JOURNAL OF Applied Polymer SCIENCE

Special Issue: Bio-based Packaging

Guest Editors: José M. Lagarón, Amparo López-Rubio, and María José Fabra
Institute of Agrochemistry and Food Technology of the Spanish Council for Scientific Research

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REVIEW

Active edible films: Current state and future trends

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ABSTRACT: Active edible films represent one of the current and future trends in the development of new polymers for selected applications, particularly food packaging. Some biopolymers show excellent performance as carriers for active compounds extracted from natural sources and are able to be released at a controlled rate to packaged food. In this review we aim to present, in a comprehensive way, the most recent advances and updates in this subject, where much research is currently ongoing and new studies are reported very often. This review focuses on innovative biopolymer matrices, their processing to obtain edible active films, and present and future applications. © 2015 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 42631.

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INTRODUCTION

The current increase in consumer demand for natural foods has forced companies and researchers to explore different ways to improve their market penetration by offering products with improvements in quality, freshness, and food safety.[1] One of the more fashionable trends consists of the development of innovative biopolymers obtained from agricultural commodities and/or food-waste products.[2] The use of biopolymers in multiple food-packaging applications has emerged as an alternative with regard to their film-forming properties to produce edible films as an environmentally friendly technology.[3]

Starches, cellulose derivatives, chitosan/chitin, gums, animal or plant-based proteins, and lipids offer one the possibility of obtaining edible films in fresh or processed food packaging to extend the food's shelf life.[4–10] These polymers offer additional advantages in their commercial use, such as biocompatibility, barrier properties to moisture and/or gases, nontoxicity, nonpolluting characteristics, mechanical integrity, and low cost.[11,12] In addition, edible films can act as carriers for antioxidant/antimicrobial additives to extend food's shelf life while maintaining their mechanical integrity and handling characteristics.[10,13] Antioxidant edible films can prevent food oxidation, the development of off-flavors, and nutritional losses, whereas antimicrobials can prevent spoilage from food-borne bacteria and organoleptic deterioration by microorganism proliferation.[14–16] The introduction of natural active additives to packaging materials provides advantages compared to the direct addition to food, such as the lower amount of active substances required, controlled release to food, and elimination of additional steps on processing.[17]

Edible films are obtained from food-grade suspensions and are usually molded as solid sheets onto inert surfaces. They are dried and put into contact with food as wrappings, pouches, capsules, bags, or casings through further processing.[18,19] However, sometimes the terms *films* and *coatings* are used interchangeably to note when a food surface is covered by relatively thin layers. Some authors have distinguished both terms by the notion that coatings are applied directly onto the food surface, whereas films are stand-alone wrapping materials.[18–21] For this reason, the current state in edible active matrices is summarized in this review, with emphasis on recent trends in protein-based and polysaccharide-based edible films. These matrices and processing methods used to obtain edible films and their role in active packaging are reviewed, whereas their industrial effectiveness in different applications is also discussed.

EDIBLE ACTIVE MATRICES

The use of edible films based on natural polymers and food-grade additives has been constantly increasing in the last few years. These films can be produced from a variety of products, such as polysaccharides, proteins, lipids, and resins, with or without the addition of other components (e.g., plasticizers and surfactants). Figure 1 summarizes a general scheme for the classification of edible films.[22,23]

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Marina Ramos graduated with a degree in chemistry from the University of Alicante (Spain). She is a Ph.D. student with the Polymer and Nanomaterials Analysis Research Group at the same university. Her current research is focused on the development of antioxidant and antimicrobial active packaging systems based on conventional polymers and biopolymers.

Nuria Burgos graduated with a degree in chemical engineering in 2002 and obtained her Ph.D. in chemistry in 2013 from the University of Alicante (Spain). She is working as a senior researcher with the Polymer and Nanomaterials Analysis Research Group (Department of Analytical Chemistry, Nutrition and Food Sciences, University of Alicante). Her main research areas are focused on the development of new sustainable composites based on biopolymers and natural compounds (plasticizers, nanofillers, and active additives) for their use as food-packaging materials. She has published several research articles in journals relating to food and polymer technologies, and she has presented their results at different international conferences.

María del Carmen Garrigós received her Ph.D. in chemistry from the University of Alicante in 2003. She has been an assistant professor in analytical chemistry at the University of Alicante since 2008. She is the author of 32 research articles published in journals in analytical chemistry, food technology, and polymer science. Her main research areas include the chemical modification of biopolymers, natural additives for active packaging, TPUs obtained from vegetable oils, and valorization of agrofood residues.

