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Edible Packaging Materials

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Key Words

edible films and coatings, composite films, permeability, mechanical properties, active packaging, extrusion

Abstract

Research groups and the food and pharmaceutical industries recognize edible packaging as a useful alternative or addition to conventional packaging to reduce waste and to create novel applications for improving product stability, quality, safety, variety, and convenience for consumers. Recent studies have explored the ability of biopolymer-based food packaging materials to carry and control-release active compounds. As diverse edible packaging materials derived from various by-products or waste from food industry are being developed, the dry thermoplastic process is advancing rapidly as a feasible commercial edible packaging manufacturing process. The employment of nanocomposite concepts to edible packaging materials promises to improve barrier and mechanical properties and facilitate effective incorporation of bioactive ingredients and other designed functions. In addition to the need for a more fundamental understanding to enable design to desired specifications, edible packaging has to overcome challenges such as regulatory requirements, consumer acceptance, and scaling-up research concepts to commercial applications.

INTRODUCTION

Permeability: the diffusion-controlled molecular exchange of low molecular solute across a homogenous polymeric material via dissolution and desorption mechanisms

Edible packaging is rapidly advancing by utilizing edible compounds, such as proteins, polysaccharides, lipids and/or resins, and other edible components, derived from diverse renewable sources. Such edible packaging materials are intended to be integral parts of food products and to be eaten with the products, thus they are also inherently biodegradable in composting and other biological recycling (Krochta 2002). Edible packaging generally consists of edible films, sheets, coatings, and pouches. Edible films (thickness <254 μm or 10 mil) or sheets (thickness >254 μm) are stand-alone structures that are preformed separately from the food and then placed on or between food components or sealed into edible pouches, whereas edible coatings are thin layers of edible materials formed directly onto the surface of the food products (Krochta & De Mulder-Johnston 1997). Soft-gel capsules, hard-gel capsules, tablet coatings, and microcapsules made from edible materials could also be considered edible packaging. The main focus of this review is on edible films, coatings, and pouches.

Considerable interest and advanced research activity in edible packaging in the food and packaging industries have been driven by both increasing consumer demand for safe, high-quality, convenient foods with long shelf lives and also ecological consciousness of limited natural resources and the environmental impact of packaging waste. Edible packaging materials have been considered as attractive alternatives for some applications because of their unique properties, including the ability to protect foods with their barrier and mechanical properties, enhance sensory characteristics, control-release active ingredients, and control mass transfer between components of heterogeneous foods.

However, edible packaging materials are not normally meant to entirely replace conventional packaging. Rather, the efficiency of food preservation can be improved by using primary edible packaging together with nonedible packaging as secondary packaging to add additional protection from the atmosphere and prevent contamination from microorganisms or foreign particles. The utilization of edible packaging can reduce the complexity of overall packaging requirements by allowing conversion from multilayer or multilevel packaging to a single-component package, resulting in source reduction and improved recyclability of the simplified packaging system without compromising protective functions (Krochta 2002).

Edible packaging materials can also be used for nonedible packaging as an O_2- or grease-barrier layer to improve protective functions and biodegradability of multilayer packaging (Hong & Krochta 2003, 2004; Han & Krochta 2001, Chan & Krochta 2001a,b; Lin & Krochta 2003, Lee et al. 2008).

The unique advantages and versatility of edible packaging materials are envisioned to create green, innovative packaging for reducing waste and/or for improving product stability, quality, safety, variety, and convenience for consumers. This paper presents a review of recent developments in biopolymer- and lipid-based food packaging materials. It includes discussion of edible packaging functions, properties, promising applications, techniques used for developing edible packaging, and detailed descriptions of their film-forming ability and their associated applications. The review will also highlight challenges and opportunities for edible packaging materials.

FUNCTIONS, PROPERTIES, AND APPLICATIONS

Barriers to Environment

The most common rationale for the use of edible films and coatings is to control mass transfer between food and the ambient atmosphere. Permeability is an important property for selecting or tailoring edible materials for packaging. The effects of temperature and moisture content on

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biopolymer materials require that the permeabilities of edible packaging reflect the conditions of intended use. Thus, measurements of permeabilities should be conducted under the specific conditions that will be encountered by a packaged product.

a_w: water activity
WVP: water vapor permeability
RH: relative humidity
OP: oxygen permeability

Moisture barrier. Edible packaging can be used to inhibit moisture exchange between finished food products and the atmosphere. Changes in water activity (a_w) of packaged food can result in problematic microbial growth, undesirable textural changes, and deteriorative chemical and enzymatic reactions.

Water vapor permeability (WVP) values of films made from various edible packaging materials are compared to commonly used plastic film in **Table 1**, which illustrates the influence of film composition and test conditions. Overall, hydrocolloid-based films have quite high WVP compared to films of edible waxes and plastics. At high relative humidity (RH) and high plasticizer concentration, WVP of hydrophilic films increase because of their substantial polarity. Therefore, these films can only be used as protective barriers to moisture exchange for short periods or in low-moisture foods. On the other hand, lipids or other hydrophobic compounds are often used to make moisture barrier coatings or enhance the moisture-barrier properties of hydrocolloid-based films, because of their low water affinity, low polarity, and dense-structured molecular matrixes.

Oxygen barrier. Much of food deterioration is due to oxidation of lipids and food ingredients, discoloration of myoglobin in fresh meat cuts, or enzymatic browning of fresh-cut produce. Using edible packaging with low oxygen permeability (OP) preserves quality and extends shelf life of O_2-sensitive foods while reducing usage of expensive nonrecyclable O_2-barrier plastics.

In addition, the advanced development of edible films with defined gas permeabililty under certain storage conditions can create a modified atmosphere, suppressing the respiration rate of horticultural products and/or the ethylene production of physiologically active climacteric produce during storage and distribution (Baldwin 2007).

The OP values of some biopolymer-based and petroleum-based films are listed in **Table 1**. Edible films possess a wide range of OP values. Hydrocolloid-based films generally have impressive gas barrier properties, particularly at low RH. **Table 1** also shows the effect of plasticizer and test conditions on OP. At high RH condition, hydrocolloid-based films and hydrophilic EVOH film are plasticized by absorbed moisture, causing compromised barrier properties.

Aroma barrier. Barriers to volatile organic compounds are very important in preventing loss of characteristic volatile flavor or aroma and the migration of external off-flavors into packaged food during storage and distribution. Generally, the barrier efficiency of packaging is optimized when a migrating compound has low affinity to film materials and low diffusivity through the polymer matrix.

The hydrophilicity of protein- and polysaccharide-based edible films makes them excellent barriers to nonpolar aroma compounds. As a result, flavor and aroma encapsulation by carbohydrate and protein emulsion-based films have been proposed (Rosenberg & Lee 2004, Pegg & Shahidi 2007, Fabra et al. 2009, Hambleton et al. 2009). The purpose of this technology is to preserve the hydrophobic organic aroma compound or active ingredient in a nonpolar lipid dispersed phase, whereas the matrix made of hydrophilic polymer prevents aroma loss to the environment or oxidation.

Although studies of the aroma permeability of edible films are relatively challenging and limited compared with measurement of moisture and gas transport, methods to determine aroma permeability have been proposed (Debeaufort & Voilley 1994, Miller & Krochta 1998).

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Table 1 Water vapor and oxygen permeability values and mechanical properties of selected edible packaging films compared to synthetic polymer films

Film[a]	Water vapor permeability		Oxygen permeability		Mechanical properties			Reference
	Test conditions[b]	(g·mm/ m^2·d·kPa)	Test conditions	(cc·μm/ m^2·d·kPa)	TS (MPa)	EM (MPa)	% E	
Protein								
WG:Gly = 5:1	30°C, 100/0% RH	5.1						Gontard et al. 1992
WG:Gly = 3.1:1			30°C, 0% RH	17				Park & Chinnan 1995
WG:Gly = 2.5:1			25°C, 0% RH	6.1				Gennadios et al. 1993b
WG:Gly = 2.5:1	26°C, 50/100% RH	108	38°C, 0% RH	6.7				Aydt et al. 1991
WG:Gly:Suc = 15:6:0	25°C, 50/100% RH	121.55			4.2		89	Cherian et al. 1995
WG:Gly:Suc = 15:4:2	25°C, 50/100% RH	99.14			5.6		39	Cherian et al. 1995
WG:Gly:Suc = 15:3:3	25°C, 50/100% RH	81.90			6.0		11	Cherian et al. 1995
WG:Gly:Suc = 15:0:6	25°C, 50/100% RH	15.52			3.8		3.2	Cherian et al. 1995
WG:Gly:Sor = 15:3:3	25°C, 50/100% RH	105.17			5.7		57.2	Cherian et al. 1995
CZ:Gly = 5.67:1	25°C, 0/100% RH	24.24						Parris & Coffin 1997
CZ:Gly = 5:1			25°C, 0% RH	12				Gennadios et al. 1993b
CZ:Gly = 4.9:1	21°C, 85/0% RH	9.6	30°C, 0% RH	31				Park & Chinnan 1995
CZ:Gly = 2.3:1	25°C, 0/100% RH	35.52			7	498	2.6	Parris & Coffin 1997
CZ:PEG400 = 2.3:1					5.7	199	44.4	Parris & Coffin 1997
CZ:PPG400 = 2.3:1	25°C, 0/100% RH	16.8			9.4	608	2.8	Parris & Coffin 1997
CZ:Gly:PEG400 = 9.3:1:3					4.9	180	26.6	Parris & Coffin 1997
CZ:Gly:PPG400 = 9.3:1:3	25°C, 0/100% RH	25.68			5.1	135	117.8	Parris & Coffin 1997
CZ:Gly:PPG400 = 9.3:3:1	25°C, 0/100% RH	34.8			5.6	275	5	Parris & Coffin 1997
CZ:Gly:PPG400 = 4.67:1:1	25°C, 0/100% RH	31.2			3	100	112	Parris & Coffin 1997
CZ:PEG+Gly = 4:1–10:1	25°C, 50/100% RH	345.6–985.0			19.5–22.7		1.3–4.3	Ryu et al. 2002
CZ:OA = 4:1–10:1	25°C, 50/100% RH	259.2–388.8			17.5–21.7		1.8–5	Ryu et al. 2002
SPI:Gly = 4:1	28°C, 0/78% RH	39						Stuchell & Krochta 1994
SPI:Gly = 1.7:1	25°C, 50/100% RH	154						Brandenburg et al. 1993

