Chapter 8 Coating and Film-Forming Properties



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Abstract Hydrocolloid-based coatings and films, produced from polysaccharides, proteins, and their blends, have emerged as alternatives to synthetic polymers for food and packaging applications, because they are edible, versatile, renewable, and biodegradable. Coatings are formed as a thin layer directly onto food surfaces by dipping, spraying, brushing, fluidized bed, or panning method. By contrast, films are standalone pre-formed materials either placed between food components or sealed into pouches, and they are manufactured by wet- or dry-casting method. Overall, hydrocolloid-based coatings and films possess excellent barrier properties to CO_2 , O_2 , and oil under certain conditions, but moderate water vapor barrier properties. Their formation mechanisms are closely correlated with conformation of biopolymers, their aggregation and crystalline state, as well as their interactions with additives and water. This chapter discusses the existing and potential applications of coatings and films, focusing on the developments and trends of hydrocolloid-based coatings and films for the food industry.

Keywords Hydrocolloids · Coatings · Films · Physicochemical properties · Applications

1 Introduction

Petrochemical-based plastics, such as polyethylene (PE), poly(ethylene terephthalate) (PET), polypropylene (PP), polyvinylchloride (PVC), have dominated the food packaging market for their functionality, lightweight, ease of processing, and low cost (Siracusa et al. 2008). Despite these advantages, increased use of plastic packaging materials has led to serious ecological problems, since they are neither fully recyclable nor biodegradable. While the materials can be incinerated to reclaim

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https://doi.org/10.1007/978-981-16-0320-4_8

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Y. Fang et al. (eds.), *Food Hydrocolloids*,

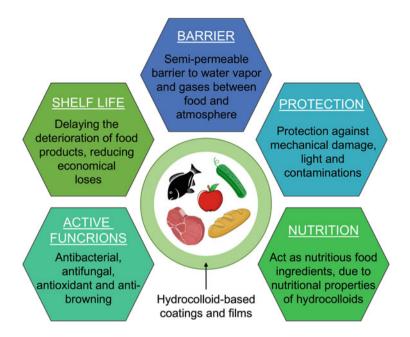


Fig. 8.1 Summary of characteristics and advantages of hydrocolloid-based coatings and films

the energy, this end-of-life approach can produce toxic compounds, including furans and dioxins, such as those produced from burning PVC (Marsh and Bugusu 2007). To address these issues, edible coatings and films have emerged as an alternative to synthetic petroleum-based polymers for food packaging because they are versatile, renewable, and biodegradable (Siew et al. 1999). They have the potential to delay the deterioration of food products and to prolong their shelf life due to their selective barrier properties against oxygen, carbon dioxide, water vapor, and flavor compounds (Giancone et al. 2008). Global edible packaging market is expected to reach USD 1097 million by 2023, from USD 697 million in 2016, growing at a compound average growth rate (CAGR) of 6.81% (Edible packaging-global market outlook from 2017 to 2023 2017).

Polysaccharides, proteins, lipids, and composites derived from these materials, can be used as base materials to prepare edible coatings and films (Gennadios et al. 1996). Hydrocolloids based on polysaccharides and proteins are used extensively for the formation of coatings and films for food preservation, because of their desirable mechanical and gas barrier properties. Besides providing protective function, coatings and films can act as nutritious food ingredients due to the unique nutritional and functional properties of hydrocolloids (Viebke et al. 2014). A scheme illustrating the main characteristics of hydrocolloid-based coatings and films is shown in Fig. 8.1. Generally, there are no fundamental differences in material composition between coatings and films, other than their method of manufacture. Coatings are formed as a thin layer directly onto food surfaces by dipping, spraying, brushing, fluidized bed,

or panning method (Andrade et al. 2012). By contrast, films are standalone pre-formed material either placed between food components or sealed into pouches, and they are manufactured by wet- or dry-casting method (Janjarasskul and Krochta 2010). The performance and functionality of hydrocolloid-based coatings and films are evaluated by their mechanical properties, barrier effects against oxygen (O_2), carbon oxygen (CO_2) and water vapor, and thermal stability. These characteristics are strongly correlated with material compositions, manufacture methods, and the end-use conditions (e.g., relative humidity, temperature, and pH) (Rojas-Graü et al. 2009).

This chapter provides an overview on different categories of hydrocolloids for coating and film formation. Methods of preparation, forming mechanisms, and the physicochemical properties for coatings and films are also discussed. Finally, recent developments and trends for packaging applications involving hydrocolloids are summarized.

2 Components of Coatings and Films

Hydrocolloid-based coatings and films are produced from polysaccharides, proteins, their blends, and/or food-grade additives. Their functional, organoleptic, nutritional, and mechanical properties are modified by addition of food-grade additives, including plasticizers, antimicrobials, antioxidants, anti-browning and crosslinking agents, nanofillers, colorants, and flavors (Otoni et al. 2017). The main components for formulation of hydrocolloid-based coatings and films are summarized in Fig. 8.2.

2.1 Polysaccharides

Polysaccharides are nontoxic and naturally occurring biopolymers. The polysaccharide film-forming materials include starch and starch derivatives, cellulose derivatives, alginate, carrageenan, chitosan, various plant gums (pectin, konjac, locust bean gum, and guar gum) and microbial gums (pullulan, xanthan) (Cazón et al. 2017). Although polysaccharide-based coatings and films have superlative barrier properties to CO₂, O₂, and oil under certain conditions, and high strength and structural integrity, they tend to present a poor barrier to water vapor due to their hydrophilic properties (Yang and Paulson 2000).

2.1.1 Starch and Starch Derivatives

Starch, an agricultural biopolymer found in a variety of plants, is a mixture of amylose and amylopectin whose content varies depending on its botanic origin (LeCorre et al. 2011). Amylose, a nearly linear biopolymer of α -1,4 anhydroglucose

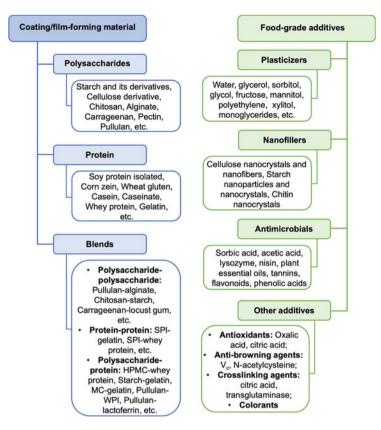


Fig. 8.2 Main components for formulation of hydrocolloid-based coatings and films

units, is known to form a coherent and relatively strong films (Campos et al. 2011). In contrast, amylopectin is a highly branched biopolymer of short α -1,4 chains linked by α -1,6 glucosidic branching points occurring every 25–30 glucose units (Durrani and Donald 1995). Its branched structure leads to form brittle and non-continuous films (De Azeredo et al. 2014). In comparison with native starch, modified starches, like acetylated starch, hydroxypropyl starch, and oxidized-starch, have been reported to form stronger and more flexible films (López et al. 2008, 2010; Hu et al. 2009).

2.1.2 Cellulose Derivatives

Cellulose, the main component of plant fibers, is essentially a linear high-molecular weight biopolymer of D-glucose units linked through β -1,4 glycosidic bonds. The close packing of cellulose chains makes it highly crystalline, fibrous, and insoluble in water (Wang et al. 2016). Water-soluble cellulose derivatives, such as methylcellulose (MC), hydroxypropyl methylcellulose (HPMC), carboxymethylcellulose

(CMC), and hydroxypropyl cellulose (HPC), possess good film-forming characteristics (Dhall 2013). Among them, MC films show the lowest hydrophilic properties, whereas the water vapor permeability of HPMC and CMC films is relatively high (Sánchez-González et al. 2009; Kester and Fennema 1989). In addition, the substitution type and degree in cellulose derivatives are critical factors determining the performance of cellulose-based films (Espinoza-Herrera et al. 2011).