Alfonso Jiménez received his Ph.D. in chemistry in 1996. He has been a full professor in analytical chemistry and food science at the University of Alicante since 2001. He is also head of the Polymer and Nanomaterials Analysis Research Group. Jiménez is also the editor of 15 books on sustainable polymers and the author of 103 research articles published in journals on analytical chemistry, food technology, and polymer science. His main research areas are the chemical modification of biopolymers, natural additives for active packaging, TPUs obtained from vegetable oils, and valorization of agrofood residues.

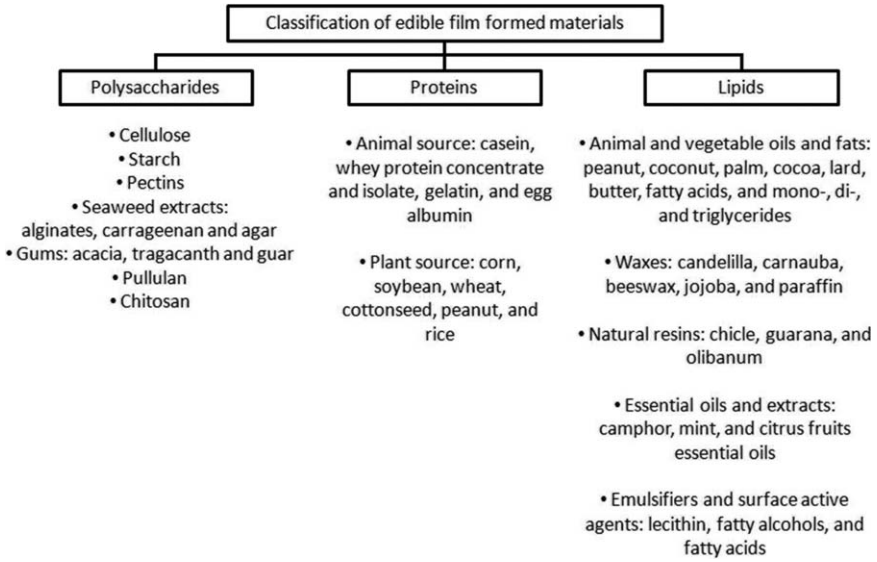

Figure 1. Classification of edible films according to their structural material.[23,28]

Polysaccharide-based films usually show poor moisture barrier properties but selective permeability to O_2 and CO_2 and resistance to oils.[24] These films can be based on cellulose, starch (native and modified), pectins, seaweed extracts (alginates, carrageenan, agar), gums (acacia, tragacanth, guar), pullulan, and chitosan. These compounds give hardness, crispness, compactness, viscosity, adhesiveness, and gel-forming ability to films.[25] Marine organisms, such as seaweeds, bacteria, and microalgae, have been considered other important sources of polysaccharide-based biomaterials.[26,27]

Film-forming proteins provide mechanical stability and can be derived from animals (casein, whey protein, gelatin, and egg albumin) or plant sources (corn, soybean, wheat, cottonseed, peanut, and rice). Plasticizers are added to improve the flexibility of the protein network, whereas the water permeability can be overcome by the addition of hydrophobic materials, such as beeswax or oils, to modify the crystallinity, hydrophobicity, surface charge, and molecular size.[24] Protein-based films exhibit poor water resistance, but they are superior to polysaccharides in their ability to form films with good mechanical and barrier properties.

A wide range of hydrophobic compounds has been used to produce edible films, including animal and vegetable oils and fats (peanut, coconut, palm, cocoa, lard, butter, fatty acids, and monoglycerides, diglycerides, and triglycerides), waxes (candelilla, carnauba, beeswax, jojoba, and paraffin), natural resins (chicle, guarana, and olibanum), essential oils (EOs), and extracts (camphor, mint, and EOs). Lipid-based edible films are used to reduce the water vapor permeability.[24]

The incorporation of active chemicals extracted from industrial wastes into edible films is a trending topic in materials research with an increasing number of results.[29] It was found that edible films may serve as carriers of active compounds, such as antimicrobials, antioxidants, and texture enhancers,[13] and different ways to obtain them have been reported (Table I). Some examples are discussed later.

New Trends in Polysaccharide-Based Edible Films

Edible films produced from polysaccharides (cellulose, starch, pectins, seaweeds, gums, chitosan, and pullulan) have been widely used in the food industry in the last few years, whereas lignocellulosic materials have been recently proven as suitable materials for edible film production. Slavutsky and Bertuzzi[29] reported the successful production of starch films reinforced with cellulose nanocrystals obtained from sugarcane bagasse. Translucent and transparent films were prepared with hemicellulose fractions from the leaves of *Pinus densiflora* by Shimokawa *et al.*[30] These authors obtained materials with properties similar to those of xylan and with high potential as edible films. Crystalline cellulose nanofibrils isolated from cotton linter by acid hydrolysis were used to prepare composite films with clear enhancements in the optical and mechanical properties, water vapor barrier properties, and thermal stability.[31] Composite alginate films were manufactured from alginate–carbohydrate solutions containing 5 wt % alginate and 0.25 wt % pectin, carrageenan (κ or ι), potato starch (modified or unmodified), gellan gum, or cellulose (cellulose extracted from soybean chaff or commercial cellulose).[32] All of those carbohydrates were able to form composite films with the alginate matrix. However, the cellulose extracted from soybean chaff could produce alginate-based composite films/casings with mechanical strengths similar to those produced from commercial microcrystalline cellulose.