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Collagen			RT, 0%RH	<0.04–0.53				Lieberman & Gilbert 1973
Collagen			RT, 63%RH	29.3				Lieberman & Gilbert 1973
Collagen			RT, 93% RH	890				Lieberman & Gilbert 1973
CC	25°C, 0/81% RH	28						Avena-Bustillos & Krochta 1993
CC:Gly = 2:1	23°C, 55/77% RH	190						Banerjee & Chen 1995
SC	25°C, 0/81% RH	37						Avena-Bustillos & Krochta 1993
SC:Gly = 2:1	23°C, 55/72% RH	310						Banerjee & Chen 1995
SC:Gly = 2:1					10.9–11.7	73.7–84.2		Siew et al. 1999
SC:Gly = 0.89:1–1.67:1	20°C, 45/0% RH	5.4–11.4						Siew et al. 1999
SC:PEG400 = 0.81:1–1.32:1	20°C, 45/0% RH	14.8–22.6						Siew et al. 1999
SC:PEG400 = 1.9:1					10.9–13.9	25.4		Siew et al. 1999
LAC:Gly = 0.6:1–1.4:1	37.8°C, 0/90% RH	54.7–59.3	23°C, 0% RH	0.73–2.18	0.42–2.51		121.4–253.6	Chick & Ustunol 1998
LAC:Sor = 0.6:1–1.4:1	37.8°C, 0/90% RH	34.0–45.0	23°C, 0% RH	0.65–0.81	2.43–11.65		50.6–170.7	Chick & Ustunol 1998
RC:Gly = 0.6:1–1.4:1	37.8°C, 0/90% RH	45.2–58.2	23°C, 0% RH	0.81–7.06	0.83–4.5		123.2–223.5	Chick & Ustunol 1998
RC:Sor = 0.6:1–1.4:1	37.8°C, 0/90% RH	39.6–49.6	23°C, 0% RH	1.84–1.02	3.83–15.12		4.9–17.9	Chick & Ustunol 1998
WPC:Gly = 2:1	23°C, 55/74% RH	255						Banerjee & Chen 1995
WPI:Gly = 2.3:1–5.7:1			23°C, 50% RH	18.5–76.1	29.1–13.9		4.1–30.8	McHugh & Krochta 1994a
WPI:Gly = 4:1	25°C, 0/77% RH	70						McHugh et al. 1994
WPI:Gly = 2:1	25°C, 55/73% RH	291						Banerjee & Chen 1995
WPI:Gly = 1:1–2:1	23°C, 50/100% RH	116–144			1–3.5	20–110	35–48	Shaw et al. 2002
WPI:Gly = 1:1–1.6:1	25°C, 0/100% RH	119.8–154.56						McHugh et al. 1994
WPI:Sor = 3.5:1			23°C, 40% RH	0.7				McHugh & Krochta 1994a

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Table 1 (*Continued*)

Film[a]	Water vapor permeability		Oxygen permeability		Mechanical properties			Reference
	Test conditions[b]	(g·mm/ m^2·d·kPa)	Test conditions	(cc·μm/ m^2·d·kPa)	TS (MPa)	EM (MPa)	% E	
WPI:Sor = 3.5:1			23°C, 70% RH	43.3				McHugh & Krochta 1994a
WPI:Sor = 1:1–2.3:1			23°C, 50% RH	4.3–8.3	14.0–14.7		1.6–8.7	McHugh & Krochta 1994a
WPI:Sor = 1:1–1.6:1	25°C, 0/100% RH	61.92–84.72						McHugh et al. 1994
WPI:Sor = 1:1–2:1	23°C, 50/100% RH	84–112			2.5–9	75–325	12–22	Shaw et al. 2002
WPI:Sor = 1:1	23°C, 50/72% RH	211						Anker at al. 1998
WPI:PEG200 = 1:1	25°C, 0/100% RH	134.64						McHugh et al. 1994
WPI:PEG400 = 1:1	25°C, 0/100% RH	129.6						McHugh et al. 1994
WPI:X = 1:1–2:1	23°C, 50/100% RH	84–89			0.5–8.5	80–275	2–15	Shaw et al. 2002
WPI:BW:S = 3.5:1.8:1			23°C, 50% RH	11.6				McHugh & Krochta 1994a
FMP:Gly = 1.9:1	20°C, 100/0% RH	6.1						Cuq et al. 1995
EWP:Gly = 2:1–3.3:1	25°C, 50/70% RH	211			4		12	Gennadios et al. 1996b
Polysaccharide								
HPMC	27°C, 0/85% RH	9.1						Hagenmaier & Shaw 1990
HPMC:PEG = 9:1	25°C, 85/0% RH	6.5						Kamper & Fennema 1984
MC	35°C, 90/0% RH	4.8						Hanlon 1992
MC:Gly = 1.52:1–5.88:1	30°C, 0/11% RH	9.07–10.80	30°C, 0% RH	101.03	20.83–37.5		50–100	Park et al. 1993
MC:Gly = 2.3:1	25°C, 0/52% RH	13.8	25°C, 52% RH	242	48.6		36.7	Donhowe & Fennema 1993
MC:PG = 1.52:1–5.88:1	30°C, 0/11% RH	8.64–30.24	30°C, 0% RH	242–1512.5	40–50		25–50	Park et al. 1993
MC:PG = 2.3:1	25°C, 0/52% RH	8.62	25°C, 52% RH	200	70.9		11.6	Donhowe & Fennema 1993
MC:PEG400 = 2.3:1	25°C, 0/52% RH	12.1	25°C, 52% RH	623	41.3		33.0	Donhowe & Fennema 1993
MC:PEG400 = 1.52:1–5.88:1	30°C, 0/11% RH	7.78	30°C, 0% RH	242	16.67–41.67		78–100	Park et al. 1993
MC:PEG1450 = 2.3:1	25°C, 0/52% RH	11.6	25°C, 52% RH	472	50.3		41.2	Donhowe & Fennema 1993

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MC:PEG8000 = 2.3:1	25°C, 0/52% RH	11.2	25°C, 52% RH	460	43.7		17.8	Donhowe & Fennema 1993
MC:PEG2000 = 2.3:1	25°C, 0/52% RH	10.3	25°C, 52% RH	351	45.0		13.3	Donhowe & Fennema 1993
HPC:Gly = 1.52:1–5.88:1	30°C, 0/11% RH	7.00	30°C, 0% RH	201.46	8.33–12.5		100–125	Park et al. 1993
HPC:PG = 1.52:1–5.88:1	30°C, 0/11% RH	12.96–20.74	30°C, 0% RH	605–1028.5	20–24		63–67	Park et al. 1993
HPC:PEG400 = 1.52:1–5.88:1	30°C, 0/11% RH	4.75	30°C, 0% RH	242	9–16.67		100–198	Park et al. 1993
Amylose	25°C, 100/0% RH	32						Rankin et al. 1958
HACS:Gly = 1:1–5:1	25°C, 50/100% RH	1011–1270			2–32		6–22	Ryu et al. 2002
HACS:Sor = 1:1–5:1	25°C, 50/100% RH	1011–1270			7–47		6–38	Ryu et al. 2002
LBG:PEG200 = 0.58:1–2.3:1	25°C, 84/0% RH	1.51–1.83						Aydinli & Tutas 2000
LBG:PEG400 = 0.58:1–2.3:1	25°C, 84/0% RH	1.51–2.19						Aydinli & Tutas 2000
LBG:PEG600 = 0.58:1–2.3:1	25°C, 84/0% RH	1.67–2.78						Aydinli & Tutas 2000
LBG:PEG1000 = 0.58:1–2.3:1	25°C, 84/0% RH	1.89–2.76						Aydinli & Tutas 2000
Gellan:Gly = 0.5:1	21°C, 0/54% RH	36			30	25	30	Yang & Paulson 2000
Gellan:PEG400 = 0.5:1	21°C, 0/54% RH				27	44	8	Yang & Paulson 2000
Pullulan:Sor = 3.3:1	21°C, 50/100% RH	2.88			29.2		2.6	Kim et al. 2002
Pullulan:Man = 3.3:1	21°C, 50/100% RH	3.6			15.7		9.5	Kim et al. 2002
Lipid								
Hydrogenated peanut oil	25°C, 100/0% RH	3.3						Lovegren & Feuge 1954
AMG	25°C, 100/0% RH	1.9–13						Lovegren & Feuge 1954
Tripalmitin	28°C, 0/100% RH	0.19						Shellhammer & Krochta 1997a
Carnauba wax	28°C, 0/100% RH	0.098						Shellhammer & Krochta 1997a
Beeswax	26°C, 0/100% RH	0.089						Shellhammer & Krochta 1997b
Paraffin wax	25°C, 100/0% RH	0.019						Lovergren & Feuge 1954
Candelilla wax	25°C, 0/100% RH	0.012						Shellhammer & Krochta 1997b

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Table 1 (*Continued*)

Film[a]	Water vapor permeability		Oxygen permeability		Mechanical properties			Reference
	Test conditions[b]	(g·mm/m²·d·kPa)	Test conditions	(cc·μm/m²·d·kPa)	TS (MPa)	EM (MPa)	% E	
Synthetic								
EVOH			25°C, 0% RH	0.027–0.18				Delassus 1997
EVOH			25°C, 100% RH	4.3–2.1				Delassus 1997
HDPE	38°C, 90/0% RH	0.025	20°C, 75% RH	390–780				Delassus 1997
LDPE	38°C, 90/0% RH	0.091	20°C, 75% RH	970–1400				Delassus 1997
Nylon-6	38°C, 90/0% RH	0.70	20°C, 75% RH	7.8–11.6				Delassus 1997
PET	38°C, 90/0% RH	0.12	20°C, 75% RH	12–16				Delassus 1997
PP	38°C, 90/0% RH	0.041	20°C, 75% RH	580–970				Delassus 1997
PS	38°C, 90/0% RH	0.047	20°C, 75% RH	970–1600				Delassus 1997
PVC	38°C, 90/0% RH	0.14	20°C, 75% RH	19–78				Delassus 1997

[a]WG, wheat gluten; CZ, corn zein; SPI, soy protein isolate; CC, calcium caseinate; SC, sodium caseinate; LAC, lactic acid casein; RC, rennet casein; WPC, whey protein concentrate; WPI, whey protein isolate; FMP, fish myofibrillar protein; EWP, egg white protein; HPMC, hydroxypropyl methylcellulose; MC, methylcellulose; HPC, hydroxylpropyl cellulose; HACS, high amylose corn starch; LBG, locust bean gum; Gly, glycerol; Suc, sucrose; PEG, polyethylene glycol; PG, propylene glycol; PPG, polypropylene glycol; Sor, sorbital; X, xylitol; M, mannitol; OA, oleic acid; BW, beeswax; AMG, acetylated monoglycerides; EVOH, ethylene-vinyl alcohol copolymer; HDPE, high-density polyethylene; LDPE, low-density polyethylene; PET, polyethylene terephthalate; PP, Polypropylene; PS, Polystyrene; PVC, polyvinylchloride.

