interaction between olive oil and chitosan improved the TS and EB. However, the TS of the film decreased with the addition of butter and animal oil, and the TS of the film did not change significantly with the addition of sunflower oil.

Permility of Oxygen and Water Vapor

Excessive oxygen or water will improve microbial growth and lipid oxidation of food in the film. Therefore, the good barrier ability of oxygen and water vapor can improve the shelf life of food [87]. Oxygen permility (OP) and Water vapor permility (WVP) are two important parameters to evaluate the oxygen resistance and water resistance of films [88]. Therefore, keeping the OP and WVP of the film as low as possible is very important for food preservation.

Cross-linking of some substances with film materials can reduce the OP. The WVP of the potato starch film decreased due to the addition of clove essential oil, this was due to the hydrogen bond between the hydrogen group of potato starch and water being replaced by the hydrogen bond of clove essential oil and starch, which reduced the hydrophilicity of the film [89]. However, the addition of hydrophilic γ-Amino acid to WPI film will increase WVP, because γ-Amino acid and WPI produced electrostatic repulsion, which leads to the formation of many pores in the film to increase WVP [90].

Different concentrations of casein phosphopeptides (CPPS, 0-0.5%, w/v) were added to gelatin-based films to observe the effect of CPPS on the WVP of films [91]. The results showed that 0.1%, 0.2% and 0.3% CPPS reduced the WVP value of the film. The phosphate group in CPPS interacts with gelatin polymer, resulting in a dense network structure in the film. This interaction changed the orientation of proteins in the film, reduced hydrophilic functional groups, increased hydrophobic amino acids, so the WVP of the film was decreased. Trinh et al. [46] discussed the effects of modified CNC on the OP and WVP of starch/beeswax edible film. The results showed that the OP of the film decreased with the increase in CNC concentration. CNC promoted the uniform mixing of beeswax and starch, and CNC has excellent oxygen resistance, which can improve the oxygen resistance of beeswax/starch. The beeswax/starch composite film prevented the fruit from oxygen and maintained the appearance, freshness and moisture content of the fruit, which prolonged the shelf life of the fruit.

Antioxidative and Antibacterial Properties

Antioxidative and antibacterial properties [92] are extremely important functional properties of edible films/coatings, which prevent food oxidative deterioration and protection from microbial contamination, maintain food quality and thus extend shelf life. Plant extracts and essential oils [93] can release active substances to protect food from free radicals, oxidative intermediates and secondary decomposition products, so both have good antioxidant and antibacterial properties to protect food freshness.

Roshandel-Hesari et al. [94] developed oregano essential oil (OEO)-chitosan-casein edible films and investigated the effects of OEO on the antioxidant and antibacterial properties of chitosan-casein composite films. The results showed that 1.5% OEO had the strongest inhibitory effect on Escherichia coli and Staphylococcus aureus with a good antioxidant effect. Therefore, adding OEO to the chitosan-casein composite film could significantly improve the antioxidant and antibacterial properties of the composite film and prolong the shelf life of food. Boeira et al. [95] developed a corn stigma extract (CSE)/gelatin/corn starch composite film, and the different concentrations (0, 15 and 25%, w/v) of CSE on the antioxidant effect of the gelatin-corn starch composite film were studied. The results showed that 25% CSE made the composite films with the strongest free radical scavenging ability and its antioxidant capacity was increased by 60% compared with the composite films without CSE. Since the corn stigma extract was rich in biologically active substances such as phenols, flavonoids and terpenes, it had the good antioxidant capacity and had the potential for food preservation and prolonged shelf life.