Pectin extracted from different vegetal sources, such as apple, carrot, and hibiscus, can be used to prepare active edible films with antioxidant and antimicrobial performances, obtained from natural additives, such as carvacrol and cinnamaldehyde[33] or lime EO.[34] In fact, lime is the most important citric fruit used for the extraction of EOs, and it is also a good source of pectin with antioxidant activity. Sánchez Aldana *et al.*[34] studied the antibacterial activity against common food-borne bacteria of edible films based on extracts from Mexican lime pectin, which increased their value by providing an antibacterial effect.

Table I. New Trends in Edible Film Matrices

Edible matrix	Industry waste	Edible film	Reference
Polysaccharides	Sugarcane bagasse	Starch (4%), glycerol (20% dry weight), water, and appropriate amount of cellulose nanocrystals obtained from sugarcane bagasse (3% dry weight)	29
	P. densiflora leaves	Hemicellulose fractions of *P. densiflora* leaves with 1% w/w polysaccharide) lecithin	30
	Cotton linter pulp	Crystalline cellulose nanofibrils from cotton linter pulp to reinforce sodium carboxymethyl cellulose films (2% w/v) and 0.9 g of glycerol (30 wt %) to 150 mL of distilled water	31
	Soybean chaff	Composite alginate films obtained from alginate–carbohydrate solutions containing 5 wt % alginate and 0.25 wt % cellulose extracted from soybean chaff	32
	Apple, carrot, and hibiscus	Apple, carrot, and hibiscus-based pectin edible films	33
	Lime bagasse and lime pomace pectic extracts	Lime bagasse pectic extract and lime pomace pectic extract at 0.70, 0.85, and 1.00% pectin equivalents with Mexican lime EO and 0.70 wt % glycerol plasticizer	34
	Pectin from citrus	Microparticles and films containing sunflower oil produced by ionic gelation with a 1:1 alginate–pectin mixture and electrostatically coated with whey and egg white proteins	35
	M. stellatus seaweeds	Hybrid carrageenan extracted from *M. stellatus* seaweeds	36
	Pyropia columbina red algae	Carrageenan/porphyran-based films obtained from a *P. columbina* aqueous fraction formed by casting from aqueous dispersions with different levels of glycerol	37
	M. stellatus seaweed	Edible active films from different *M. stellatus* crude aqueous extracts	38
	Porphyra columbina seaweed	Antioxidant phycobiliprotein/phycocolloid-based films obtained from mixtures of two aqueous fractions extracted from *P. columbina* red seaweed	39
	Brown seaweeds *Laminaria digitata* and *Ascophyllum nodosum*	Film-forming carbohydrate-rich extracts from the brown seaweeds *L. digitata* and *A. nodosum* obtained with Na_2CO_3 or NaOH at different temperatures and with different acid pretreatments (H_2SO_4 and HCl)	40
	Wheat bran AXs	1.5% w/v chitosan in 1% v/v lactic acid under agitation for 1 h at 25°C. Glycerol (0.5 wt %) and Tween 80 (0.1 wt %) were added at 60°C under agitation for 30 min. Then, AXs (0.2%) was added, and the solution was stirred for 72 h.	24
	Chitosan and protein concentrate from shrimp waste	Chitosan solution (2% w/w) dissolved in a 0.15*M* lactic acid solution (pH 3.2) and sonicated	41
	Chitosan and potato and cassava starches	Starch and chitosan films obtained by the variation of the starch source (potato and cassava starches), starch concentration (0.5 and 1.0 wt %), and type of plasticizer (glucose and glycerol)	42
	Marine industry byproducts: chitosan and fish gelatin	Chitosan and fish gelatin (1:1 w/w), entrapping natural antioxidants [ferulic acid, quercetin, and tyrosol (~50 mg/g)], used to prepare edible active films by casting	43, 44
	Chitosan and zein	Composite edible films fabricated with zein and chitosan and supplemented with phenolic compounds (ferulic acid or gallic acid) and dicarboxylic acids (adipic acid or succinic acid)	45