2.1.3 Chitosan

Chitosan is a functional biopolymer derived from chitin by deacetylation in alkaline media. It consists of randomly distributed β -(1,4)-2-acetamido-D-glucose and β -(1,4)-2-amino-D-glucose units, with the latter usually exceeding 60% (Kim et al. 2006). Chitosan has a wide spectrum of activity and high killing rate against Grampositive and Gram-negative bacteria (Chung and Chen 2008). The antimicrobial activity and film-forming ability of chitosan are correlated to its degree of acetylation or deacetylation, and molecular weight (Hosseinnejad and Jafari 2016). Owing to its outstanding characteristics, chitosan could be potentially utilized as the antimicrobial packaging materials to improve food quality and shelf life.

2.1.4 Polysaccharides Extracted from Seaweed

Alginate, a linear polysaccharide extracted from brown seaweed, is composed of variable proportions of β -D-mannuronic acid (M block) and α -L-guluronic acid (G block) linked by 1,4 glycosidic bonds. The block copolymer consists of homopolymeric regions of M- and G-blocks, separated by regions that contain M and G units (Fu et al. 2011). The proportion and distribution of these blocks determine the physicochemical properties of the biopolymer (Lacroix and Le Tien 2005). Alginate dissolves readily in water to form homogeneous film-forming solutions, which upon drying can yield coherent, and transparent films that have a wide range of food applications (Xiao et al. 2012).

Carrageenan is an anionic linear polysaccharide, extracted from edible red seaweeds of *Rhodophycea* class. It is formed by alternate units of D-galactose and 3,6-anhydrogalactose linked by α -1,3 and β -1,4 glycosidic linkage (Cosenza et al. 2014). There are three types (κ , 1, and λ) of carrageenan with varying number and position of sulfate groups on the galactose dimer (Liu et al. 2015). In comparison with 1-carrageenan films, κ -carrageenan films showed the higher moisture barrier and mechanical properties, except for its flexibility (Paula et al. 2015).

2.1.5 Pectin

Pectin consists of linear homo-galacturonan (α -1,4-galacturonic acids) chains interspersed with branched rhamnogalacturonan (α -1,4-galacturonic acid to α -1,2-rhamnose) chains (Jolie et al. 2010). According to its degree of esterification (DE), pectin can be classified as high-methoxyl pectin (HMP, DE > 50%) and low-methoxyl pectin (LMP, DE < 50%) (Espitia et al. 2014). The mechanical, water barrier properties and thermal stability of HMP films are better than that of LMP films (Lorevice et al. 2016).

2.1.6 Pullulan

Pullulan is an extracellular and water-soluble microbial polysaccharide produced by *Aureobasidium pullulans*. The linear polymer mainly consists of maltotriose units interconnected to each other by α -(1,6) glycosidic bonds, which are responsible for the flexible conformation and the ensued amorphous character of this polysaccharide in the solid state (Sutherland 1998). This unique linkage pattern endows pullulan with distinctive physical properties to form film that is strong, transparent, and with low permeability to oil and oxygen (Xiao et al. 2012, 2015).

2.2 Proteins

Proteins used for film-forming materials can be categorized into two groups based on their origin of sources: plant-derived proteins, such as corn zein, soy protein, and wheat gluten, or animal-derived proteins like casein, whey protein, gelatin, and collagen proteins (Han 2014). Depending on amino acid composition and sequence, the structure of protein can be random coil, fibrous, or globular. For globular proteins (i.e., soy protein, wheat gluten), they must be denatured by heat, acid, and/or solvent to shape extra extended structures that are required for film formation (Dhall 2013). Overall, protein-based coatings and films display considerably lower O_2 and CO_2 permeability and CO_2/O_2 permeability ratio, and moderate mechanical and water vapor barrier properties (Song and Zheng 2014).

2.2.1 Corn Zein

Corn zein, a prolamin protein, has a molecular weight ranging from 18 to 45 kDa. As a relatively hydrophobic protein, the hydrophobicity of zein is related to its high content of non-polar amino acids residues including leucine, alanine, and proline (Shukla and Cheryan 2001). Corn zein dissolves in aqueous ethanol solution to form the glossy, greaseproof, and brittle films through the hydrophobic, hydrogen, and limited disulfide (SS) bonds between zein chains (Ghanbarzadeh et al. 2007).

2.2.2 Wheat Gluten

Wheat gluten, an ethanol-soluble protein in wheat flour, is composed of gliadin and glutenin. Gliadin is monomeric protein with molecular weight of 28–55 kDa, while glutenin is aggregated protein linked by interchain SS bonds with molecular weight of about 500 to 10,000 kDa (Wieser 2007). Glutenin films presented higher mechanical strength and lower water vapor permeability than gliadin films (Hernández-Muñoz et al. 2003). Moreover, the purity of wheat gluten has positive effect on the appearance and mechanical attributes of wheat gluten films (Gennadios et al. 1993).

2.2.3 Soy Protein

Soy protein is comprised of two major components, 7S (β -conglycinin) and 11S (glycinin), representing 37% and 31% of soy protein, respectively. 7S is rich in asparagine, glutamine, leucine, and arginine residues with a molecular weight of 180 kDa. 11S has a molecular weight of 320–360 kDa and contains 20 intramolecular SS bonds (Kumar et al. 2002). Films made from 11S fraction are smooth and opaque, whereas 7S films exhibit transparent and creased appearance (Kunte et al. 1997). At low relative humidity (RH), O₂ permeability of soy protein isolate (SPI) films was lower than that of films based on low-density polyethylene (LDPE), methylcellulose, starch and pectin, respectively (Song et al. 2011a).

2.2.4 Casein and Caseinate

Casein mainly consists of five fractions including α_{s1} , α_{s2} , β , κ , and δ -casein, and their sizes vary from 11.5 to 25 kDa. Among them, β -casein is the most interesting one, as it produces films with lower permeability to water vapor than other milk protein (Mauer et al. 2000). Caseinate is a mixture of casein monomers and small aggregates formed after removing of colloidal calcium phosphate from casein micelles. Compared to casein, caseinate, particularly for sodium caseinate, is more soluble and has better film-forming capacity. Films produced from sodium caseinate possess excellent barriers to O₂, CO₂, and aromas, and thermal resistance (Khwaldia et al. 2004a).

2.2.5 Whey Protein

Whey protein includes β -lactoglobulin, α -lactalbumin, bovine serum albumin, immunoglobulins, lactoferrin, and proteose-peptones (Mulvihill and Ennis 2003). Films prepared from whey protein isolates (WPI) exhibited promising mechanical features, as well as moderate moisture permeability and good oxygen barrier properties, compared to the synthetic polymer films, e.g., low-density polyethylene (LDPE), high density polyethylene (HDPE), PVDC, cellophane, and polyester (Khwaldia et al. 2004b).

2.2.6 Gelatin

Gelatin is an animal protein obtained by hydrolysis of collagen. It is a combination of many fractions varying in size, including the whole α -chain of tropocollagen molecule (a trimer of around 330 kDa that aggregates to form the larger collagen structures) and hydrolytic fragments of parts of the α -chains (Boran and Regenstein 2010). Gelatin films display effective barriers against O₂ and aromas at low or intermediate RH, but weak water resistance due to its hydrophilic nature. Furthermore, their mechanical properties are closely related to the renaturation level of gelatin (Bigi et al. 2004).

2.3 Food-Grade Additives

2.3.1 Plasticizers

Plasticizers are low molecular weight compounds with non-volatile compounds. Their primary role is to enhance the flexibility and processability of hydrocolloidbased coatings and films. However, their barrier properties are impaired as result of the increased free volume and molecular mobility after plasticizers addition (Sothornvit and Krochta 2005; Vieira et al. 2011). Food-grade plasticizers mainly include glycerol, sorbitol, polyethylene glycol, sucrose, glucose, fructose, mannitol, xylitol, fatty acids, and monoglycerides (Vieira et al. 2011).