[b]RHs on top and bottom sides of film (top/bottom).

Adapted from Krochta 2002, Sothornvit & Krochta 2005.

Oil barrier properties. Edible packaging can provide grease resistance to any lipid-containing products. Although standard test method ASTM F119 for rate of grease penetration of flexible barrier materials (ASTM International 2008) can be applied to edible films, quantitative data regarding oil permeability is very limited (Krochta 2002). Inherent hydrophilicity of protein- and carbohydrate-based polymer films is expected to give grease-resistant property to such films. For example, zein (Trezza & Vergano 1994) and whey protein (De Mulder-Johnston 1999) were shown to have excellent grease resistance.

Barrier to Mass Transfer During Food Processing

Edible coatings can potentially be used to improve product quality and efficiency of several food processing unit operations. Hydrocolloid-based edible coatings can be utilized to retain moisture and reduce fat uptake in deep-fat fried foods (Balasubramaniam et al. 1997, Mallikarjunan et al. 1997, Dragich & Krochta 2009).

Application of edible coatings to food products prior to osmotic dehydration can prevent valuable water-soluble ingredient loss as a result of diffusion into dehydration fluids and the penetration of the dehydrating agent into the food itself (Dabrowska & Lenart 2001). To maximize selective dehydration, the chosen edible coatings must have high WVP but low permeability to valuable ingredients and the dehydration agent. This process-aiding function could also be applied to other food preservation techniques, e.g., minimizing salt migration into foods during the brine-freezing process (Guilbert 1986) and limiting flavor and aroma loss during the freeze-drying operation.

Barrier to Mass Transfer Between Components in Multidomain Food Products

Intercomponent mass transfer phenomena in heterogeneous food affect sensory attributes and microbiological safety of the packaged food products. The classic method to limit moisture diffusion between multicomponent foods is reducing the a_w gradient by adding food-grade humectants, which often results in drastic sensory and physiochemical characteristic changes (Guilbert et al. 1997). A novel approach of placing edible film between domains can retard migration phenomena between food components of different a_w, flavor, aroma, and/or oil content, thus increasing both quality and shelf life (Krochta 2002). Furthermore, enrobing food products with edible coatings can simplify packaging of assorted selections of food products to satisfy consumer demand for variety.

Carrier of Antimicrobials, Antioxidants, and Other Food Additives

The ability of edible packaging to carry and control-release active compounds is a promising active food packaging function attracting current research interest and much attention from both food and pharmaceutical industries. Formulations can be designed to carry desired food additives (including antioxidants, antimicrobials, pigments, flavors, spices, salts, nutrient, light absorbers) as well as pharmaceutical or nutraceutical ingredients in the form of hard capsules, softgel capsules, microcapsules, soluble films/strips, flexible pouches, or coatings (Han & Gennadios 2005).

Depending on individual application, specific controlled-release rates of active solutes are required to fully perform their assigned functions. For example, limiting the migration rate of a preservative can reduce surface microbial growth. By slowing diffusion from the food surface into the food bulk, effective surface concentration of preservative can be maintained while decreasing the total concentration required. In certain cases, modification of an edible film matrix such as cross-linking could be used to reduce bioactive compound mobility. A controlled rate of release

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of active solute could be designed based on the chemical affinities between active ingredient, film-forming materials, and food product and the conditions (e.g., temperature, pH, a_w, time) to which the edible packaging containing the active ingredient would be subjected.

Antimicrobial edible packaging is a promising application that can be used alone or synergistically in combination with other preservation such as refrigeration, modified atmosphere packaging (Caillet et al. 2006), or irradiation (Ouattara et al. 2001, Kang et al. 2007) to improve microbial stability. Extensive effort has been given to incorporating natural and synthetic antimicrobial agents (**Table 2**) and antioxidants (**Table 3**) into various edible film-forming materials.

Tensile strength (TS): the largest stress (force/cross-sectional area) required to break the film

Elongation (E): the degree to which film specimen can stretch before breaking

Elastic modulus (EM): the ratio of stress to strain, indicating the material's resistance to elastic deformation

Glass transition temperature (Tg): the temperature at which amorphous phase of the polymer converts between rubbery and glassy states

Enhancer of Product Structural Integrity and Handling Characteristic

Structural reinforcement of fragile food products by edible coatings is a means to improve yield, facilitate handling, and protect food product from mechanical damage during the processing, transportation, storage, marketing, and end use. Coating fresh produce can lessen epidermal cell damage, thereby reducing surface browning and decay. Enrobing frozen foods, freeze-dried foods, or multicomponent foods with edible coatings can improve mechanical integrity and prevent fragmentation.

Favorable mechanical properties are essential for edible packaging to perform their protective functions efficiently. A standard method, ASTM D882 (ASTM International 2008), originally developed to evaluate tensile properties such as tensile strength (TS),% elongation (E), and elastic modulus (EM) of commercial plastic film structures, is also applied to edible films with special attention to test conditions. Mechanical properties for a selection of edible and synthetic films are listed in **Table 1**. Generally, protein films have lower TS than most polysaccharide films and petroleum-based films. The mechanical properties of edible films depend on structural cohesion, which is governed by the type and ratio of film-forming materials used, film-forming process, and experimental conditions.

Protein and carbohydrate films are often brittle and susceptible to cracking as a result of the strong cohesive energy density of the polymers. Plasticizers are commonly required to increase film flexibility by reduction of intermolecular H-bonding along polymer chains and increased intermolecular spacing. It is theorized that the resulting increase in molecular mobility lowers the glass transition temperature (Tg) and decreases the ratio of the crystalline region to the amorphous region (Guilbert et al. 1997). Both type and amount of plasticizers affect the interactions between polymer molecules. Increasing plasticizer concentration in the formula results in decreased TS and EM, and increased film elongation. Plasticizer efficiencies of polyols on mechanical properties of milk protein-based film were related to plasticizer size, shape, number, and spacing of oxygen atoms and water-binding ability (Sothornvit & Krochta 2001).

Commonly, the environmental or testing conditions affect both composition and structure of polymers, which directly affect mechanical and other properties of the resulting film. Hydrophilic edible packaging materials are normally sensitive to moisture because water molecules in the films can also function as plasticizers. Thus, increased RH lowers their TS and increases flexibility and stretchability. Temperature is also an important environmental variable affecting tensile properties of edible films. The mechanical strength of edible films decreases drastically when temperature rises above their Tg. High RH and plasticizer concentration can lower Tg of edible packaging.

Enhancer of Product Appearance

Application of edible coatings can enhance sensory attributes, including visual quality (e.g., color, glossiness) and tactile features (e.g., surface smoothness, nongreasy/sticky surface). Surface gloss

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value of biopolymer coating can be determined using a standard testing method ASTM D523 for specular gloss (ASTM International 2008). Lipid-based coatings such as shellac and wax coating have been ordinarily used to provide high-gloss finish on fruits. The most common polishing agents used in the finishing coat on candies and confectionary products are ethanol-based shellac and corn zein coatings. However, edible packaging production that requires ethanol must take into account appropriate safety measures, attention to environmental release of solvent to the atmosphere and solvent recovery in planning commercial operations (Krochta 2002). Furthermore, ethanol exposure can produce irreversible flavor changes in coated food products, hence limiting ethanol-based coating only to highly-flavored confectionery products. Utilizing water-based coating materials to impart a gloss characteristic for food products is considered to be a significant alternative to ethanol-based materials. Carbohydate coatings, e.g., cellulose or pectin, can impart an attractive gloss to the coated product when dry. Trezza & Krochta (2000) showed that ethanol-free, high-gloss whey protein coating has the same gloss as shellac, zein, dextrin, or hydroxyl methylcellulose (HPMC) coatings, and more stable gloss at high RH without blushing (whitening when exposed to moisture) like shellac and zein and less sticky than dextrin or HPMC coatings. Unlike zein glaze, with inherent native color and flavor, flavorless and colorless whey protein coatings do not interfere or cause flavor or color change to product, thus indicating feasible uses for foods such as chocolate and nuts (Lee et al. 2002a,b; Dangaran et al. 2006).

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Dispenser of Food Ingredients

New products can employ edible packaging to meet consumer demands for variety and convenience, including a quick-dissolving edible film in the form of a strip or pouch/sachet used to deliver a single dose of dry food ingredients or drugs. While providing primary protection to the premeasured portion, these quick-dissolving edible films provide consumer convenience for instant foods. They also facilitate accurate consistent formulation and mixing operations in manufacturing plants, thereby saving production time and cleaning requirements (Gennadios & Weller 1990, Janjarasskul & Krochta 2009a).

Sealability and instant solubility, preferably at cold temperatures, are crucial functional properties for edible pouches that serve as dispensers. A secondary package for edible pouches is necessary to protect these moisture sensitive pouches from environmental contamination and humidity. Evaluation methods for film total solubility (Handa et al. 1999) and protein solubility (Roy et al. 1999) have been proposed. The standard test method ASTM F88 for seal strength of flexible barrier materials (ASTM International 2008) is appropriate for edible pouches.