Table I. *Continued*

Edible matrix	Industry waste	Edible film	Reference
	Cashew tree gum	Starch–cashew tree gum nanocomposite film; sage seed gum edible films with two different plasticizers (glycerol and sorbitol: 20, 40, 60, 80, and 100% w/w)	46
	Locust bean gum	0.4 and 0.6% w/v κ-carrageenan and locust bean gum suspended in distilled water under agitation for 1 h at 25°C with 0.3% w/v glycerol (87% v/v) in solution and homogenized at 80°C for 30 min	47
	Basil seed gum	Basil seed gum and different plasticizer concentrations added to deionized water and heated to 80°C under mild stirring	48, 49
	Brea gum	Brea gum (10% w/v), glycerol (25% w/w Brea gum), water, and montmorillonite (5% w/w Brea gum)	50, 51
	C. myxa gum	Glycerol, sorbitol, PEG 200, and PEG 400 in the range 0.1–0.3 g/g of dry polymer (weight basis) dried at 40°C for 48 h	52
	Turmeric dye solvent residue	Turmeric flour films and sorbitol (30 g of sorbitol/100 g of flour)	53
	Cassava starches	Cassava starch (5% w/w) and soy protein concentrate with glycerol (20%)	54
	Dark cush-cush yam and cassava starch	Edible films prepared from a film-forming solution made by the mixture of 2% w/v starch and 1.9% w/v glycerol in distilled water	55, 56
	Waxy and normal corn starches	2% w/v of either native or modified starch, 1.9% w/v glycerol, and 500 mL of distilled water	57
	Wheat starch	Wheat starch and whey-protein isolates (100:0, 75:25, 50:50, 25:75, and 0:100 ratios) with glycerol used as a plasticizer at 50 wt %	58
	Wheat starch	Lamination of a wheat starch solution and rapeseed oil	59
	Cassava starch, carnauba wax, and stearic acid	Formulations containing cassava starch, glycerol, carnauba wax, and stearic acid based edible coatings/films	60
	Perilla seed oil residue	Perilla seed meal protein and different amounts of red algae 1.5% w/v of chitosan	61
	Canola meal	Cast canola protein isolate edible films (5.0 and 7.5%) and glycerol (30–50%)	62
	Chicken feet	Chicken feet protein films with a 3:2 w/w ratio of glycerol to sorbitol	63
	Cold-water fish skin	4 g of gelatin in 100 mL of distilled water with glycerol (0.3 g/g of gelatin)	64
	Surimi and skin gelatin from silver carp	Surimi solution with a gelatin solution (10:0, 8:2, 6:4, 5:5, 4:6, 2:8, and 0:10), 4 g of gelatin in 100 mL of distilled water with glycerol (0.3 g/g of gelatin)	65
Proteins	Zein particles and fatty acid oleic acid	1.4 g of zein dissolved in 8.1 mL of ethanol (96%), catechin, and lysozyme	66

Citrus pectin was used to obtain one emulsion based on alginate–pectin–sunflower oil by spraying over a 2% w/w calcium chloride solution at room temperature with a double-fluid atomizer. Then, films were fabricated in two different ways by crosslinking and hardening, such as in the production of microparticles.[35]

Carrageenan describes a family of polysaccharides produced by some red seaweed (*Rhodophyta*) as their main cell-wall material. Paula *et al.*[26] studied the effects of the relative proportions of κ-carrageenan, ι-carrageenan, and alginate on the physical properties of glycerol-plasticized edible films. They reported that κ-carrageenan was the component that improved the moisture

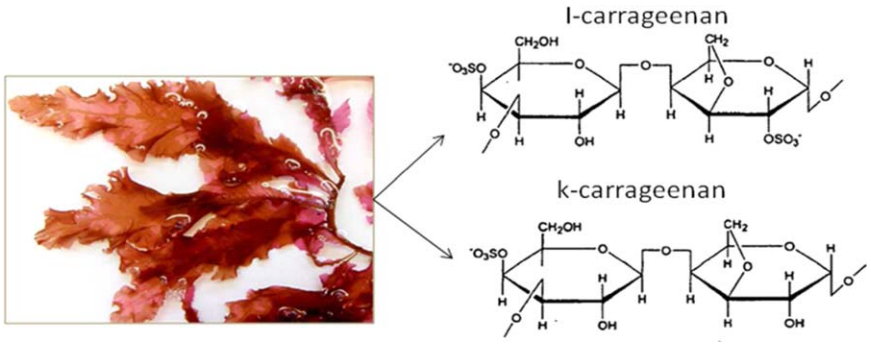

Figure 2. Structures of κ-carrageenan and ι-carrageenan extracted from the red algae *Hypnea musciformis* and *Sagittaria filiformis*. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

barrier and overall tensile properties, whereas ι-carrageenan was the component that impaired them (Figure 2).