Application of Edible Films/Coatings in Food

Fruits and Vegetables

Due to the high water content and rich nutrition, fruits and vegetables are prone to water loss, shrinkage and mechanical damage during transport and storage, leading to quality deterioration and short storage life. After picking, some fruits reach maturity through their production of ethylene [96]. For example, bananas, kiwis and peaches produce more and more ethylene over time after they are picked, which converts starch into sugar, making it more palatable. But in the process, a lot of heat was generated which caused the fruit to spoil in a short time. Therefore, for these climacteric fruits, edible coatings are used to inhibit the growth of microorganisms and maintain the flavor of the fruit before climacteric. La et al. [97] successfully coated bananas with chitosan/arabic gum/ZnO coating solution by dipping method. They studied the effect of the compound solution at 35 ℃ and 54% relative humidity on the storage life of bananas. The results showed that the smooth surface of banana treated with chitosan/arabic gum/ZnO coating could maintain the hardness, weight and shelf life of the banana for a long time, the best protection performance was found when the ZnO content was 0.5% (w/w).

Nonclimacteric fruits such as cherries, oranges and grapes produce less ethylene and do not continue to ripen once picked. These fruits have high nutritional properties, good sensory properties and limited shelf life. To extend the shelf life of these fruits, edible coatings are applied to these fruits. Zhang et al. [92] studied the effects of gelatin/carboxymethyl chitosan/calcium chloride/ascorbic acid composite edible coating on cherry quality and antioxidant activity. The results showed that compared with cherries without coating, coated cherries had a lower weight loss rate, respiration rate and browning rate. In addition, coated bananas kept the hardness, water content and high antioxidant activity. When fruits and vegetables are treated with edible coatings, water loss is prevented and the surface of vegetables is maintained by reducing oxygen and water vapor permility. Antimicrobials in edible coatings also prevent spoilage, thus increasing storage time. Zahedi et al. [98] studied the effects of edible coatings prepared with different concentrations of chitosan and polyamine imine on improving the shelf life and quality of mango. Compared with the control group, chitosan (2%) and spermidine (2 mM) can significantly improve the hardness and shelf life and delay the deterioration process of mango.

Hernández-Carrillo et al. [99] studied the effect of reuterin in pectin-based edible coated films added with lemon essential oil on the quality of frozen strawberries, preparing pectin-based solutions (3%, w/v) (T1), T2 preparations incorporating 2% (v/v) lemon essential oil (TEO), while T3 was admixed with LEO and reuterin (10 mM). Physicochemical and microbiological parameters were evaluated during 31 days of storage. The coated samples showed a more intense and reddish color than the control samples (Fig. 7a). Edible coatings with pectin as the main raw material, added with LEO and reuterin, have a great potential for strawberry preservation, since these coatings are able to avoid fungal spoilage without degrading the quality. Chen et al. [100] established an anthocyanin treatment method that can significantly delay the ripening of banana fruits stored at 23 ± 2 ℃. The results showed that anthocyanin treatment could delay the yellowing of banana peel and decrease of flesh hardness (Fig. 7b). Mei et al. [101] prepared a composite film containing sericin, chitosan as well as carbon dots, and used it for lychee preservation. The results showed that the composite film used in the postharvest preservation of lychee was helpful to delay the loss of nutrients during storage and keep the good quality of lychee fruit (Fig. 7c).