2.3.2 Polysaccharide Nanofillers

Nanofillers (at least one dimension smaller than 100 nm) provide reinforcement effects due to their high aspect ratio and surface-to-volume ratios (Crosby and Lee 2007). Considering the application and safety for hydrocolloid-based coatings and films in food packaging, the polysaccharide nanofillers, e.g., cellulose nanoparticles, cellulose nanocrystals, starch nanoparticles, starch nanocrystals, chitin nanowhiskers, and chitin nanofibers, have been used as excellent candidates for improvement of their mechanical, barrier, and thermal properties (Otoni et al. 2017).

2.3.3 Antimicrobial Additives

Incorporation of antimicrobial compounds into packaging materials provides inhibitory effects against spoilage and pathogenic bacteria by maintaining active compounds on food surface (Gennadios et al. 1997). There are several categories of antimicrobial compounds that have been employed in hydrocolloid-based coatings and films, including organic acids (sorbic and its potassium salt, acetic acid, and malic acid), polypeptides and bacteriocins (lysozyme and nisin), plant essential oils (cinnamon, oregano, rosemary, and lemongrass), and polyphenols (flavonoids and phenolic derivatives) (Franssen and Krochta 2003).

3 Preparation Methods

3.1 Preparation of Hydrocolloid-Based Coatings

3.1.1 Spray Coating

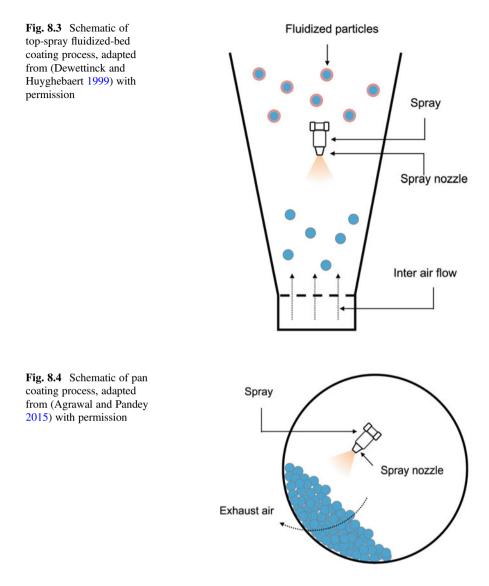
Spray coating is a commonly used technique for food coatings, especially for fruits and vegetables. In this process, food products are placed on a rotating platform, then the coating-forming solution forms droplets and distributes them over the food surface by means of a set of spraying nozzles (Debeaufort and Voilley 2009). The main advantages of this technique offer uniform coating, thickness control, and the possibility of multilayer applications, such as using alternating sodium alginate and chitosan solutions (Ustunol 2009).

3.1.2 Dip Coating

Dip coating involves submerging food products into a vat containing coating solution. After dipping the products and draining away excess coating, it is dried either at room temperature or with the aid of a dryer (Andrade et al. 2012). The advantage of this method is to obtain good uniformity around the irregularly-shaped and rough food surface. Several problems may occur by using this method, such as coating dilution, build-up of trash or dirt, and microorganism growth in the dipping tank (Andrade et al. 2012).

3.1.3 Fluidized-Bed Coating

Fluidized beds are categorized by three different configurations: top spray, bottom spray, and rotating-fluidized bed. The conventional top-spray method has a greater possibility of success in the food industry compared to other methods (Andrade et al. 2012). As presented in Fig. 8.3, the coating solution is sprayed through a set of nozzles onto the surface of fluidized particles to form a shell-type structure. Its application focuses on the functional ingredients and food additives, i.e., leavening agents, enzymes, vitamins, minerals, and spices (Chen et al. 2009).



3.1.4 Pan Coating

The schematic of pan coating process is displayed in Fig. 8.4. As shown, the coating solution is sprayed into a rotating bowl (referred to as pan), and the food particles are tumbled within the pan to distribute the coating solution over their surface. Forced air, either ambient or elevated temperature, is utilized to dry the coating (Agrawal and Pandey 2015). Pan coating is mainly used for the confectionery and chocolate industries or particularly small food items like nuts and raisins (Andrade et al. 2012).

3.2 Preparation of Hydrocolloid-Based Films

3.2.1 Wet Method

The wet method, also known as solvent casting, can be sub-classified to bench casting and continuous casting, respectively. The bench casting is commonly utilized to fabricate films at laboratory scale as it is simple and cost effective. In this method, the film-forming solution is deposited over a rimmed plate, and then followed by drying to produce a cohesive and free-standing film.

Continuous casting is more suitable for industrial applications, because it requires less space and labor. As shown in Fig. 8.5, film-forming solution is uniformly spread on a continuous steel belt that passes through a drying chamber. The dried film is then stripped from the steel belt and wound into film roller. The advantage of this method is optimizing uniformity, heat transfer, and drying efficiency, while avoiding expense of a separate substrate (Rossman 2009).

3.2.2 Dry Method

Dry method, i.e., compression molding and extrusion processing, is based on the thermoplastic properties of polysaccharides and proteins. In the presence of plasticizers, at low moisture levels and high temperatures and with pressure, biopolymers acquire a viscoelastic behavior that allows them to be shaped for the production of films (Gómez-Estaca et al. 2016). In general, compression molding is studied at laboratory scale as a precursor to extrusion with the aim of determining the suitable processing conditions (Hernandez-Izquierdo and Krochta 2008).

Extrusion processing is a highly efficient manufacturing method with commercial potential for large-scale production of biopolymer films (Fishman et al. 2000). The

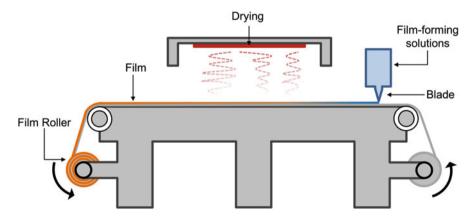


Fig. 8.5 Schematic of continuous casting technique to prepare hydrocolloid-based films, adapted from (Borges et al. 2015) with permission

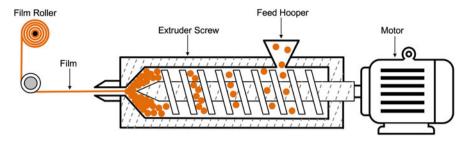


Fig. 8.6 The configuration of one-screw extruder, adapted from (Borges et al. 2015) with permission

configuration of one-screw extruder is presented in Fig. 8.6. The extruder basically consists of an endless screw inside a barrel with a double casing that permits control of temperature. The biopolymer is fed from a hopper and pushed by the screw towards a die (Nur Hanani et al. 2012). To date, dry method has been successfully used in preparation of starch, alginate, wheat gluten, soy protein, and whey protein films (Mendes et al. 2016; Hernandez-Izquierdo and Krochta 2008; Azevedo et al. 2017; Ciannamea et al. 2014).

4 Microstructural and Physicochemical Characterization

The microstructural characteristics (such as chemical, crystalline structure, and morphology) of hydrocolloid-based coatings and films are closely correlated with their packaging performance (e.g., mechanical, barrier, and thermal properties).

4.1 Structural Analysis

Microscopy and spectroscopic techniques have been utilized to study the architecture and structure of hydrocolloid-based films at micro and nanometric scales. Ultrastructural and internal structure in films have been characterized by confocal laser scanning microscopy (CSLM), while scanning electron microscopy (SEM) and atomic force microscopy (AFM) are more used to study their surface and crosssection morphology (Arzate-Vázquez et al. 2012; Andreuccetti et al. 2009). Fourier transform infrared spectroscopy (FTIR) analyzes the possible functional chemical groups, conformational transitions, and molecular interactions (Yadav et al. 2014). Nuclear magnetic resonance (NMR) spectroscopy provides information about the chemical and physical properties of atoms or their related molecules, as well as reaction state, dynamics, structure and chemical environment (Karbowiak et al. 2008). For instance, for hsian-tsao gum (HG)-casein films, the hydrogen bonding interactions and Maillard reactions between HG and casein were revealed by FTIR data. Meanwhile, NMR analysis indicated that HG addition significantly changed the mobility of water molecule in casein films (Yang et al. 2015). Other complementary techniques are also utilized for structural analysis of hydrocolloid-based coatings and films, such as X-ray diffraction (XRD) to identify the information about crystalline/amorphous structures, and small-angle X-ray scattering (SAXS) to monitor crystalline and aggregate structures of membrane materials (Bodnár et al. 2007).