Commercial examples of edible film-dispensing applications include hydrocolloid-based oral strips for vitamins, over-the-counter medications and breath strips (Dixit & Puthli 2009), HPMC- and carboxymethyl cellulose (CMC)-based spice carrier films for delivering marinades, and pouches for unit-dose packaging of dry ingredients for food service, the food processors industry, and consumers.

FILM-FORMING PROCESS

Generally, formulations of edible packaging include at least one natural polymer capable of forming a sufficiently cohesive and continuous structural matrix. Functional properties of edible films and film coatings and their applications to food products depend on cohesion forces, including covalent bonds (e.g., disulfide bond cross-linking), ionic bonds, and H-bonding, between film-forming polymer molecules. Cohesive strength depends upon biopolymer structure and chemistry (e.g., molecular weight, regularity, branching, polarity), film-formation procedures (e.g., solution

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Table 2 Research on antimicrobial edible packaging

Antimicrobial agents	Edible packaging materials	Foods	Target microorganisms	References
Chitosan	Chitosan	Culture media	*Listeria monocytogenes*	Coma et al. 2002
Organic acids				
Acetic, propionic acid	Chitosan	Cooked ham, bologna, pastrami	Migration test, *Lactobacillus sakei*, *Serratia liquefaciens*	Ouattara et al. 2000
Acetic, citric, sorbic acid	Hydroxypropyl methylcellulose	Tomato	*Salmonella montevideo*	Zhuang et al. 1996
Sodium benzoate, potassium sorbate	Cellulose based	Cut apples, cut potatoes	Natural flora	Baldwin et al. 1996
Sodium benzoate, potassium sorbate	Methylcellulose, chitosan	Water-glycerol solution; culture media	Migration test; *Penicillium notatum*, *Rhodotorula rubra*	Chen et al. 1996
Benzoic acid	Methylcellulose	Fruit preserves	*Zygosaccharomyces rouzii*, *Z. mellis*	Chen et al. 1999
p-Aminobenzoic, sorbic acids	Whey protein isolate	Culture media	*L. monocytogenes Escherichia coli*, *Salmonella typhimurium*	Cagri et al. 2001
Potassium sorbate	Whey protein isolate	Water-glycerol	Migration test	Ozdermir & Floros 2001
Potassium sorbate	Whey protein isolate	Water-glycerol	Migration test	Franssen 2002
Potassium sorbate	Carrageenan	Buffer solution	Migration test	Choi et al. 2005
Sorbic acid	Corn zein	Cooked sweet corn	*L. monocytogenes*	Carlin et al. 2001
Sorbic acid	Wheat gluten, acetylated monoglyceride, beeswax	Aqueous medium	Migration test	Redl et al. 1996
Enzyme				
Lactoferrin, Lactoferrin hydrolysate, lactoperoxidase system	Whey protein isolate	Culture media	*Polytrichum commune*	Min & Krochta 2005
Lactoperoxidase system	Whey protein isolate	Culture media	*Salmonella enterica*, *E. coli O157:H7*	Min et al. 2005a
Lactoperoxidase system	Whey protein isolate	Culture media	*L. monocytogenes*	Min et al. 2005b
Lactoperoxidase system	Whey protein isolate	Smoked salmon	Migration test	Min et al. 2007
Lysozyme	Whey protein isolate	Smoked salmon	Migration test	Min et al. 2008b
Lysozyme, nisin, EDTA	Soy protein, corn zein	Culture media	*E. coli*, *Lactobacillus plantarum*	Padgett et al. 1998
Ovotransferrin	κ-Carrageenan	Chicken breast	*E. coli*, *S. typhimurium*, *Staphylococcus aureus*, *Candida albicans*	Seol et al. 2009

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Bacteriocins				
Nisin	Alginate	Ground beef	*Brochothrix thermosphacta*	Cutter & Siragusa 1997
Nisin	Alginate	Ground beef, beef slices	*S. aureus*	Millette et al. 2007
Nisin, EDTA, citric acid	Alginate	Poultry	*S. typhimurium*	Natrajan & Sheldon 2000
Nisin	Hydroxypropyl methylcellulose	Culture media	*L. monocytogenes, S. aureus*	Coma et al. 2001
Nisin	Hydroxypropyl methylcellulose	Culture media	*L. monocytogenes, S. aureus*	Sebti et al. 2002
Nisin	Corn zein, wheat gluten	Culture media	*L. plantarum*	Dawson et al. 2003
Nisin, lauric acid, EDTA	Corn zein	Culture media	*L. monocytogenes, Salmonella enteritidis*	Hoffman et al. 2001
Nisin, lauric acid	Soy protein	Turkey bologna	*L. monocytogenes*	Dawson et al. 2002
Nisin, pediocin	Cellulose casing	Turkey breast, ham, beef	*L. monocytogenes*	Ming et al. 1997
Nisin, sodium lactate, potassium sorbate	Sodium caseinate	Culture media	*L. monocytogenes*	Kristo et al. 2008
Nisin, grape seed extract, malic acid, EDTA	Whey protein isolate	Turkey frankfurter system	*L. monocytogenes, E. coli O157:H7, S. typhimurium*	Gadang et al. 2008
Pediocin	Corn zein, whey protein isolate	Culture media	*Listeria innocua*	Quintero-Salazar et al. 2005
Natural extracts				
Grape seed extracts	Pea starch films	Pork loins	*Brochothrix thermosphacta*	Corrales et al. 2009
Cinnamaldehyde	Chitosan	Bologna, ham	*Enterobac., Lactic acid bacteria, Lb. sakei, Serratia. Spp.*	Ouattara et al. 2000
Oregano, pimento, essential oils	Whey protein isolate	Whole beef muscle	*E. coli O157:H7*	Oussalah et al. 2004
Thyme oil, trans-cinnamaldehyde	Whey protein isolate, soy protein	Shrimp	*Pseudomonas putida*	Ouattara et al. 2001
Carvacrol (oregano oil), cinnamaldehyde (cinnamon oil), citral (lemongrass oil)	Alginate-apple puree	Culture media	*E. coli O157:H7*	Rojas-Grau et al. 2007
Carvacrol	Tomato puree	Culture media	*E. coli O157:H7*	Du et al. 2008
Green tea extract	Pectin	Cooked pork patty	*Aerobic bacteria*	Kang et al. 2007

Table 3 Research on antioxidant edible packaging

Antioxidant	Edible packaging material	Foods	Reference
Ascorbic acid	Whey protein isolate	Roasted peanut	Min & Krochta 2007
Ascorbic acid	Gellan	Stability test	Leon & Rojas 2007
Ascorbic acid	Whey protein isolate	Baby formula, peanut butter, mayonnaise	Janjarasskul & Krochta 2009b
Ascorbic acid, TBHQ	Cellulose based	Cut apples, cut potatoes	Baldwin 1996
α-Tocopherol, ascorbyl palmitate	Whey protein isolate	Physical properties test	Han & Krochta 2007
α-Tocopherol, ascorbyl palmitate	Whey protein isolate	Roasted peanut	Han et al. 2008
Natural extracts			
Borage extract	Sole skin gelatin, commercial fish gelatin	Antioxidant ability test	Gomez-Estaca et al. 2009
Oregano extract	SPI	Antioxidant ability test	Pruneda et al. 2008
Oregano, pimento essentials oil	WPI	Whole beef muscle	Oussalah et al. 2004
Fenugreek, rosemary, α-tocopherol	Egg albumen film	Cooked and uncooked poutry	Armitage et al. 2002

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casting, extrusion), film-formation parameters (e.g., temperature, pressure, solvent type, dilution, application technique, drying technique), and the concentrations of plasticizers and additives.

Edible coatings based on solid fats, waxes, or resins can be formed by melting and solidification, solubilizing in an organic solvent and then evaporating the solvent, or preparing an emulsion in water and then evaporating the water. On the other hand, forming hydrocolloid-based edible biopolymer films can be achieved by two basic technologies: the wet and dry processes.

The wet process, called solution casting, is based on a coacervation mechanism where hydrocolloid dispersed in aqueous suspension is precipitated after drying (Kester & Fennema 1986). It can also result from thermal gelation or coagulation followed by cooling (e.g., in the case of gelatin or agar). Depending on the number of biopolymers in the solvent system, the coacervation could be simple or complex. If a film formulation based on an emulsion is desired, a lipid material, and possibly a surfactant, is added to a biopolymer solution. The mixture is heated to above the lipid melting point, homogenized, and cast or directly applied while melted or after cooling (Krochta 2002). Food grade solvents for edible packaging are generally limited to only water and ethanol.

The degassed casting solution can be spread in a thin layer on a suitable base material surface that will release the film after drying to produce a preformed stand-alone film, or applied to form a coating directly on food products or drug tablets by spraying, dipping and subsequent draining, falling-film enrobing, brushing, pan coating followed by solvent removal for aqueous solutions, or cooling for lipid-based coatings (Krochta 2002, Baldwin 2007). Various drying methods have been developed for solution casting to produce self-supporting films including air drying, hot surface, infrared, and microwave techniques.

The film-coating formulation and conditions must be optimized to obtain a sufficient cohesiveness of the film matrix and adequate adhesion and coverage on the food surface in order for the coating to perform its intended functions. Although the thickness of the coatings depends primarily on the application technique and viscosity of the casting solutions, the adhesiveness of the coatings on the surface of food product is essentially governed by chemical affinity between the coatings and their supporting surfaces (Baldwin 2007). For example, hydrophilic film-forming solutions have poor adhesion on hydrophobic product surfaces. To enhance the adhesion in these cases, surface-active agents such as emulsifiers can be coated on the food or added to the film-forming solution.

Solution casting is often used as a method to evaluate the film-forming potential of biobased materials. The properties of cast films are measured using standard methods to predict how coatings from the same formulation would perform on the food.