In this context, different authors have reported the use of hybrid carrageenan, extracted from *Mastocarpus stellatus* seaweeds, as an alternative to commercial κ-carrageenan in new edible film formulations. These hybrid materials have shown promising possibilities for the production of edible coatings and biodegradable films.[36] Other seaweed-based edible films obtained from several extracts have been recently studied[37–40] (Table I).

Chitosan is produced by the alkaline deacetylation of chitin, the material that comprises the exoskeleton of crustaceans and mollusks. Costa *et al.*[24] evaluated the properties of chitosan-based films with different fractions of arabinoxylans (AXs) obtained through three different processes. In this sense, AXs could be obtained from the pretreatment of low-cost agricultural residues, such as cereal crops and wheat bran. The incorporation of 0.2 wt % AXs into the chitosan matrix allowed the production of films with prebiotic and/or dietary fiber properties and potential health benefits. Arancibia *et al.*[41] used mildly processed chitosan and a protein concentrate obtained from shrimp (*Litopenaeus vannamei*) for the development of active edible films with antioxidant and antimicrobial properties. Different chitosan-based edible films in combination with several biopolymers, such as starch,[42] fish gelatin,[43,44] and proteins obtained from zein,[45] have been recently reported.

Polysaccharide gums have been recently studied in their possibilities as edible film promoters because of their sustainable, biodegradable, and biocompatible characteristics. The term *gum* refers to polysaccharides that form gels, make viscous solutions, or stabilize emulsion systems. Several plant gum exudates obtained from different agricultural wastes have been used recently to obtain edible films. In this sense, Razavi *et al.*[9] reported that the gum extracted from sage (*Salvia macrosiphon*) seeds could be used to obtain edible films with increased thickness, ductility, moisture content, and uptake and decreased surface hydrophilicity. Films with carvacrol exhibited higher antimicrobial activity than those with cinnamaldehyde, in particular in apple-based films. Pinto *et al.*[46] developed films from starch and cashew tree gum, a water-soluble heteropolysaccharide, with montmorillonite, and they tested their application as a coating to increase the stability of cashew nut kernels, tensile properties, and water vapor barrier properties. Smart thermosensitive poly(nisopropyl acrylamide) nanohydrogels with or without acrylic acid incorporated into polysaccharide-based films obtained from κ-carrageenan and locust bean gum were recently reported by Fuciños *et al.*[47] They showed the possibilities of these biocomposites to transport natamycin and their controlled release as a response to environmental triggers. These results were promising because this system makes it possible to reduce the natamycin concentrations in food products while improving their antifungal effect. The seeds of *Ocimum basilicum L.*, also known as basil, contain a considerable amount of gum composed of two major fractions of polysaccharides (43% glucomannan and 24.3% xylan) with outstanding functional properties in developed edible films.[48,49] Brea gum is the exudate obtained from the Brea tree (*Cercidium praecox*), and it has been used to study the effect of montmorillonite incorporation into Brea gum-based films through thermodynamic and phenomenological analysis.[50,51] The effect of the addition of plasticizers into edible films obtained from *Cordia myxa* gums with different effects has been also reported.[52]

Finally, starch is the most important polysaccharide used in the formulation of biodegradable edible films. Different starch formulations may lead to the formation of edible films with particular characteristics and properties. For example, it was reported that *Curcuma longa L.*, commonly known as turmeric, generates a residue that consists predominantly of starch and fibers that may present residual levels of curcuminoids with antioxidant properties.[53] It was concluded that turmeric films could act as active packaging materials because of the presence of curcuminoids with antioxidant characteristics. Among starches, cassava, corn, and wheat starches have been recently proposed for the formulation of edible films thanks to their availability and relatively low price. Starch is normally used in mixtures with different biomaterials, such as soybean protein concentrates,[54] native and modified cush-cush yam and cassava starches,[55,56] wax and normal starches,[57] wheat starch and whey-protein isolates,[58] wheat starch solution and rapeseed oil,[59] and cassava starch, glycerol, carnauba wax and stearic acid.[60]

New Trends in Protein-Based Edible Films

Several agricultural wastes have been reported as new sources of proteins for use in edible films. For example, perilla seed, consisting of 51% fat and 17% protein, with this value increased

Table II. Recent Trends in Active Edible Film Production (Wet Method)