4.2 Mechanical Properties

Favorable mechanical properties are essential for packaging materials to perform their protective functions efficiently. Mechanical properties of selected hydrocolloid-based films are listed in Table 8.1. A standard method, ASTM-D882–91, originally developed to evaluate mechanical properties such as tensile strength (TS), elongation at break (EAB), elastic modulus (EM), and toughness of commercial plastic, is also applied to hydrocolloid-based films (ASTM-D882-91 1991). As shown in Fig. 8.7, the mechanical parameters are calculated by determining the relationship between stress and strain, when film is stretched at a set rate (distance/time). EM, a measure of intrinsic film stiffness, is the slope of the linear range of the stress–strain curve (Mauer et al. 2000). Toughness refers to the ability of a material to absorb energy during deformation up to fracture, determined as the area under the stress–strain curves (Fig. 8.7b). TS is the maximum strength measuring the resistance of the film, whereas the percentage of EAB is a measure of the stretching capacity of flexibility of the film prior to breaking. They are calculated by using Eqs. 8.1 and 8.2:

$$TS = F/A \tag{8.1}$$

where TS is the tensile strength (MPa), F is the force (N) at maximum load, and A is the initial cross-sectional area (m²) of the film specimen.

$$EAB = 100 \times (l - l_1)/l_1 \tag{8.2}$$

where EAB is the elongation at break (%), l_1 is the initial length, and l is the length of the film at breaking point.

4.3 Barrier Properties

The basic function of packaging materials is to control mass transfer between food and the ambient atmosphere. Water vapor in environment transferring to packaged food results in problematic microbial growth, and undesirable textural changes. Oxygen can cause deterioration of food due to oxidation of lipids and other

	Mechan	Mechanical properties	perties	WVP		Gas permeability	bility		
Composites ^a	TS (MPa)	EAB (%)	EM (MPa)	Test conditions	$(g.m.Pa^{-1}.h^{-1}.m^{-2})$	Test conditions	$O_2P (cm^3.mm. m^{-2}.d^{-1}.Pa^{-1})$	$CO_2P (cm^3.mm. m^{-2}.d^{-1}.Pa^{-1})$	Reference
HMw chitosan	61.82	4.59	1	38 °C, 90% RH	1.34×10^{-10}	23 °C, 50% RH	6.65	1	(Leceta et al. 2013)
LMw chitosan	55.83	4.58	1	38 °C, 90% RH	1.53×10^{-10}	23 °C, 50% RH	7.70	1	(Leceta et al. 2013)
Pullulan	73.89	1.67	4938	23 °C, 100% RH	7.05×10^{-6}	23 °C, 23% RH	0.48	1	(Xiao et al. 2012, 2015)
Alginate	74.04	4.60	3047	23 °C, 100% RH	$25.75 imes 10^{-6}$	23 °C, 23% RH	8.94	1	(Xiao et al. 2012, 2015)
Sweet potato starch- chitosan	33.8	2.02	I	25 °C, 75% RH	7.09×10^{-6}	23 °C, 50% RH	4.09	1	(Shen et al. 2010)
WPC-GLY (40% w/w)	0.72	50	78	20 °C, 50% RH	4.50×10^{-6}	20 °C, 50% RH	0.2	1.02	(Ramos et al. 2013)
WPI-GLY (40% w/w)	0.9	55	95	20 °C, 50% RH	3.43×10^{-6}	20 °C, 50% RH	0.41	1.58	(Ramos et al. 2013)
Vetch seed protein	1.59	32.08	78.14	1	1	25 °C, 55% RH	20.16	24.84	(Porta et al. 2015)
Papaya puree-gelatin (8:1)	6.7	18.09	1	38 °C, 90% RH	6.62×10^{-5}	23 °C, 50% RH	8.24	10.25	(Tulamandi et al. 2016)
Papaya puree-defatted soy protein (8:4)	5.13	28.11	I	38 °C, 90% RH	7.52×10^{-5}	23 °C, 50% RH	8.92	10.89	(Tulamandi et al. 2016)
WPI-okra polysaccha- ride (1:1)	2.3	31.4	68.2	23 °C, 100% RH	3.23×10^{-5}	20 °C, 50% RH	5.67	1	(Prommakool et al. 2011)
WPI-LBG (0.025% w/w)	2.66	11.4	I	20 °C, 100% RH	4.97×10^{-7}	20 °C, 50% RH	0.78	4.71	(Silva et al. 2016)

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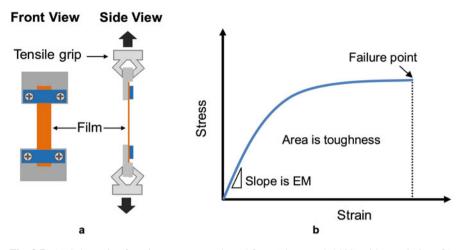


Fig. 8.7 (a) Schematic of tension test setup, adapted from (Pham et al. 2008) with permission, (b) mechanical properties determined from the typical stress-strain curve

oxygen-sensitive components. Thus, water vapor and gas permeability is a vital property for selecting or tailoring the hydrocolloid-based films.

4.3.1 Water Vapor Permeability (WVP)

Table 8.1 shows WVP values of selected hydrocolloid-based films. These data are obtained gravimetrically following the ASTM Standard Test Method E96, known as the "cup method" (ASTM-E96-92 1990). According to this method, a cup with an open mouth is filled with distilled water or desiccant. The film is sealed on the open mouth of the cup, the assembly is weighed, and placed under controlled temperature and RH conditions (Cazón et al. 2017). WVP is calculated according to the combined Fick–Henry laws for gas diffusion through films (Eq. 8.3).

$$WVP = \frac{\Delta w}{\Delta t \times A} \times \frac{L}{\Delta p}$$
(8.3)

where $\Delta w/\Delta t$ is the rate of water gain (g/h), A is the exposed area of the film (m²), L is the mean thickness of film specimens (m), and Δp is the difference in partial water vapor pressure between the two sides of film specimens.

4.3.2 Gas Permeability

Oxygen permeability (O_2P) and carbon dioxide permeability (CO_2P) are evaluated on the basis of the ASTM D 3985–02 method (ASTM-D3985-02 2002). The films are sealed between two chambers with each having two channels to the exterior. In the lower chamber, O_2 or CO_2 is supplied at a controlled flow rate to maintain the pressure constant in that compartment. The other chamber is purged by a stream of nitrogen, also at a controlled flow. In the case of O_2P measurement, the nitrogen flow leaving this chamber is connected to an O_2 sensor installed on-line which measures the O_2 concentration. For CO_2P measurement, the nitrogen flow leaving this chamber is collected in a syringe for CO_2 quantification by a gas chromatograph (Cerqueira et al. 2009). The O_2P and CO_2P of selected hydrocolloid-based films are listed in Table 8.1.

4.4 Thermal Properties

One key factor that influences the processing and operating temperatures of hydrocolloid-based coatings and films is their thermal properties. The properties are investigated by differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), and dynamic mechanical analysis (DMTA). DSC technique is used to determine the glass transition temperature (T_g), melting temperature (T_m), crystallization temperature, heat capacity difference at T_g of hydrocolloid-based coatings and films (Cheng 2002). TGA is widely employed to examine their decomposition temperature, weight loss, and activation energy of decomposition (Cheng 2002). Furthermore, the structural and viscoelastic properties of films are investigated by DMTA. Dynamic modulus, dynamic loss modulus, temperature of main chain relaxation, and temperature of local mode relaxation are measured as functions of temperature and frequency by forced oscillation method (Brown and Gallagher 2011).