In addition to advances in the wet process, noteworthy developments have been made in the dry thermoplastic extrusion process, which is based on the thermal properties of the film's biopolymer, including phase transitions, glass transitions, and gelatinization characteristics. The dry process is of considerable interest because it is a more cost-effective, with higher throughput than the wet process for making stand-alone films. In the extrusion process, the biopolymers are plasticized and heated above their Tg under low water content conditions to form a uniform melt by using heat, pressure, and shear in the barrel of the extruder. Besides formation of films, this soft and rubbery melt can be shaped into other useful forms using heat and pressure upon cooling or by using conventional processing techniques such as thermal compression molding or injection molding. In thermal extrusion, temperature, dry biopolymer feed rate, glycerol concentration, and screw speeds are important parameters that determine the extent of conformational changes, aggregation, and chemical cross-linking (Rhim & Ng 2007, Hernandez-Izquierdo & Krochta 2008, Hernandez-Izquierdo et al. 2008).

Many proteins exhibit thermoplastic behavior, e.g., collagen, gelatin (Park et al. 2008), wheat gluten (Zhang et al. 2008), corn zein (Wang & Padua 2003), casein, whey protein (Sothornvit et al. 2007, Hernandez-Izquierdo et al. 2008, Rauch 2008), soy protein (Cunningham et al. 2008), and fish myofibrillar protein (Cuq 2002). Carbohydrate-based films such as hydroxypropyl cellulose (Kester & Fennema 1986), starch (Pushpadass et al. 2009), pectin, and sodium alginate (Liu et al. 2006) are also potential materials for film formation by extrusion. Thermoplastic processing of two or more edible thermoplastic resins (Fishman et al. 2000) and incorporation of lipid components (Rauch 2008) and active ingredients (Liu et al. 2008) are gaining research interest in enhancing functional properties of films.

FILM-FORMING MATERIALS

Diverse materials used in edible packaging formulations generally fall into the categories of polysaccharides, proteins, lipids, and resins. Often, a plasticizer is added to increase flexibility. Other additives can be combined to modify and enhance physical properties or functionality of the films. An important research trend has been exploring food industry by-products and waste as potential edible packaging materials: whey protein from cheese production, chitosan from crustacean shells, corn zein from ethanol production, fish proteins from surimi wash water (Bourtoom et al. 2006), potato starch from potato chip waste, mung bean protein from mung bean starch (Bourtoom 2008), and fruit pomace from beverage production (Park & Zhao 2006). These avoid competition for food resources and reduce environmental impacts and waste disposal costs. Films can also be produced from fruit and vegetable purees (Rojas-Grau et al. 2006, Du et al. 2008, Sothornvit & Rodsamran 2008, Azeredo et al. 2009). Descriptions of common materials that form films are given below.

Proteins

Proteins are linear, random copolymers built from up to 20 different monomers. Proteins have attracted research attention and gained importance as potentially the most significant edible packaging materials. Molecularly, they provide an almost unlimited variety of film-forming materials that come from diverse sources that naturally differ in amino acid sequence. Thus, proteins lend themselves to producing a wide range of desirable properties and modifications by various

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methods. Depending on amino acid sequence, native protein structures are random coil, fibrous, or globular. The main random-coil protein is milk casein. Fibrous proteins (e.g., collagen) are fully extended and associate through H-bonding in parallel structures to form fiber. Globular proteins (e.g., wheat gluten, corn zein, soy protein, and whey protein) fold into spheroidal structures created by combinations of hydrophobic interactions and hydrogen, ionic, and disulfide bonds.

Many proteins can be used directly to form films from solution. A major mechanism of protein film formation involves the denaturation of the protein initiated by heat, solvents, or change in pH. The protein film matrices are subsequently formed when the extended peptide chains associate through new intermolecular interactions, depending on the protein, treatment, and fabrication conditions, resulting in modified film properties.

Protein-based films generally have good mechanical and optical properties. They are good barriers against the transport of O_2, CO_2, aroma, and lipid, but have high WVP. The barrier and mechanical properties of protein films are compromised by moisture owing to their inherent hydrophilic nature.

Many methods based on physical, chemical, and enzymatic treatments have been investigated to improve properties of edible protein films. A diverse variety of protein moieties were modified to create their final chemical properties and structure, for example, by adjusting the pH of a film-forming solution (Gontard et al. 1992, Brandenburg et al. 1993, Avena-Bustillos & Krochta 1993), breaking intramolecular disulfide bonds to form intermolecular cross-linking by heat denaturation (McHugh et al. 1994, Perez-Gago et al. 1999), and modifying protein side chains by adding salt or changing solvents. Using enzymatic treatments, such as transglutaminase to catalyze ε-(γ-glytamyl)-lysyl cross-links between the glutamine and lysyl groups (Stuchell & Krochta 1994, Lim et al. 1999, Chen 2002, Park et al. 2002b), also appears feasible.

Physical modifications include lamination, formation of composites, addition of emulsions or nanoparticles, aging, orientation, and annealing/heat curing (Gennadios et al. 1996a, Kim et al. 2002). Irradiation has been used to covalently cross-link aromatic amino acids (Gennadios et al. 1998, Vachon et al. 2000). Enhancing intermolecular interactions and improving molecular order with mechanical energy by ultrasound and microfluidization techniques have been attempted (Banerjee et al. 1996).

Wheat gluten (21CFR184.1322). Wheat gluten (WG) is composed mainly of gliadins and glutenins (alkali- and acid-soluble) fractions. Gliadin has large proline and glutamine contents but is low in charged amino acids, making gliadin a poorly ionized species across the entire pH range. Glutenin has similar amino acid composition to gliadin, with a slightly lower content of hydrophobic amino acids. Cysteine and cystine residues account for 2–3% of gluten's total amino acid residues.

Film formation from WG dispersions has been extensively studied. Because WG is water-insoluble, a complex solvent system with basic or acidic conditions in the presence of alcohol and disulfide bond-reducing agents is required to prepare casting solutions of WG films (Cuq et al. 1998). Generally, changing the pH of the medium disrupts hydrogen and ionic interactions, whereas ethanol disrupts hydrophobic interactions. Intermolecular and intramolecular disulfide bridges are cleaved and reduced to thiol groups when dispersing WG in alkaline environments or addition of reducing agents, e.g., sodium sulfite, cysteine, or mercaptoethanol, in acidic environments (Guilbert et al. 2002).

During drying of film-forming solutions, volatile solvents evaporate and WG becomes concentrated. Consequently, active sites for bond formation become free, allowing new intermolecular interactions to form. New H-bonds, hydrophobic interactions, and disulfide bonds contribute to the formation of a cohesive network. The moisture, gas, and solute barrier properties of

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WG-based films could be useful for active packaging, drug delivery systems or modified atmosphere packaging (Guilbert et al. 2002).

Corn zein (21CFR184.1984). Corn zein comprises a group of prolamins (alcohol-soluble proteins). Zein is insoluble in water except at very low or high pH and is insoluble in anhydrous alcohols (Gennadios & Weller 1990).

Zein coating or casting solutions are usually prepared by dissolving zein in warm, aqueous ethyl alcohol or isopropanol. Formation of films is believed to involve development of hydrophobic, hydrogen, and limited disulfide bonds between zein chains in the film matrix as the alcohol evaporates from the film surface (Padua & Wang 2002).

Zein is one of a few proteins used as a commercially successful finishing agent imparting surface gloss and acting as an O_2, lipid, and/or moisture barrier for nuts, candies, confectionery products, and other foods (Krochta & De Mulder-Johnston 1997). Pharmaceutical tablets are zein coated to achieve controlled release of active ingredients (Gennadios & Weller 1990) and to mask the taste of orally administered drugs (Meyer & Mazer 1997). Use of zein-based coatings has been suggested for reducing oil uptake by deep-fat fried foods (Mallikarjunan et al. 1997). Zein films of moderate gas barrier reduce moisture loss and respiration rates of vegetables (Park et al. 1994).

Soy protein isolate (21CFR182.90). Soy protein is commercially available in soy flour (50–59% protein), soy concentrate (65–72%), or isolate (>90%) (Park et al. 2002b). Fractionation of soy proteins using a solubility procedure yields albumin, globulin, and glutelin fractions. The globulin (saline soluble) is the predominant fraction that can be separated into 7S and 11S fractions.

Soy protein isolate (SPI) is most often used to prepare film-forming solutions, although 7S and 11S fractions have also been used (Kunte et al. 1997). When heat denatures casting solutions, soy protein films are formed via intermolecular disulfide bonds and hydrophobic interactions. Cysteine residues, present in both 7S and 11S fractions, undergo polymerization via a sulfhydryl-disulfide interchange reaction during the heat treatment to form a cohesive continuous covalent film network upon cooling.

SPI films can be applied on precooked meat product to control lipid oxidation and limit surface moisture loss (Wu et al. 2000). These films have good potential to carry flavoring agents (Kunte et al. 1997) or antimicrobial and antioxidant compounds (**Tables 2** and **3**). Also, SPI films may find application as microencapsulating agents of flavors and medications or in coatings of fruits, vegetables, and cheese (Petersen et al. 1999). Protective SPI coatings could also be used on certain food products, such as meat pies and high-moisture cakes, which require films that are highly permeable to water vapor (Gennadios et al. 1993a).

Collagen and gelatin. Collagen is a hydrophilic protein rich in glycine, hydroxyproline, and proline, thus it swells in polar liquids with high solubility parameters.

Collagen sausage casing is one of the most commercially successful edible protein films. Collagen film overwrap on refrigerated and thawed beef round steak reduced exudation without significantly affecting color or lipid oxidation (Farouk et al. 1990). The use of collagen-based films has been proposed for processed meats to reduce shrink loss, increase juiciness, allow for easy removal of nets after cooking or smoking, and absorb fluid exudates for a variety of cooked meat products.

Gelatin (21CFR172.230) is obtained by hydrolysis of collagen. The gelatin films can be produced by drying thermally reversible gelatin gel formed from cross-linking between amino and carboxyl components of amino acid residue side groups.

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Gelatin coatings can reduce O_2, moisture, and oil migration or carry bioactive ingredients (Krochta & De Mulder-Johnston 1997). Gelatin is used widely as an encapsulating agent in hard and soft gel capsules for low-moisture or oil-based food ingredients, dietary supplements, and pharmaceuticals (Baldwin 2007).