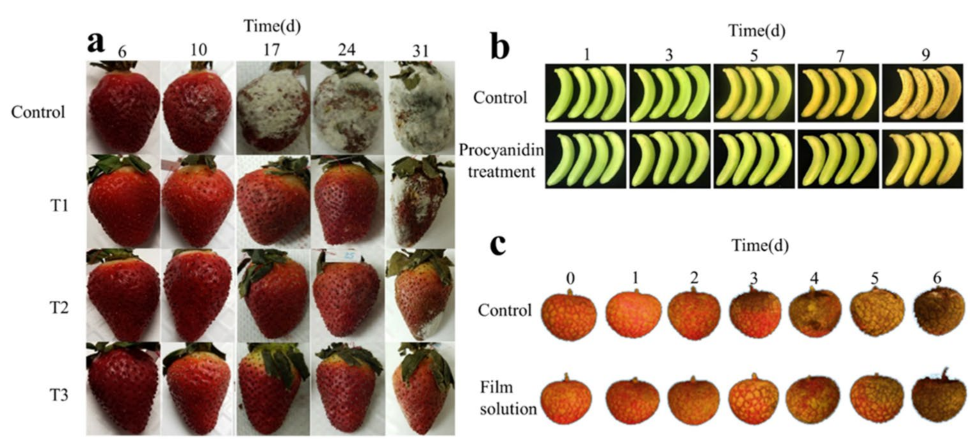

Fig. 7 **a** Visual appearance of uncoated (Control) and coated strawberries during cold-storage (T1: Pectin; T2: Pectin + LEO; T3: Pectin + LEO + Reuterin); copyright permission from [98], **b** Changes in appearance; copyright permission from [85], **c** The appearance changes of lychee fruits treated with film solution and control lychee fruits during storage. copyright permission from [95]

Meat

Meat provides a good environment [102] for the growth of microorganisms due to the presence of various nutrients (protein, minerals, vitamins, etc.) and sufficient moisture, which will cause the degradation of fresh meat. Edible films/coatings for meat can act as a gas barrier, with antimicrobials and antioxidants that maintain the quality and safety of fresh meat and delay its deterioration.

Ruan et al. [103] prepared edible coatings containing epigallocatechin gallate, sodium alginate and CMC to study the effects on the quality of pork. The results showed that the edible coating could reduce weight loss, improve the color and odor of pork. In addition, the edible coating significantly inhibited the lipid oxidation and microbial growth of pork to prolong its shelf life. Criado et al. [104] prepared cellulose nanocrystals/sodium alginate edible films by casting method. The results showed that the edible film could effectively reduce OP and prevent the oxidation of chicken breasts. Du et al. [105] added rosemary extract into fish myofibrillar protein/chitosan composite films for the preservation of fish meat. The results showed that pH value, free amino acids and total volatile nitrogen in the rosemary extract group were lower than those in the control group at the same storage, which indicated that the composite film could reduce lipid oxidation, inhibit microbial growth and prolong the shelf life of fish. Velásquez et al. [106] wrapped beef in k-carrageenan films containing bee pollen and honey extracts and evaluated the effect of the films on the preservation of beef. The total phenolic content, antioxidant, radiation resistance and antibacterial activity of the films were determined. The films did not significantly improve the antibacterial properties, but significantly improved the antioxidant properties. In addition, the physical properties of the films were improved.

Other Foods

In addition to being widely used in fruits, vegetables and meat, edible films/coatings are also used in other foods such as baked goods, eggs and cheese, etc. In general, baked food was vacuum-packed in plastic to extend its shelf life, but plastic packaging can cause serious environmental problems due to the non-degradability of plastics, edible films/coatings are biodegradable and can be applied to bakery products as an alternative to plastic packaging to extend shelf life [107]. Gregirchak, Stabnikova, & Stabnikov [108] found that bread treated with edible coatings containing lactic acid bacteria had a better inhibitory effect on microorganisms, which could prevent the mold contamination of bread.

The surface of cheese has a high water content and a suitable pH level, which provides a good environment for the growth of microorganisms [109]. Mold growth is an important factor affecting the shelf life of cheese, so edible coatings can be used to inhibit mold growth to extend the shelf life of cheese. Seydim et al. [110] prepared WPI edible films oils with the addition of OEO, garlic oil, nisin and natamycin respectively to coat cheese, and then the cheese was inoculated in the environment of various microorganisms and the inactivation of microorganisms within 15 days was determined. The results showed that nisin /WPI films had the best inactivation effect on listeria monocytogenes, natamycin/WPI film had the best inactivation effect on the Penicillium genus and OEO/WPI had the best inactivation effect on escherichia coli.

With the porous structure, eggshells are easily contaminated by bacteria in the environment [111], escherichia coli and staphylococcus are the two major bacteria to contaminate eggshells. Therefore, protecting the eggshell surface from microbial contamination is the key to extending the shelf life of eggs [112]. Sun et al. [113] added basil essential oil and beeswax to chitosan edible coatings, the results showed that the addition of basil essential oil and beeswax could improve the stability of the coating, and could sterilize the bacteria on the eggshell surface and prolong the shelf life of eggs. The chitosan coating containing 1% basil essential oil and 0.5% beeswax had the best preservation effect on eggs.