Biopolymer	Solvents	Additives	Homogenization conditions	Drying	Reference
Chitosan	Acetic acid (1%)	Glycerol, Tween 80, and clove oil	Chitosan solution stirred (at 40°C for 2 h), mixing with additives by Ultra Turrax, ultrasound for 30 min to remove bubbles	42°C for 15 h in a forced-air incubator	78
Chitosan	Acetic acid (1%)	Rosemary EO and Tween 80	Chitosan solution stirred (at 90°C for 20 min); cooling; mixing with Tween 80 (40°C, 30 min) and EO (Ultra Turrax, 4000 rpm, 2 min); cooling; degassing *in vacuo* for 5 min	72 h at 25°C on a Teflon-coated steel plate	93
Methyl cellulose	Water	PEG, clove and oregano EOs, and Tween 80	Mixing EOs with water and Tween (at 1000 rpm for 5 min), ultrasonication of coarse emulsions (20 kHz, 400 W, 10 min), homogenizing with methyl cellulose and PEG (30 min at 6 rpm), rested for 2 h	Overnight at 25°C on a glass plate	85
Whey-protein isolate	Water	Glycerol and cinnamon, cumin, and thyme EOs	pH 8 (NaOH), stirring (at 90°C for 30 min), cooling, degassing *in vacuo* (for 30 min)	Overnight (at 35°C and 45% RH) on a glass dish	91
Cassava starch	Water	Glycerol and cellulose fibers	Hydration of fibers for 24 h; stirring for 10 min at 14,000 rpm; mixing with all components by stirring (at 71°C for 5 min at 90 rpm); tape casting at a spreading speed of 40 cm/min	Heat conduction (60°C) for 5 h in PMMA protected by a silicone-coated PET film	69
WPC	Water	Glycerol, cinnamon EO, and Tween 80	WPC solution (90°C, 30 min), mixing with glycerol and Tween 80 for 30 min, homogenizing with CEO (at 25°C for 2 min at 7000 rpm), keeping overnight at 4°C	72 h at 25°C on a glass dish	74
ISP	Water	Glycerol, oregano, and thyme EOs	pH 10 (NaOH) of ISP solution with glycerol, mixing with EO (at 90°C for 30 min), cooling to 40°C	Oven (30°C, 72 h) on a PS Petri dish	94
Fish-skin gelatin	Water	Glycerol and natural extracts	Gelatin solution with glycerol and extracts, mixing at 45°C for 30 min, ultrasound for 15 min	Oven (at 22°C for 36 h) on a methacrylate dish	79
Sodium alginate	Water	Glycerol and EO surfactant	Sodium alginate solution (a t100°C for 1 h), cooling, mixing with glycerol and EO (vortex)	Ambient conditions on a glass dish	80
NaCas	Water	Glycerol and natural EOB	NaCas solution with glycerol stirred for 2 h, mixing with EOBs (2 h), homogenizing at 700 bar for 5 min, keeping overnight at 4°C	37°C for 24 h on a Petri dish	75
SSPS	Water	Glycerol, EOs, and Tween 80	SPSS solution (at 300 rpm for 40 min on a hot plate), mixing with glycerol (for 15 min at 82°C), homogenizing in a rotor–stator (at 20,000 rpm for 3 min)	18 h at 25°C on a PS Petri dish	76
κ-Carrageenan	Water	Glycerol, EOs, and Tween 80	κ-Carrageenan solution (for 15 min at 82°C), mixing with glycerol (for 25 min at 82°C),	30 h at 30°C on a glass dish	71

Table II. *Continued*

Biopolymer	Solvents	Additives	Homogenization conditions	Drying	Reference
			homogenizing in a rotor–stator (at 13,500 rpm for 3 min at 80°C); degassing at 65°C		
Chitosan	Acetic acid (1%)	EOs and Tween 80	Chitosan solution (at 250 rpm and 45°C), stirring overnight at room conditions, homogenizing in a dual-range mixer (at 2500 rpm for 4 min), vacuum	48 h at 22°C and 30% RH on a PS dish	72
Cassava starch	WaterEthanol	Glycerol, clove, and cinnamon EOs; emulsifier; and clay nanoparticles	Clay NPs suspended in water (for 1 h at 500 rpm), resting for 24 h and blending with cassava starch solution; glycerol, EO, and emulsifier (200 rpm); both mixtures being homogenized in a domestic microwave oven (until 69°C); cooling and dilution with ethanol	Forced-air oven at 35°C for 18–24 h on a Teflon plate	84
Fish protein	Water	Glycerol, clove, garlic, and oregano EOs	Protein concentrate and glycerol (stirred 30 min), emulsifying with EO in a homogenizer (at 13,500 rpm for 2 min), vacuum (20 min)	Ventilated drying chamber (at 30°C and 50% RH for 20 h)	83
Fish gelatin	Water	Glycerol and cinnamon EO nanoliposomes	Gelatin solution, glycerol, and EO stirred 30 min at 45°C; ultrasound	22°C at 50% RH for 24–48 h on a methacrylate dish	86
Chitosan	Lactic acid (1%)	Glycerol, Tween 80, and AX/AXOS	Chitosan solution stirring (for 1 h at 25°C), mixing with glycerol and Tween (at 60°C for 30 min); mixing with AX/AXOS for 72 h	30°C for 60 h on a PS plate	95
Fish gelatin	Water	Glycerol–sorbitol (1:1), lignin	Gelatin solution at 40°C, mixing with plasticizers and lignin at 40°C for 15 min at pH 11	Forced-air oven at 45°C for 15 h	88
SSG	Water	Glycerol, sorbitol, and PEG 400	Plasticizers in water (at 150 rpm and 80°C), mixing with SSG (at 1200 rpm and 80°C for 10 min), homogenizing (at 3200 rpm for 1 min), mixing on a rotating roller–mixer (for 24 h at 25°C)	23°C at 53% RH on a polyethylene dish	73
Turmeric flour	Water	Sorbitol	Turmeric in water (30 min of stirring), adjusted different pHs (6.5–9.5) and temperatures (78–92°C) for 4 h, homogenization cycles (at 12,000 rpm for 2 min) every hour, mixing with sorbitol (20 min of heating), sonication (20 min)	Forced-air oven (at 35°C for 7 h) on an acrylic plate	53