5 Film-Forming Mechanism

Understanding the film-forming mechanism is important to predict material properties of hydrocolloid-based films, which is essential for the optimization of drying and processing condition. As previously mentioned, both processing methods (wet and dry) have been widely used to prepare the films. The wet method requires solubilizing the hydrocolloids in a solvent, spreading the solution onto a flat surface, and then followed by drying to produce a film. The film-forming mechanism involves conformational change of the biopolymer, as well as solvent-biopolymer and biopolymer-biopolymer interactions that continue to evolve as the solvent evaporates under different drying conditions (Watanabe et al. 2006; Xiao et al. 2014b). However, a number of polysaccharides and proteins have capacity to form gel during film-forming process, and their film-forming mechanism is related to the gelation mechanism. Although a few researchers proposed that the transition from wet gel (biopolymer-in-water) to dry film (water-in-biopolymer) is a critical stage during film-forming process, the complete transition mechanism after gelation have not yet been fully explained (Szabó et al. 2012).

By contrast, dry method involves heating and mixing biopolymers and plasticizers by extrusion and/or compression molding techniques. Over the course of extrusion, biopolymer chains denature, dissociate, unravel and align, and then recombine, crosslink, and aggregate via specific linkages with heat and pressure, which result in film formation through complete restructuring of biopolymer molecules. Thus, the film-forming mechanism is correlated with conformation changes of biopolymers, their aggregation and crystalline state, as well as the interactions among biopolymer, plasticizer, and water.

5.1 Polysaccharide-Based Films

Polysaccharides (with the exception of glycogen, etc.) are long-chain biopolymers formed from mono- or disaccharide repeating units joined together by glycosidic bonds. Owing to the presence of a large number of hydroxyl and other polar groups in their structure, hydrogen bonds and/or electrostatic interactions have a crucial function in film formation (Han 2014). Polysaccharide films are fabricated by disrupting interactions among polysaccharide segments and forming new intermolecular hydrophilic interactions and hydrogen bonding (Rhim and Ng 2007).

5.1.1 Formation Mechanism of Solvent Casting Films

For starch films, their formation mechanism depends on the starch concentration and amylose content. At relatively high concentration, aggregation and packing of swollen granules dominated the film formation, whereas both coil-to-helix transition and aggregation of double helices were operative during the film formation from dilute starch solutions (Liu 2005). Xiao et al. (2014a, b) elaborated the formation mechanism of pullulan and alginate films by monitoring the conformational change of polysaccharides, water-polysaccharide, and polysaccharide-polysaccharide interactions during drying. As pullulan drying process progressed, the oxygen atoms at the C₅ and C₆ carbons of the D-glucopyranose ring might preferentially form hydrogen bond with water or pullulan molecules, resulting in more-ordered structure with increased interchain interactions in pullulan films. Moreover, the less-ordered structure domain of the pullulan was first affected during drying, followed by pullulan skeleton segments. Finally, conformational changes in pullulan chains occurred as the drying process completion (Xiao et al. 2014b). In the course of the formation of alginate film, the oxygen atoms at the C_2 and C_3 carbons of the pyranose ring preferentially formed hydrogen bond with water or alginate molecules, while the skeletal vibrations of pyranose ring (e.g., C-C and C-O-C groups) were less perturbed than the stretching vibrations of COO⁻ group and O-H bending vibration of alginate with drying (Xiao et al. 2014a). The film-forming mechanism of *Flammulina velutipes* polysaccharide might be associated with the intermolecular and intramolecular hydrogen bonds between polysaccharide chains and the formation of β -glycosidic bonds upon drying (Du et al. 2016). Li et al. (2019) proved that the electrostatic interactions and hydrogen bonds are crucial in fabricating the multilayer films based on chitosan and alginate by layer-by-layer (LbL) technique. Strong intermolecular interactions occurred among the amino, carboxyl, and hydroxyl groups of the chitosan and alginate.

5.1.2 Formation Mechanism of Extruded and Compression-Molded Films

Pushpadass et al. (2009) reported that glycerol and/or water destroyed the crystallinity of native starch, then the starch fragmentation converted into thermoplastic starch with heat and shear. During extrusion process, the inter- and intra-hydrogen bonds of starch would be unraveled when the glycerol was added into starch, and the new hydrogen bonds between starch and glycerol were formed simultaneously (Pushpadass et al. 2009). Afterwards, the starch recrystallization induction process among the helical amylose molecule occurred during cooling, which led to the Vh-type crystalline arrangement (Azevedo et al. 2017). According to Gao et al. (2017), neat alginate granules were largely de-structured by glycerol and water, and glycerol increased the mobility of alginate chains while promoting the crystallization of alginate chains with structural reorganization during compression molding.

5.2 Protein-Based Films

The main formation mechanism of protein films involves denaturation of the protein initiated by heat, solvent, or change in pH, followed by association of extended peptide chains through new intermolecular interactions, such as covalent (SS bond or crosslinking) and electrostatic, hydrophobic, or ionic interactions between protein chains (Janjarasskul and Krochta 2010).

5.2.1 Formation Mechanism of Solvent Casting Films

The formation of intact and water-insoluble WPI films was realized by heat denaturation of aqueous protein solution (Pérez-Gago and Krochta 2002). Heat denaturation unfolded whey protein and promoted the exposure of SH and hydrophobic groups. The unfolded protein might then undergo intermolecular interactions (hydrogen bonds, hydrophobic, covalent and electrostatic interactions). It is noteworthy that the cohesion of WPI films relied principally on the intermolecular SS bonds via sulphydryl/disulphide (SH/SS) exchange reactions (Guckian et al. 2006). On the other hand, WPI had the ability to form water-soluble films without heat denaturation. Since most of the hydrophobic and SH groups are buried in the interior of WPI molecule, their film-forming mechanism involves the intermolecular hydrogen bonding between protein molecules, rather than the hydrophobic and covalent interactions (Guckian et al. 2006; Pérez-Gago et al. 1999). Ciannamea et al. (2014) also proved that hydrogen bonds and hydrophobic interactions played a more important role in the formation of soy protein films. During the film formation of 11S, along with disappearance of its α -helices and disordered structures, the intermolecular hydrogen bonds between β -sheet segments predominated the aggregation of 11S (Robert et al. 2001; Subirade et al. 1998). Similar to the 11S films, the high density of intermolecular hydrogen-bonded β -sheets were conducive to the formation of gliadin network during drying (Mangavel et al. 2001). According to Pankaj et al. (2014), the film-forming mechanism of caseinate was attributed to their random coil structure which allowed them to form extensive intermolecular hydrogen, electrostatic, and hydrophobic bonds, resulting in increased interchain cohesion.

In comparison, the formation mechanism of gelatin films is related to the temperature during drying due to thermo-reversible gelation behavior of gelatin. When the gelatin films were prepared below the helix-coil transition temperature, partial renaturation of collagen in gelatin took place, which resulted in the formation of a collagen-like triple-helix structure. Moreover, the partial renaturation only took place during the advanced stage of drying (Ghoshal et al. 2014). On the contrary, a helix structure was rarely formed in gelatin films when they were dried above the helix-coil transition temperature.

5.2.2 Formation Mechanism of Extruded and Compression-Molded Films

For compression-molded soybean protein films, the high temperature promoted the crosslinking between soybean proteins through intermolecular SS bonds, either from free sulfhydryl (SH) groups or through SH/SS exchange reactions, which predominated the formation of film matrix (Ciannamea et al. 2014). During extrusion process, the aggregation and reorganization of wheat gluten molecules were principally related to the formation of intermolecular SS crosslinking bonds via oxidation of SH groups and SH/SS exchange reactions between glutenin and gliadin (Lagrain et al. 2010). The formation schematic of intermolecular SS bonds between glutenin and gliadin during heat processing is illustrated in Fig. 8.8.