Milk protein. With an average concentration of ~3% (w/w wet basis) total protein in cow's milk, the two main classes of milk proteins are caseins (80%) and whey proteins (20%).

Caseins (21CFR182.90). Caseins are mostly phosphoproteins that form colloidal micelles in milk stabilized by calcium phosphate bridges. Caseins are characterized by low cysteine levels. Consequently, they cannot form extensive covalent inter- or intramolecular disulfide bonds to deliver water-insoluble films by heat denaturation (Chen 2002). Casein molecules have an open, flexible, random conformation that facilitates film formation from aqueous solution through their ability to form extensive intermolecular hydrogen and electrostatic bonds and hydrophobic interactions (McHugh & Krochta 1994b).

Casein films have been investigated to function as moisture barriers for water-soluble pouches, fresh produce, dried fruits, and frozen foods. The excellent O_2 barrier of casein-based films at low a_w has been shown to retard lipid oxidation of nuts. Furthermore, casein films have a great potential as carriers of flavor, nutrients, or bioactive ingredients (Gennadios et al. 1994).

Whey proteins (21CFR184.1979). Whey proteins (WPs) remain soluble after casein is precipitated at pH 4.6 during the cheese-making process. WPs are commercially available as whey protein concentrates (WPCs; 25–80% protein) and whey protein isolates (WPIs; >90% protein). WPs are globular and heat labile in nature. β-lactoglobulin, the predominant protein in whey, contains one free thiol group and two disulfide groups per monomer; four hydrophobic groups are located inside the globular structure.

Research on formation of WP films has mainly involved heat-induced molecular thiol-disulfide interchange reactions. Heating modifies the native globular structure to unfolded protein strands, exposing internal SH and hydrophobic groups, which promote intermolecular SS bonding and hydrophobic interactions upon drying to form water-insoluble films (McHugh & Krochta 1994b).

Pérez-Gago et al. (1999) observed that native WPI had ability to form water-soluble edible film without heat denaturation. Cohesion of native WPI films relies mainly on the intermolecular H-bonding created upon coacervation and solvent evaporation between native globular molecules, with most of the hydrophobic and SH groups buried in the interior of the molecule.

Both native and heat-denatured WPI films are transparent, flavorless, and have similar WVP and OP; however, they possess different solubility and mechanical properties. The unfolded structure and the disulfide bonding of heat-denatured WP film contribute to water insolubility and stronger, stiffer, tougher, and more stretchable films (Pérez-Gago & Krochta 2002). The low-energy bondings and the globular structure of native WP films account for complete solubility in water and lower strength, stiffness, and stretchability (Pérez-Gago et al. 1999).

WP films and coatings have been used as protective barriers to reduce O_2 uptake and rancidity in roasted peanuts (Maté et al. 1996) and frozen king salmon (Stuchell & Krochta 1995) and to reduce disintegration of fragile freeze-dried food. WPI films with O_2-scavenging function by incorporation of ascorbic acid have been developed (Janjarasskul & Krochta 2006). Ascorbic acid–incorporated WPI coatings have reduced OP (Janjarasskul & Krochta 2006, Min & Krochta 2007) and proven to notably retard lipid oxidation in coated peanut (Min & Krochta 2007), baby formula, peanut butter, and mayonnaise (Janjarasskul & Krochta 2009b). The ability to carry and control-release antimicrobial agents has been investigated (Min et al. 2005a,b). WPI

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lipid composite emulsion coatings were developed (Min et al. 2008a) and have proven to reduce enzymatic browning of fresh-cut apples (Pérez-Gago et al. 2005).

Other proteins. There are other proteins of limited availability or application, such as fish myofibrillar protein (Cuq 2002), egg white protein (Lim et al. 2002), keratin (Yamauchi & Yamauchi 2002), peanut protein, rice protein, pea protein, and sorghum protein (Park et al. 2002b), that may be of interest as a result of their unique properties or advantages.

Polysaccharides

Polysaccharides are long-chain polymers formed from mono- or disaccharide repeating units joined together by glycosidic bonds. As a result of the large number of hydroxyl groups and other hydrophilic moieties present in their structure, H-bonds play significant roles in film formation and characteristics. Generally, polysaccharide films are formed by disrupting interactions among long-chain polymer segments during the coacervation process and forming new intermolecular hydrophilic and H-bonding upon evaporation of the solvent to create a film matrix.

A variety of polysaccharides and their derivatives have been tested for potential use as edible packaging because they are abundant, low cost, and easy to handle. Polysaccharides have good film-forming properties with a wide range of coating solution viscosities. Polysaccharide films exhibit good mechanical and gas barrier properties (Baldwin et al. 1995) and are efficient barriers against oil and lipids but offer little resistance to water migration. Similar to other hydrophilic films, humidity greatly affects their functional properties.

Polysaccharides can easily be modified to improve their physiochemical properties by salt addition, solvent changes, heat gelatinization, pH changes, chemical modification of hydroxyl groups, cross-linking of polysaccharides, hydrolysis of polysaccharides, and employing nanotechnology (De Moura et al. 2009).

Cellulose derivatives. Cellulose is composed of linear chains of (1→4)-β-D-glucopyranosyl units. Cellulose is insoluble in aqueous solution by virtue of tightly packed polymer chains and a highly crystalline structure imparted by its regular structure and array of hydroxyl groups. Water solubility can be enhanced by etherification to produce cellulosic derivatives. Substitution of hydroxyl groups on the glucosyl units with bulkier groups helps separate the polymer chains in crystalline structure by disrupting intramolecular H-bonding. The common commercial water-soluble cellulose ethers, including methyl cellulose (MC; 21CFR182.1480), hydroxypropyl cellulose (HPC; 21CFR172.870), hydroxypropylmethyl cellulose (HPMC; 21CFR172.874), and carboxymethyl cellulose (CMC; 21CFR182.70), all possess good film-forming properties. The degree of substitution, types of substitution for functional groups, and polymer chain length affect permeability, mechanical properties, and solubility (Sanderson 1981).

Edible coatings from these cellulose ethers have been applied to a variety of foods to provide barriers to moisture, O_2, or oil. Owing to their ability to form thermally induced gelatinous coatings, MC and HPMC have been used as batter ingredients to lessen oil uptake and moisture loss during deep-fat frying (Sanderson 1981, Balasubramaniam et al. 1997). MC and HPMC solutions are widely used in the pharmaceutical industry for tablet coating. Water-soluble, edible pouches from MC and HPMC are used commercially to deliver preweighed dry food ingredients.

Starch (21CFR182.70). Amylose is the liner polymer composed of (1→4)-α-D-glucopyranosyl monomers. Amylopectin is a highly branched molecule that contains the amylose backbone with side units of D-glucopyranosyl linked by α-1,6-glycosidic bonds. Native starch molecules arrange

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themselves in the form of starch granules in which amylose and amylopectin are structured by H-bonding in the orderly manner of semicrystalline domains alternating with amorphous rings. To enhance water solubility, partial etherification of high-amylose starch with propylene oxide can be performed to yield hydroxypropylated derivatives. Amylose, high amylose starch, and hydroxypropylated high amylose starch have been used to form self-supporting films by casting from aqueous solutions of gelatinized starch.

Amylose and hydroxypropyl amylose films have been formed as protective edible coatings on foods and encapsulating agents, to provide an O_2 or lipid barrier and to improve appearance, texture, and handling. Edible starch films and coatings are commonly used in bakery, confectionary, batters, and meat products.

Chitosan. Chitin is a β-1,4-linked linear polymer of 2-acetamido-2-deoxy-D-glucopyranosyl residues. Chitosan is produced by fusion of chitin with alkalies. It is not currently approved for use in the United States as a food additive. Chitosan films can be formed by casting acidic aqueous solutions. The viscosity of chitosan solutions may differ with the type of organic acid solvent used, thus affecting film properties (Rhim et al. 1998).

Chitosan coatings with semipermeability can be used to increase postharvest life of fresh fruits (Wang et al. 2007). Amino groups of chitosan confer opportunities for chemical modification because cationic groups can react with any negatively charged substances, e.g., fats, cholesterol, basic ions, and proteins. The cationic property of chitosan offers antimicrobial and antioxidant activities, as well as the ability to carry and slow-release functional ingredients (Coma et al. 2002). The use of chitosan coatings to delay enzymatic browning in fresh produce has been reported (Zhang & Quantick 1997).

Pectin (21CFR184.1588). Pectins are water-soluble anionic polymers composed mainly of (1→4)-α-D-galactopyranosyluronic acid units. Pectins with a degree of esterification (DE) above 50% are labeled high-methoxyl pectin (HMP) and below 50% are termed low-methoxyl pectin (LMP). The differences in methyl ester content and DE affect solubility and gelation properties of pectin (Baldwin et al. 1995). HMP forms gels with sugar and acid, especially in jams and jellies. LMP, produced from chemical de-esterification, forms gels in the presence of divalent cations. Calcium cations bridge adjacent LMP chains via ionic interactions and with interchain H-bonding yield a 3D-gel network.

Edible pectin film can be formed by evaporating water from pectin gel. Although pectinate coatings are not adequate moisture barriers, they can retard water loss from enrobed food by acting as a sacrificing agent when moisture evaporates from their gel matrix rather than dehydrating the food significantly. Pectin coatings have been investigated for their ability to retard moisture loss and lipid migration and improve handling and appearance of foods (Kester & Fennema 1986).

Alginate (21CFR184.1011). Alginates are salts of alginic acid that is a linear (1→4) linked polyuronic acid containing three types of block structures: poly-β-D-manopyranosyluronic acid (M) block, poly-α-L-gulopyranosyluronic acid (G) blocks, and MG blocks containing both polyuronic acids. These highly anionic polymers have the ability to form instantaneous gel structures by reacting with divalent or trivalent cations, without heating or cooling, similar to LMP.

Alginate films can be formed from evaporating solvent from alginate gel or by a two-step procedure that involves drying of alginate solution followed by treatment with a calcium salt solution to induce instantaneous cross-linking at the interface.