Problems and Future Trends

With the improvement of people's lives and the development of food processing technology, there is an increasing demand for food preservation, environment and transport conditions, preservation methods could all influence food shelf-life. As a result, edible films/coatings are receiving a lot of attention from researchers. The films/coatings help to inhibit microbial decay and biochemical damage, maintain the freshness of foods during transport and storage. The main materials [22] used to prepare edible films/coatings include polysaccharides, proteins, lipids, and some additives [34] such as antibacterial agents, antioxidants and plasticizers, etc. Films/coatings can be prepared by extrusion and casting methods, while edible coatings are applied to the food surface by dipping method, spraying method, fluidized bed methods, etc. Once the film has been prepared, the mechanical properties and functional properties of the film need to be evaluated. Edible films/coatings or coatings are not only used on fruits, vegetables and meat products, but also cheese, poultry eggs, bakery products, etc.

The edible films/coatings have great potential for use in the food industry. However, there are still several issues that need to be addressed. Firstly, the process of extracting materials from plants and animal resources is complicated and expensive, and requires professional technology, which will lead to the high preparation cost of edible film, thus limiting its wide commercial application. As to commercialization, the realization ability of special films/coatings material lies in its easy extraction, eco-friendly nature, efficacy at low concentration and having protection potential against biological and chemical damage. Non-toxic, biocompatible and biodegradable microbial biopolymers, such as gellan gum, xanthan gum and gel polysaccharide, have been used as edible food films in the food industry for coating and packaging purposes. The films prepared by these polymers are transparent, have good mechanical and oxygen barrier properties, and need additives to enhance the water barrier properties, thus improving the functional characteristics of edible films [114]. In the next few years, more research should be carried out and improved extraction and separation technologies should be developed to better recover materials from plants, animals and microorganisms.

Secondly, single material cannot fulfill all the requirements of desirable films/coatings and combinations of coatings may help to optimize performance by balancing the benefits and deficiencies of the individual coating ingredient [115]. For example, the combined application of protein and polysaccharide that form high interaction between molecules can improve the barrier performance and delay breathing and aging in many fruits [116]. Therefore, the structure of each biopolymer and their interaction needed to be deeply studied, so as to develop multifunctional and safe edible films to meet the needs of consumers. In addition, all components of the edible films/coatings must have good biocompatibility and interaction to prevent the separation and stratification of the multiphase solution, which is very significant for the mechanical properties of the films/coatings.

Thirdly, applying nanotechnology to edible films/coatings can develop nano-carriers of various bioactive agents to control the release of bioactive agents, protect their stability and prolong food shelf life. However, several studies have shown that food matrix may adversely affect the bioavailability of nano-carriers. Therefore, the relative compatibility and interaction of nano-carriers with certain foods must be considered for the future applicability research of nano-carriers in foods [117]. In addition, the safety of some nanomaterials used in food applications requires appropriate measures and new technologies [118]. Future review and analysis should determine the overall impact of nano-carriers of food bioactive compounds on global environmental sustainability and economy.

In general, biodegradable packaging materials based on natural polymers have attracted more and more attention. To a certain extent, edible films/coatings reduce the economic losses caused by the decay of fruits and vegetables and decrease the risks to human health and the environment [96]. In the coming years, the modification of film materials will be studied in depth to improve the physical properties and overall performance of films/coatings. In-depth research into the interactions of various film substrates and food spoilage mechanisms will provide a theoretical basis for the production of edible films/coatings with better overall performance.

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Data Availability No data was used for the research described in the article.

Declarations

Conflict of Interest I declare that the authors have no competing interests as defined by Springer, or other interests that might be perceived to influence the results and/or discussion reported in this paper.

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