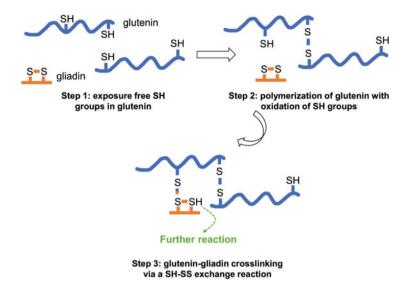


Fig. 8.8 Schematic of the formation of intermolecular SS crosslinking bonds between glutenin and gliadin during heat processing. Adopted and modified from (Lagrain et al. 2010) with permission

6 Applications and Recent Developments

Oxidation, microbial spoilage, and metabolism are the main causes of deterioration of food products. Thus, the primary function of packaging materials based on hydrocolloids is to maintain the quality and safety of food products during storage and conveyance. Normally, fruits and vegetables have short shelf life due to its perishable nature. Hydrocolloid-based coatings and films may act as a semipermeable barrier to selectively control the exchange of CO₂, O₂, and ethylene, resulting in the reduction in ethylene levels, ripening, respiration rate, and water loss on fruits and vegetables (Valencia-Chamorro et al. 2011). Several studies shown in Table 8.2. have demonstrated the ability of hydrocolloid-based coatings and films carrying bioactive compounds to retard browning reactions and microbial growth in fruits and vegetables, especially the minimally processed (MP) fruits and vegetables. Ramos-García et al. (2012) reported that lime essential oil incorporated into chitosanbeeswax blend coatings on tomato showed strong inhibitory effect against *Rhizopus* stolonifer and Escherichia coli DH_{5α} during storage at 12 and 23 °C. Sarengaowa et al. (2018) coated the fresh-cut "Red Fuji" apples with alginate coatings containing thyme oil, cinnamon oil, and/or oregano oil, and observed that reduction of total coliform, yeast and mold counts in comparison with control and alginate-coated samples. Meanwhile, the respiration rate, weight loss, firmness, and browning reactions in fresh-cut apples stored at 4 °C were significantly decreased.

Recently, the development of multilayer and nanomultilayer coatings based on hydrocolloids, formed by LbL deposition technique, gained much attention for the preservation of fruits and vegetables. For instance, the multilayer coatings based on gelatin and chitosan predominantly enhanced physiological quality and reduced the

Coating/film composites	Form	Food products	Main benefits	References
-		Fruits and vegetables		
Pectin-pullulan- chitosan with sodium benzoate and potassium sorbate	Coatings	Strawberry (Fragaria ananassa)	Reduced weight loss, fruit softening and microbial growth (total aerobic counts, molds, and yeasts), delayed alteration of color and total soluble solids content	(Treviño- Garza et al. 2015)
Alginate with carvacrol and methyl cinnamate	Coatings	Strawberry	Inhibited the Escherichia coli O157: H7 and Botrytis cinereal	(Peretto et al. 2014)
Alginate-pectin	Coatings	Blueberry	Improved the firmness, significantly reduced growth kinetics of yeasts and mesophilic aerobic bacteria	(Mannozzi et al. 2017)
Gum arabic- <i>Aloe vera</i> - chitosan com- bined with thyme oil	Coatings	Avocado (Persea ameri- cana Mill.)	Reduced the anthrac- nose incidence during the postharvest supply chain, inhibited myce- lial growth of <i>Colletotrichum</i> <i>gloeosporioides</i>	(Bill et al. 2014)
Chitosan- carrageenan	Coatings	Longan (Dimocarpus longan)	Reduced weight loss, respiration rate and color changes	(Lin et al. 2018)
Chitosan-cas- sava starch with essential oil extract from <i>Lippia gracilis</i>	Coatings	Guavas (Psidium guajava L.)	Reduced total aerobic mesophilic bacteria, mold and yeast counts, exhibition lower titrat- able acidity value	(de Aquino et al. 2015)
Pea starch-guar gum with shellac and oleic acid	Coatings	'Valencia' oranges	Reduced fruit respira- tion rate, ethylene pro- duction, weight and firmness loss, and peel pitting	(Saberi et al. 2018)
CMC-chitosan	Bilayer coatings	Citrus fruit	Increased fruit firm- ness, and enhanced fruit gloss	(Arnon et al. 2014)
Gum arabic with cinnamon oil	Coatings	Banana and papaya	Delayed ripening, weight loss, fruit firm- ness, and titratable acidity, fungicidal effects against	(Maqbool et al. 2011)

 Table 8.2
 Recent applications of hydrocolloid-based coatings and films in fruits and vegetables

Coating/film composites	Form	Food products	Main benefits	References
			Colletotrichum musae and Colletotrichum gloeosporioides	
HPMC-beeswax	Coatings	Plums (Cv. Angeleno)	Reduced water loss, flesh softening and internal breakdown	(Navarro- Tarazaga et al. 2011)
HPMC with oregano essen- tial oil	Coatings	'Formosa' plum	Reduced the respiration rate, ethylene produc- tion, total weight loss	(Choi et al. 2016)
Chitosan with Artemisia annua oil	Coatings	Cherry tomato	Decreased Escherichia coli O157:H7	(Cui et al. 2017)
Chitosan-bees- wax with lime essential oil	Coatings	Tomato	No growth of <i>Rhizopus</i> stolonifer and Escherichia coli $DH_{5\alpha}$	(Ramos- García et al. 2012)
Pectin-chitosan	Nanomultilayer coatings	'Tommy Atkins' mangoes	Presented a lower mass loss, lower total soluble solids and higher titrat- able acidity	(Medeiros et al. 2012a, b)
Starch-gelatin	Coatings	Refrigerated red crimson grapes	Enhanced appearance and decreased weight loss	(Fakhouri et al. 2015)
<i>Aloe vera</i> -gum tragacanth	Coatings	Button mushroom	Reduced weight loss, color changes and softening	(Mohebbi et al. 2012)
Chitosan-banana flour	Films	Asparagus and corn	Inhibited growth of <i>Staphylococcus aureus</i>	(Pitak and Rakshit 2011)
		MP fruits and vegetables		
Cassava starch- potassium sorbate	Coatings	Minimally processed strawberry	Reduced respiration rate, increased water vapor resistance	(Garcia et al. 2010)
Gellan with geraniol	Coatings	Fresh-cut strawberry	Significantly reduced microbial counts	(Tomadoni et al. 2018)
Alginate- chitosan	Nanomultilayer coatings	Fresh-cut mangoes	Lower values of mass loss, pH, malondialdehyde con- tent, browning rate, soluble solids	(Souza et al. 2015)
к-Carrageenan- lysozyme	Nanomultilayer coatings	Whole and fresh-cut 'Rocha' pear	Presented lower mass loss, and total soluble solids and higher titrat- able acidity	(Medeiros et al. 2012a, b)
SPI with ferulic acid	Coatings	Fresh-cut apples	Effective in controlling their weight loss and firmness	(Alves et al. 2017)

Table 8.2 (continued)

(continued)

Coating/film composites	Form	Food products	Main benefits	References
Alginate with thyme/ cinna- mon/oregano oil	Coatings	Fresh-cut 'Red Fuji' apples	Significantly inhibited the microbial growth, respiration, weight loss, firmness and browning	(Sarengaowa et al. 2018)
Gelatin-chitosan	Multilayer coatings	Fresh-cut melon	Effective inhibition of the total microbial growth	(Poverenov et al. 2014)
Sodium alginate with lemongrass essential oil	Nanoemulsion coatings	Fresh-cut Fuji apples	A greater inactivation of <i>Escherichia coli</i> during storage time	(Salvia- Trujillo et al. 2015)
Whey protein- pectin	Crosslinked coatings	Fresh-cut apples	Reduced the weight loss, prevented micro- bial growth	(Rossi Marquez et al. 2017)
Chitosan	Coatings	Fresh-cut broccoli	Decreased in total mesophilic and psychrotrophic bacteria counts, inhibited open- ing florets	(Moreira et al. 2011b)
Starch with carvacrol	Coatings	Minimally processed pumpkin	Decreased counts of Escherichia coli O157: H7, and Staphylococ- cus aureus	(Santos et al. 2016)
Whey protein- pectin	Crosslinked coatings	Fresh-cut potatoes and carrots	Reduced the weight loss, prevented micro- bial growth	(Rossi Marquez et al. 2017)