Film strength and permeability can be altered by the concentration of polyvalent cations, the rate of its addition and time of exposure, pH, temperature, and the presence of composite

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constituents (Kester & Fennema 1986). Gelatinous alginate coatings effectively lessen desiccation in enrobed meats by acting as a sacrificing agent. Owing to their good O_2 barrier properties, alginate coatings can protect foods against oxidation. Alginates also have versatile uses in encapsulation (Hambleton et al. 2009).

Carrageenans (21CFR172.620). Carrageenan is a complex mixture of several polysaccharides. Three principal carraggeenan fractions, kappa (κ), iota (ι), and lambda (λ), differ in sulfate ester and 3,6-anhydro-α-D-galactopyranosyl content, which results in various degrees of negative charge and solubility in water. Thermoreversible carrageenan gels can be used as food coatings to retard moisture loss from an enrobed food by acting as a sacrificing agent (Kester & Fennema 1986). Carrageenan has been applied to a variety of foods to carry antimicrobials and to reduce moisture loss, oxidation, or disintegration (Nieto 2009) and has been studied for flavor encapsulation (Fabra et al. 2009).

Exudate gums. Gum arabic (21CFR184.1330), gum ghatti (21CFR184.1333), gum karaya (21CFR184.1349), and gum tragacanth (21CFR184.1351) are structurally complex heteropolysaccharides commonly used as an encapsulation coating material (Pegg & Shahidi 2007).

Seed gum. Guar gum (21CFR184.1339) is a polysaccharide having a straight chain of β-D-mannopyranosyl with single α-D-galactopyranosyl units attached as side chains in a ratio of 1:2. Guar gum is soluble in cold and hot water, giving high viscosity solutions. Calcium can cross-link guar gum, causing it to gel. Locust bean gum (21CFR184.1343) is a neutral polysaccharide consisting of β-D-mannopyranosyl and α-D-galactopyranosyl units in a ratio of 4:1. It is soluble in hot water, producing high viscosity solutions over a wide range of pH values and temperatures.

Microbial polysaccharides. Gellan gum (21CFR172.665) is a water-soluble polysaccharide, principally composed of a tetrasaccharide repeating unit of (1→3)-β-D-glucopyranosyl, (1→4)-β-D-glucopyranosyluronic acid, (1→4)- β-D-glucopyranosyl, and (1→4)-α-L-rhamnopyranosyl units. Gel of gellan gum can be formed by heating the solution in the presence of cations. Upon cooling, the polymer chains can assume double helices, which aggregate into a weak gel structure supported by van der Waals attractions. In the presence of cations, the double helices form cation-mediated aggregates leading to the formation of strong gel associations. The gel properties are strongly affected by cation type, ionic strength, and gum concentration. Pullulan (21CFR170.36) and xanthan gum (21CFR172.695) are also extracellular microbial polysaccharides exhibiting good film-forming ability (Nieto 2009).

Lipids

Many edible lipid materials have been utilized as protective coatings against moisture transfer and to add sheen. Unlike other macromolecules, lipid and resin compounds are not biopolymers. They do not have a large number of repeating units connected by covalent bonds to form a large molecular structure. Thus, they are fragile and do not generally form cohesive, self-supporting film structures. Owing to their relatively low polarity, lipids and resins have been incorporated into edible film-forming materials to provide a moisture barrier within composite films (Greener & Fennema 1989). Nevertheless, there are disadvantages of employing lipids in edible packaging materials, such as their waxy taste and texture, greasy surface, and potential rancidity.

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Glycerol esters. Neutral esters of glycerol and fatty acids, including mono-, di-, and triacylglycerides have been used alone or in combination with other edible ingredients to coat food products. The properties, e.g., solubility and resistance to water vapor, of fatty acids and the lipids derived from them are markedly dependent on their physical state, chain length, and degree of saturation. Generally, increased degree of unsaturation or branching of acyl chain and/or reduction in carbon chain length result in increased WVP (Kamper & Fennema 1984). This is a consequence of the enhanced mobility of hydrocarbon chains and less efficient lateral packing of acyl chains caused by interchain reductions in van der Walls attraction.

Acetylated monoglycerides (21CFR172.828, 21CFR175.230) are modified fats in which a single fatty acid attached to the glycerol molecule is substituted by acetic acid. The degree of acetylation and the type of monoglyceride yield acetic acid esters with different properties. The WVP of acetoacylglycerol improved as the degree of acetylation increased, which was hypothesized to be a consequence of differences in crystal packing or removal of free hydroxyl groups that would otherwise interact with migrating water. Certain associated problems include an acidic, bitter aftertaste and the tendency of highly saturated acetylated glycerides to crack and flake during storage (Bourlieu et al. 2008). Fatty acids (21CFR172.86), fatty alcohols (21CFR178.3480), and sucrose fatty acid esters (21CFR172.859) can also be used as lipid coatings.

Waxes. Waxes are esters of a long-chain fatty acid with a long-chain alcohol. They are substantially more resistant to diffusion of water than most lipid or nonlipid edible films, owing to the very low level of polar groups. (Kester & Fennema 1986).

Both natural waxes, e.g., carnauba wax (21CFR184.1978), candelilla wax (21CFR184.1976), rice bran wax (21CFR172.890), and beeswax (21CFR184.1973), and synthetic waxes, e.g., paraffin wax (21CFR175.250; 21CFR175.300) and petroleum wax (21CFR172.886), have been used as protective coatings, alone or in combination with other ingredients (Baldwin 2007).

Resin. Edible resins, such as shellac (21CFR175.300), terpene resin (21CFR172.280), and wood rosin (21CFR175.300) are used to impart gloss to food commodities (Baldwin 2007). Shellac is composed of a complex mixture of aliphatic and alicyclic hydroxyl acid polymers, such as aleuritic and shellolic acids. It is soluble in organic solvents and in alkaline solutions. Shellac has been used extensively as edible coating for confectionary and fresh produce, and as an enteric coating for pharmaceuticals (Rhim & Shellhammer 2005, Baldwin 2007).

Composite Materials

Edible composite packaging materials have been developed by blending biocomponents for specific applications, aiming to take advantage of complementary functional properties or to overcome their respective flaws (Krochta & De Mulder-Johnston 1997). Edible films based on polar biopolymers, i.e., polysaccharides and proteins, are generally efficient gas barriers and have moderately good mechanical properties at low RH. However, proteins and polysaccharides give water-sensitive films with poor moisture-barrier performance and markedly degraded gas barrier and mechanical properties at high humidity. On the contrary, hydrophobic lipids are effective against moisture migration, but their mechanical properties are much inferior to those of hydrocolloid films because of their nonpolymeric nature. Most of the composite films studied to date involve combining lipid compounds with a hydrocolloid-based structural matrix. The lipid components in the formulation reduce water transmission, whereas the hydrocolloid components serve as selective gas barriers and provide strength and structural integrity (Gontard et al. 1994).

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Generally, composite films can be fabricated as either a hydrocolloid-lipid bilayer or a stable lipid emulsion in a hydrocolloid matrix. The bilayer composite films have a distinct second layer made of a lipid component over the hydrocolloid-based film. In the emulsified lipid composite films, lipid globules are uniformly dispersed, sometimes with the aid of an emulsifier, and entrapped throughout the dried continuous support matrix of hydrocolloid components (Perez-Gago & Krochta 2005). The bilayer film can be formed by two different techniques: the coating technique or the emulsion technique. The coating technique involves casting or laminating a molten lipid or film-forming lipid solution onto a dried, preformed polysaccharide or protein film to prepare bilayer films, similar to bilayered composite plastic films. Alternatively, the emulsion technique involves solubilizing the lipid into the film-forming solution prior to film casting. The bilayer film is formed later during the drying process from phase separation, as the film-forming solution becomes an unstable emulsion (Perez-Gago & Krochta 2005).

Both bilayer and stable emulsion films offer unique advantages and limitations. Commonly, bilayer films have better water vapor barrier efficiencies than stable emulsion films and coatings owing to the continuous hydrophobic layer in the film (Debeaufort & Voilley 1995). The main disadvantage of the coating technique is that the bilayer structure has a tendency to crack and/or delaminate. Furthermore, the coating technique requires two different castings and the use of lipid solvent or handling of molten lipid at high temperature. However, fabricating stable emulsion-based film could be viewed as more challenging, as it must take into account lipid melting temperature, gelation/cross-linking, and solvent volatilization of the structural network.

The moisture resistance of both bilayer and emulsion films strongly depends on the polarity and the degree of saturation of the lipid component. Furthermore, variations in homogeneity, location, volume fraction, and polymorphism of the lipid phase affect the barrier properties of emulsion films (Perez-Gago & Krochta 2005).

Noteworthy advances in composites, especially in recent publications, seem to be correlated with the evolution of nanotechnology. The use of bionanocomposite materials for edible packaging promises to improve barrier and mechanical properties beyond what could be achieved by utilizing macroscopic reinforcing components (Rhim & Ng 2007, Sorrentino et al. 2007, Azeredo et al. 2009, De Moura et al. 2009). A uniform dispersion of nanoparticles leads to a very large matrix/filler interfacial area, which changes the nanostructure, molecular mobility, the relaxation behavior, and the consequent thermal and mechanical properties of the material (Dalmas et al. 2007). Furthermore, the nanocomponents are expected to improve barrier properties by increasing the toruosity of the path that water, gases, or low-molecular-weight compounds take to penetrate the films.

Film Additives

Film additives are materials other than film formers incorporated to enhance structural, mechanical, and handling properties or to provide active functions to the films.

Plasticizers. Plasticizers are typically small-molecular-weight hydrophilic agents added to film-forming preparations to improve film mechanical properties by situating themselves in their polymeric network and competing for chain-to-chain H-bonding along the polymer chains.

Commonly used plasticizers in edible packaging are mono-, di-, or oligosaccharides (e.g., glucose, fructose-glucose syrups, and sucrose), polyols (e.g., glycerol, sorbitol, glyceryl derivatives, and polyethylene glycols), and lipids and derivatives (e.g., phospholipids, fatty acids, and surfactants). Generally, the selection of plasticizers requires considering plasticizer compatibility, efficiency, permanence, and economics (Sothornvit & Krochta 2005).