AXOS, arabinoxylan-oligosaccharide; EOB, emulsified oil body; ISP, isolated soy protein; PET, poly(ethylene terephthalate); PMMA, poly(methyl methacrylate); SSG, sage seed gum; SSPS, soybean polysaccharide.

up to approximately 40% after oil extraction. Perilla seed is currently used as an animal feed or fertilizer and would increase the added value in edible films. Song *et al.*[61] reported that perilla seed protein combined with 3 wt % red algae resulted in suitable mechanical properties. Among the EOs incorporated into the composite films, clove oil exhibited the highest level of antimicrobial performance. Canola (*Brassicaceae* species) proteins showed functional attributes, but the oil is currently used for cooking and biodiesel synthesis with no further added-value applications. Once the oil is pressed, the remaining meal (high in proteins and fibers) is typically used for feeding animals. Canola proteins can be extracted from the meal as a byproduct for food and nonfood applications, such as edible films.[62] Other proteins can be extracted from animal sources. For instance, Lee *et al.*[63] prepared antimicrobial and antioxidant films using proteins extracted from chicken feet and proposed their use in sliced cheddar cheese packaging.

Among all of the protein sources, gelatin has been the most extensively studied for its high film-forming capacity and applications as outer coatings to protect food against light and oxygen. Fish gelatin has gained interest in recent years as an alternative to bovine and porcine gelatin because of social and health reasons, such as the bovine spongiform encephalopathy crisis. Furthermore, fish skin, a major byproduct of the fish-processing industry, could provide a valuable source of gelatin. As consequence, the elaboration of edible films from fish gelatin has been recently studied by several authors.[64,65]

Zein has been used as a supporting carrier of antimicrobial enzymes, including lysozyme, lactoperoxidase, and glucose oxidase, bacteriocins, and natural antimicrobial and antioxidants, such as plant phenolics and EOs. Lysozyme–zein films showed high performance in the inhibition of Gram-positive pathogenic bacteria, such as *Listeria monocytogenes,* which might cause deadly infections. Zein-based edible films have recently gained interest, because (1) zein is a major coproduct in bioethanol production, (2) it is one of the rare proteins soluble in organic solvents, and (3) it provides an effective delivery system for lysozyme. In this context, Arcan and Yemenicioğlu[66] developed films by forming emulsions of zein with oleic acid in the presence of lecithin. Microspheres within these films were observed, and these increased the barrier against diffusion of the encapsulated lysozyme.

ACTIVE EDIBLE FILM PROCESSING

Traditionally, the methods used for the production of edible films have been divided in two main groups: wet and dry processes. The wet process needs solvents for the solution and dispersion of the polymer onto a flat surface; this is followed by drying in controlled conditions for the removal of the solvent and the formation of the film. It is a high-energy-consuming procedure, adequate for laboratories but not for the industrial scale-up. The production of edible films by dry methods includes extrusion, injection, blow-molding, and heat-pressing processes as those most commonly used.[12,67] The combination of efficiency and high productivity provided by these thermal processes have induced an increase in their application for active edible film manufacturing.[68] However, the high temperatures used in the dry processes could affect the presence and concentration of some active compounds on the films,[12] whereas the addition of plasticizers is sometimes necessary to decrease the glass-transition temperature for polymer matrices. The method selected for the production of active edible films could affect and modify the final properties of the material.