Table 8.2 (continued)

bacteria, yeast, and fungi counts of fresh-cut melons (Poverenov et al. 2014). Souza et al. (2015) reported that the nanomultilayer coatings, made of alginate and chitosan, considerably inhibited putrefaction of fresh-cut mangoes during 14 days at 8 °C. At the end of the storage period, the lower values of mass loss, pH, malondialdehyde content, and browning rate were observed in the coated mangoes. Furthermore, nanoemulsion-based sodium alginate coatings with lemongrass essential oil at 0.5% or 1% (v/v) were created to completely inhibit the natural microflora of fresh-cut Fuji apples during 2 weeks at 23 °C. The application of this coating on fresh-cut apples exhibited a faster and greater inactivation of *Escherichia coli* during storage time compared with conventional emulsions (Salvia-Trujillo et al. 2015). Rossi Marquez et al. (2017) reported that transglutaminase crosslinked coatings prepared from whey protein and pectin were able to totally prevent the weight loss of fresh-cut potato and carrot at least until the sixth day of storage, which also maintained the phenolic and carotenoid content of fresh-cut carrot during storage.

Meat, poultry, and seafood products are common sources of proteins, yet susceptible to the spoilage microorganisms and food-borne pathogens. Thus, the hydrocolloid-based coatings and films with antimicrobial and/or antioxidant

Coating/film composites	Form	Food products	Main benefits	References
Chitosan with <i>Satureja</i> plant essential oil	Nanoencapsulation coatings	Lamb meat	Retention of the good quality char- acteristics, improvement of microbiological safety, and exten- sion of shelf life	(Pabast et al. 2018)
Chitosan– nanocelullose	Nanocomposite films	Ground meat	Decreased lactic acid bacteria population	(Dehnad et al. 2014)
Sodium caseinate with pomegranate peel extract	Films	Ground beef	More pronounced against gram- positive bacteria compared with gram-negative bacteria	(Emam- Djomeh et al. 2015)
Distiller dried grains-soluble protein with tea extract	Films	Pork meat	Decreased lipid oxidation	(Yang et al. 2016)
Perilla seed meal protein with clove oil	Films	Pork sausages	Reduced the micro- bial growth, and decreased peroxide value and thiobarbituric acid value	(Song et al. 2015)
Chitosan with thymus moroderi and piperella essential oil	Films	Cooked cured ham	Decreased the counts of aerobic mesophilic bacteria and lactic acid bac- teria, and lipid oxidation	(Ruiz- Navajas et al. 2015)
WPI with oreg- ano/clove essen- tial oils	Coatings	Chicken breast fillets	Decreased counts of total mesophilic aerobic, enterobacteriaceae, <i>Pseudomonas spp.</i> , and lactic acid bacteria	(Fernández- Pan et al. 2014)
Sodium caseinate with ginger essen- tial oil	Nanoemulsion coatings	Chicken breast fillets	Significantly decreased the total aerobic psychro- philic bacteria, maintained food color	(Noori et al. 2018)

 $\label{eq:stable} \textbf{Table 8.3} \hspace{0.1 cm} \text{Recent applications of hydrocolloid-based coatings and films in meat, poultry, and seafood}$

(continued)

Coating/film composites	Form	Food products	Main benefits	References
Chitosan-cyclo- dextrin with carvacrol	Films	Chicken breast fillet	A bactericidal effect against <i>Staphylococcus</i> <i>aureus</i> and <i>Escherichia coli</i> O157:H7	(Higueras et al. 2014)
Skate skin gelatin with thyme essen- tial oil	Films	Chicken tenderloin	Inhibited the growth of <i>Listeria</i> monocytogenes and <i>Escherichia coli</i> O157:H7	(Lee et al. 2016b)
Chitosan with lauric alginate ester	Coatings	Ready-to-eat deli Turkey meat	Reduced the growth of <i>Listeria</i> <i>innocua</i>	(Guo et al. 2014)
Sunflower seed protein-red algae with grapefruit seed extract	Films	Smoked duck meat	Decreased popula- tion of <i>Listeria</i> monocytogenes	(Song et al. 2013)
Chitosan with citric acid/licorice extract	Coatings	Japanese sea bass (Lateolabrax japonicas)	Reduced the TVB-N levels, showed antioxidant and antimicrobial effects	(Qiu et al. 2014)
Alginate with V _c / tea polyphenols	Coatings	Refrigerated bream (Megalobrama amblycephala)	Efficiently inhibited the growth of total via- ble counts, chemi- cal spoilage, and water loss	(Song et al. 2011b)
Chitosan with grape seed extract and tea polyphenols	Coatings	Refrigerated red drum (<i>Sciaenops</i> <i>ocellatus</i>) fillets	Maintained lower pH values, inhibited the degra- dation of ATP and lipid oxidation	(Li et al. 2013)
Chitosan	Coatings	Frozen Atlantic salmon	Maintained the color, controlled microbial activity	(Soares et al. 2015)
Chicken feather protein-gelatin with clove oil	Films	Smoked salmon	Decreased the populations of <i>Escherichia coli</i> O157:H7 and <i>Listeria</i> <i>monocytogenes</i> , decreased peroxide and thiobarbituric acid value	(Song et al. 2014)

Table 8.3 (continued)

(continued)

Coating/film composites	Form	Food products	Main benefits	References
Gelatin with lem- ongrass essential oil	Films	Sea bass slices	Retarded growth of lactic acid bacteria, psychrophilic bac- teria and spoilage microorganisms, lowered changes of color, K value, and total volatile base nitrogen	(Ahmad et al. 2012)
Quince seed mucilage with thyme/oregano essential oil	Films	Rainbow trout fillets	Decreased peroxi- dation values, reduced the changes of color, texture, and lipid oxidation	(Jouki et al. 2014)
Alginate-chitosan with grapefruit seed extract	Multilayer coatings	Shrimp (Litopenaeus vannamei)	Reduced the bacte- rial count and the off-flavor	(Kim et al. 2018b)
Chitosan/gelatin with Ziziphora clinopodioides essential oil and pomegranate peel extract	Films	Fresh shrimp	Decreased counts of bacterial, and population of <i>Listeria</i> <i>monocytogenes</i>	(Shahbazi 2018)
Starfish gelatin with vanillin	Films	Crab sticks	Inhibited the populations of <i>Listeria</i> monocytogenes	(Lee et al. 2016a)

Table 8.3 (continued)

compounds are produced to prolong their shelf life (Table 8.3). The incorporation of grape seed extract and tea polyphenols into chitosan coatings predominantly delayed the degradation of ATP and lipid oxidation of red drum during refrigerated storage (Li et al. 2013). Song et al. (2011b) reported the efficacy of alginate coatings enriched with V_c and tea polyphenols in inhibiting the growth of total viable counts, reducing chemical spoilage, and improving sensory quality of refrigerated bream compared to uncoated samples. According to Kim et al. (2018b), the multilayer coatings, based on alginate, chitosan, and grapefruit seed extract, were fabricated to reduce the bacterial counts and off-flavor of shrimp stored at 4 °C.