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Emulsifiers. Emulsifiers are surface active compounds, with both polar and nonpolar character, capable of modifying interfacial energy at the interface of immiscible systems, such as a water-lipid interface or a water-air surface.

Emulsifiers are essential for the formation and stabilization of well-dispersed lipid particles in composite emulsion films or to achieve sufficient surface wettability to ensure proper surface coverage and adhesion to the coated surface (Krochta 2002). Some common emulsifiers are acetylated monoglyceride, lecithin, glycerol monopalmitate, glycerol monostearate, polysorbate 60, polysorbate 65, polysorbate 80, sodium lauryl sulfate, sodium stearoyl lactylate, sorbitan monooleate, and sorbitan monostearate. Many proteins have emulsifying properties owing to their amphiphilic nature.

Antimicrobials. Incorporation of both natural and synthetic antimicrobial agents into various edible packaging has been developed as an effective alternative for controlling the growth of microorganisms (**Table 2**).

Organic acids and their salts. Benzoic acid (21CFR184.1021) and its salts are most effective in the undissociated form at pH 2.5–4.0. This preservative is more active against yeasts and molds than bacteria. Sorbic acid (21CFR182.3089) and its salts are effective in pH range 3.0–6.5 against a broad spectrum of yeast and molds and lactic acid bacteria. Acetic (21CFR 184.1005), lactic (21CFR 184.1061), propionic (21CFR 184.1061), and fumaric (21CFR 172.350) acids also can be used in coatings and contribute to antimicrobial activity.

Chitosan. Chitosan's antimicrobial activity is most effective against yeasts and molds, followed by gram-positive bacteria and gram-negative bacteria. Its mechanism of antimicrobial action was proposed to be a deleterious leakage of microbial proteinaceous and intercellular components as a result of the interaction between positively charged chitosan and the negatively charged microbial cell membrane. Chitosan also chelates trace metals, thus preventing microbial growth and toxin production (Cuero et al. 1991).

Plant extracts. Essential oil extracts from plants, e.g., grapefruit seed, cinnamon, allspice, clove, thyme, rosemary, onion, garlic, radish, mustard, horseradish, and oregano, are rich in phenolic compounds such as flavonoids and phenolic acids, which exhibit a wide range of biological effects, including antioxidant and antimicrobial activity (Oussalah et al. 2004). These naturally occurring antimicrobial agents can be added into foods without labeling as antimicrobial agents or preservatives (Suppakul et al. 2003).

Bacteriocins. Bacteriocins are protein-containing macromolecules produced by various bacteria and possessing different antibacterial spectra, modes of action, and chemical properties. They are generally heat stable, hypoallergenic, and readily degraded by proteolytic enzymes in the human intestinal tract. Numerous bacteriocins have been characterized, such as colicins (*Escherichia coli*), lacticin (*Lactococcus lactis*), pediocins (*Pediococcus acidilactici*), and nisin (*Lactococcus lactis*). Nisin (21CFR184.1538) remains the most commercially important bacteriocin because of its history of safe use and documented effectiveness against gram-positive pathogenic and spoilage bacteria. Nisin interacts with the sulfur-containing compounds in bacterial membranes, disrupting their semipermeable function and causing cell lysis (Thomas et al. 2000).

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Enzyme. Lysozyme and lactoperoxidase are widely studied antimicrobial enzymes isolated from various natural sources, e.g., milk. Lysozyme is a single-chain protein that possesses the ability to hydrolyze β (1→4) glycosidic linkages between *N*-acetylmuramic acid and *N*-acetylglucosamine found in peptidoglycan cell walls of both gram-positive and gram-negative bacteria. The loss of structural integrity of the cell walls causes lysis of bacterial cells (Shah 2000). Lysozyme is less effective against gram-negative bacteria, because of the lipid-based outer membrane over their cell walls.

Lactoperoxidase catalyzes the oxidation of the thiocyanate ion, generating oxidizing products such as hypothiocyanite and hypothiocyanous acid. These inhibit microorganisms by oxidizing sulphydryl groups of microbial enzymes and other proteins, resulting in structural damage to cytoplasmic membranes that causes detrimental leakage of potassium ions, amino acids, and peptides from microbial cells (Kussendrager & van Hooijdonk 2000).

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Antioxidants. Antioxidants are chemical compounds that delay the start or slow the rate of oxidation reactions (**Table 3**). Antioxidants are broadly classified by mechanism of action as primary and secondary antioxidants. Primary or chain-breaking antioxidants are free radical acceptors that delay the initiation or propagation step of autoxidation. Examples are phenolic antioxidants, including butylated hydroxyanisole (21CFR182.3169, 21CFR172.115), butylated hydroxytoluene (21CFR182.3173), propyl gallate (PG,; 21CFR184.1660) and tertiary butylhydroquinone (21CFR172.190). Tocopherols (21CFR182.8890) are the most commonly used natural primary antioxidants.

Secondary or preventive antioxidants retard oxidation by several different actions, e.g., chelating pro-oxidant metals, replenishing hydrogen to primary antioxidants, decomposing hydroperoxides to nonradical species, deactivating singlet O_2, absorbing UV radiation, scavenging O_2, or promoting antioxidant activity of primary antioxidants. Citric acid (21CFR182.1033, 21CFR182.6033), ascorbic acid (21CFR182.8013, 21CFR182.5013), ascorbyl palmitate (21CFR182.3149, 21CFR172.110), and tartaric acid (21CFR184.1099) are good examples of synergists.

Metal sequesters, e.g., ascorbic acid, citric acid, phosphoric acid (21CFR182.1073), and erythorbic acid (21CFR182.3041), inhibit polyphenol oxidase activity that causes browning of fresh cut produce. Numerous plants have been identified as sources of natural phenolic compounds with antioxidant activity, e.g., herb extracts (e.g., rosemary, sage, thyme), sesamol (sesame seed), tea catechins, and gossypol (cotton plant).

CURRENT STATUS, CHALLENGES, AND OPPORTUNITIES

The potential of edible packaging has been well recognized by many research groups and by the food and pharmaceutical industries as an alternative or synergistic addition to conventional packaging to enhance food protection and/or package recyclability. Diverse innovative utilizations of edible films and coatings have been proposed as novel applications as well as alternatives to existing technologies. The dry thermoplastic process is advancing rapidly as a feasible commercial edible packaging manufacturing process, as numerous edible packaging materials derived from by-products or waste from the food industry are being explored and developed. Nanocomposites are also at the forefront of edible packaging research and development. Nanotechnology allows scientists to engineer the nanostructure of packaging materials to achieve desirable barriers and mechanical properties, to carry bioactive ingredients, and to better perform their designed functions. Nevertheless, edible packaging still has to overcome several challenges to achieve significant commercial application. In general, lack of knowledge and data prevents design of edible films

for desired specifications. Given that edible packaging serves both as a packaging and a food component, it must fulfill rigorous requirements and challenges.

Regulation

Components of edible packaging must meet all required regulations regarding food products (Guilbert & Gontard 1995). Depending on their applications, edible films and coatings could be classified as food products, food ingredients, food additives, food contact substances, or food packaging materials (Debeaufort et al. 1998). In the case of pharmaceutical and nutraceutical applications, there may be other regulations.

Materials used in formation of edible packaging must be generally recognized as safe (GRAS) for intended use or sanctioned by the United States Food and Drug Administration (FDA) Code of Federal Regulations or the U.S. Pharmacopoeia/National Formulary. The edible packaging materials and additives must be used in accordance with good manufacturing practice (GMP) (i.e., food grade, prepared and handled as a food ingredient) and within any limitations specified by the FDA (Krochta & De Mulder-Johnston 1997). The current classification and regulations of food ingredients can be found at FDA's Everything added to Food in the United States (EAFUS) database (**http://www.accessdata.fda.gov/scripts/fcn/fcnNavigation.cfm?rpt=eafusListing)**.

The ingredients of edible packaging must be declared on the label under regulation of the Federal Food, Drug, and Cosmetic Act (21 USC 343). Edible packaging made from common allergens (i.e., milk, eggs, peanuts, tree nuts, fish, shellfish, soy, wheat) must be clearly labeled to provide information to consumers with allergies or intolerances to particular food components according to Food Allergen Labeling and Consumer Protection Act of 2004.

Consumer Acceptance

The potential uses of edible materials are significantly affected by consumer acceptance issues, including sensory properties, safety, marketing, and cultural and religious restrictions regarding the use of new materials and applications. As food components, edible packaging materials must have neutral sensorial properties or be compatible with edible-packaged food in order to be undetected during consumption.

Consumers are becoming increasingly more aware of labels on what they eat. Although FDA requires food manufacturer to list ingredients and allergens on labels, the new edible film-forming materials or application can present problems for sensitive groups of consumers with allergies or cultural/religious restrictions. Such labeling regulation is only limited to the top eight allergens and is poorly enforced in fresh produce, bulk, institutional, or food-service applications. Other safety issues related to the potential microflora changes of packaged/coated food products with new edible packaging application must also be addressed.

Commercialization of edible packaging also depends on marketing factors including price, consumer reluctance to use new materials, and special attention to any special instructions required for opening, cooking, consuming the packaged/coated foods, or disposing of the packaging.

Feasibility of Commercialized Systems

At present, production of edible films is mainly at the laboratory scale and considered to be expensive compared with synthetic plastic films. Research on cost reduction and production in larger scales are necessary to promote the feasibility of commercialized edible packaging. Feasibility of commercialized systems depends on the complexity of the production process, size of investment

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for film-production or coating equipment, potential conflicts with conventional food packaging systems, and manufacturer resistance to the use of new materials (Han & Gennadios 2005).

Additionally, food manufacturers demand a long shelf life for products in interstate or international commerce. Edible packaging materials are themselves inherently susceptible to biodegradation, thus their protective functions are stable for shorter durations than conventional packaging. Therefore, the stability and safety of edible packaging under the intended storage/use conditions require investigation.

DISCLOSURE STATEMENT

The authors are not aware of any affiliations, memberships, funding, or financial holdings that might be perceived as affecting the objectivity of this review.

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Errata

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