As shown in Table 8.3, the hydrocolloid-based coatings and films with nanoemulsion, nanoencapsulation, and nanocellulose have been created to extend shelf life of meat and seafood products. Dehnad et al. (2014) proved that the application of nanocomposite films based on chitosan and nanocellulose on ground meat decreased lactic acid bacteria population up to 3.1 logarithmic cycles (compared with nylon packaged sample) at 25 °C during 6 days of storage. Noori et al. (2018) showed that the addition of ginger essential oil nanoemulsion into sodium

caseinate coatings caused significant decrease of total aerobic psychrophilic bacteria of refrigerated chicken fillets during 12 days. The chitosan coatings included with nanoencapsulated *Satureja* plant essential oil were developed by Pabast et al. (2018) to improve the microbiological safety and prolong shelf life of lamb meat during chilled storage. Additionally, new plant extracts, as well as hydrocolloids based on non-conventional sources have been developed as potential ingredients of coatings and films (Shahbazi 2018; Jouki et al. 2014; Lee et al. 2016a; Ruiz-Navajas et al. 2015).

Cheese is nutritious food derived from milk. The shelf life of cheese is limited due to the uncontrolled and extensive fungal and bacterial proliferation on its surface. Table 8.4 shows some recent applications of antimicrobial coatings and films based on hydrocolloids in cheese. WPI coatings included with thyme and clove essential oils were produced by Kavas et al. (2015) to prolong the shelf life of semi-hard kashar cheese. The application of this coating on cheese retarded the growth of Listeria monocytogenes, Staphylococcus aureus, and Escherichia coli O157:H7 during 60 days of storage. Nanolaminate coatings based on alginate and lysozyme by LbL technique were fabricated to preserve "Coalho" cheese (Medeiros et al. 2014). After 20 days, coated cheese showed lower values of mass loss, pH, lipidic peroxidation and higher titratable acidity in comparison with uncoated cheese (Medeiros et al. 2014). Kim et al. (2018a, b) wrapped the Mozzarella cheese with chicken bone gelatine films containing cinnamon bark oil (1% w/v) and observed the reduction in the population of *Listeria monocytogenes* on mozzarella cheese during 20 days storage. In the current market, the commercialized hydrocolloid-based coatings, RIOCOBERT and RIOCOBERT PLUS (Becor Barbanza Ltd., A Coruña, Spain) effectively inhibited the growth of fungi on cheese (Fuciños et al. 2017).

For bakery and nuts products, most applications are hydrocolloid-based coatings rather than films. The coatings made from potato starch with potassium sorbate and citric acid were applied to extend shelf life of mini panettone (Ferreira Saraiva et al. 2016). Pinto et al. (2015) coated the cashew nuts with starch-cashew tree gum blend coatings to reduce moisture absorption, lipid oxidation, and the loss of crisp texture of nuts. Apart from that, hydrocolloid-based coatings are an additional method to improve unit operation efficiencies in the food industry. For example, they were applied in frying pre-treatments to reduce oil content in deep-fat fried products, such as chicken breasts (Dragich and Krochta 2010), potato chips (Hua et al. 2015), and fish cake (He et al. 2015). In osmotic dehydration processes of fruits and vegetables, hydrocolloid-based coatings can prevent large solute uptake without noticeably affecting water loss (Rodriguez et al. 2016; Azam et al. 2013).

7 Future Perspectives

Although hydrocolloid-based coatings and films have been utilized in food products, their mechanical and water barrier attributes are still weaker compared to those of synthetic plastic materials. Several approaches (e.g., bilayer, multilayer,

Coating/film composites	Form	Food products	Main benefits	References
Water chestnut starch-chitosan containing perilla oil	Coatings	Mongolian cheese	Delayed weight loss and the microbial growth	(Mei et al. 2013)
WPI-guar gum-sunflower oil with natamycin and lactic acid	Coatings	Cheese	Decreased water loss, hard- ness, and color change, inhibited pathogenic or contaminant microorganisms	(Ramos et al. 2012)
Sodium casein- ate-chitosan	Coatings	Cheese	Significantly inhibited the growth of mesophilic bac- teria, psychrotrophic, yeasts, and molds	(Moreira et al. 2011a)
Galactomannan with nisin	Coatings	Ricotta cheese	Against Listeria monocytogenes	(Martins et al. 2010)
WPI with thyme and clove essen- tial oils	Coatings	Semi-hard kashar cheese	Significant effect on the antimicrobial activity against <i>Listeria</i> monocytogenes	(Kavas et al. 2015)
WPI with ginger essential oil	Coatings	Kashar cheese	Inhibited the growth of Escherichia coli O157:H7 and Staphylococcus aureus	(Kavas et al. 2016)
Alginate- lysozyme	Nanolaminate coatings	'Coalho' cheese	Lower values of mass loss, pH, lipidic peroxidation, microorganisms' prolifera- tion and higher titratable acidity	(Medeiros et al. 2014)
Sodium caseinate with nisin	Films	Mini red Babybel cheese	Against <i>Listeria innocua</i> during storage at refriger- ated temperatures	(Cao-Hoang et al. 2010)
Starch with natamycin and nisin	Films	Port Salut cheese	Controlled Saccharomyces cerevisiae and Listeria innocua growth	(Ollé Resa et al. 2016)
Zein-carnauba wax with lysozyme	Films	Fresh Kashar cheese	A significant reduction in initial <i>Listeria</i> monocytogenes counts	(Ünalan et al. 2013)
Puffer fish skin gelatin with <i>Moringa oleifera</i> <i>Lam.</i> leaf extract	Films	Gouda cheese	Inhibited the <i>Listeria</i> <i>monocytogenes</i> growth, retarded the lipid oxidation	(Lee et al. 2016c)
Chicken bone gelatine with cin- namon bark oil	Films	Mozzarella cheese	Displayed antimicrobial and antioxidant activities, inhibited <i>Listeria</i> <i>monocytogenes</i>	(Kim et al. 2018a)
Red algae with grapefruit seed extract	Films	Cheese	Inhibited the growth of <i>Escherichia coli</i> O157:H7 and <i>Listeria monocytogenes</i> , decreased peroxide and thiobarbituric acid values	(Shin et al. 2012)

 Table 8.4
 Recent applications of hydrocolloid-based coatings and films in cheese

crosslinking, and bio-nanocomposite films, etc.) are employed to ameliorate properties of hydrocolloid-based coatings and films. Among them, incorporation of polysaccharide nanofillers into hydrocolloids to produce bio-nanocomposites has gained increasing attention in recent years, due to their edibility, remarkable physical performance, and functional properties (Otoni et al. 2017). Thus, this type of bio-nanocomposites is expected to be a promising area of research in the future.

On the other hand, the nanodelivery systems, such as nanoencapsulation, nanoliposomes, nanoemulsion, and nanolaminate, have emerged to enhance the performance of bioactive agents and improve their effectiveness in preserving food products. Currently, they are developed as the effective tools to augment the functionality of hydrocolloid-based coatings and films (Aloui and Khwaldia 2016). Future research should focus on the development of hydrocolloid coatings and films based on nanodelivery systems as well as their interactions with food products.

As a bottom-up approach, the structure-properties of hydrocolloid coatings and films should be studied further. Practically important properties such as WVP, TS, and EAB must be correlated with molecular structure and mobility in the solid state to further develop the utilization of polysaccharides. For instance, dextran, consisting of α -1,6 glycosidic linkages, shows a poor film-forming capacity in comparison with pullulan or amylose. In addition, dextran shows the largest molecular mobility in the solid state, followed by pullulan and amylose. The physico-chemical properties and molecular mobility of dextran, pullulan, and amylose in the solid state are quite different from each other because of the different modes of glucosidic linkages (Nishinari et al. 1985, 1992). Overall, hydrocolloids as packaging materials still need scientific research to improve their properties, quality and marketability. Further studies include (1) embracing big data and artificial intelligence (AI) in research and development, e.g., for process simulation, classification, pattern recognition, and transfer learning; (2) developing new techniques, equipment, machines for large-scale industrial implementation and applications.

Acknowledgemensts The author thanks Prof. Loong-Tak Lim of University of Guelph (Guelph, Canada), for his helpful suggestions and review of manuscript.

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