Wet Processing

Casting is a simple method for the production of edible films, but it is a batch procedure used on a very small scale. Nevertheless, a continuous casting method (knife coating or tape casting) can be used on the industrial scale[3] because the film-forming suspension is prepared on continuous carrier tapes with effective control of the thickness. The formed film is dried by heat conduction, convection, or radiation in short times. De Moraes *et al.*[69] showed that tape casting is a suitable technology for scaling up the production of cassava-starch-based edible films.

In general, wet processes could be divided into four steps: dispersion or gelatinization, homogenization (in the case of emulsions or mixtures), casting, and drying. Several factors have influence and should be optimized depending on the polymer matrix: the solvent, plasticizer, and/or other additive contents, the method for granule disruption, the temperature, and the time.[70] If different hydrocolloids or other nonmiscible components are added, a mixing step is also required to obtain stable emulsions and homogeneous film-forming solutions. Homogenization is currently performed with new homogenizer devices, such as a rotor–stator or Brabender viscographs, to induce high pressures that can enhance the disruption and interactions between all components.[53,71–76] Table II summarizes the recent trends in active edible film production according to the wet method.

Because the final product should be edible and biodegradable, only water and ethanol or their combination, are suitable solvents.[67,77] Furthermore, all of the components of the film-forming solutions should be homogeneously dispersed in solvents to produce edible films without phase separation. Emulsifiers can be added to prevent this situation, even when incompatible compounds are used.[72,74,76,78] The addition of food-grade plasticizers rich in hydroxyl groups, such as glycerol, sorbitol, or poly(ethylene glycol) (PEG), at concentrations between 15 and 30 wt % reduce the polymer rigidity and glass-transition temperature and enhance the distribution of film-forming solutions.

EOs extracted from plants have been used in recent years as active compounds for edible film manufacturing (Table II). Different strategies for incorporating active components into water-soluble polymers, including direct addition to the film-forming solution followed by a casting method,[74,79–84] nanoemulsions through ultrasonication,[85] and encapsulation into nanoliposomes through the sonication of their aqueous dispersions,[86,87] have been reported. Otoni *et al.*[85] prepared coarse emulsions (diameter = 1.3–1.9 μm) and nanoemulsions (diameter = 180–250 nm) of clove EO through low-speed mixing and ultrasonication, respectively, with emulsifiers for

Table III. Recent Trends in Blending Methods for the Production of Active Edible Films

Biopolymer	Solvents	Additives	Homogenization	Drying	Reference
Corn starch/NaCas	Water	Glycerol, orange EO, and limonene encapsulated in nanoliposomes	Starch stirred in water (at 95°C for 30 min); mixing with NaCas solution, glycerol, and nanoliposome solution (for 1 h at 300 rpm)	For 48 h at 20°C at 45% RH in a PET Petri dish	87
Chitosan/protein concentrate	Lactic acid (1%)		Chitosan and protein solutions, direct mixing	In an oven at 45°C for 12 h on a methacrylate plate	96
Chitosan/gelatin	Water/acetic acid (1%)	Glycerol	Blend stirred at 40°C for 30 min	At 25°C for 12 h (air blown) and at 25°C and 50% RH for 48 h on a silicone plate	97
Chitosan/methyl cellulose	Water, acetic acid (0.25%), and ethanol	Resveratrol	Methyl cellulose in water, chitosan stirred with acetic acid (0.25%) overnight at 25°C, homogenizing with resveratrol in ethanol in a rotor–stator (at 13,500 rpm for 4 min), vacuum (10 min)	Natural convection (48 h at 25°C and 60% RH) in darkness	92
CFP/gelatin	Water	Clove oil, cinnamaldehyde, and Tween 20	CFP solution with gelatin and sorbitol stirred for 1 h, ultrasonicating for 8 min, hot-water bath (at 75°C for 30 min), all components stirred for 20 min at 40°C	For 24 h at 25°C on a Teflon-coated glass plate	99
Fish gelatin/chitosan	Water/acetic acid (1%)	Glycerol	Gelatin in water stirred for 30 min at 25°C and for 30 min at 45°C, chitosan and acetic acid (1%) stirred overnight at 25°C, both solutions mixed at 45°C for 30 min, glycerol added (at 45°C for 15 min), vacuum (for 15 min)	For 72 h at 23–25°C on a PS dish	90

PET, poly(ethylene terephthalate); RH, relative humidity.

Table IV. Recent Trends in the Application of Active Edible Films in Food Packaging

Edible matrix	Active compound	Application	Reference
Carregeenan	Lemon EO	Trout	111
Whey protein	Oregano and clove EOs	Chicken	112
Sunflower protein	Clove EO	Fish patties	82
Argentine anchovy protein	Sorbic and benzoic acids	Meat	113
Chitosan	Thyme EO	Cured ham	114
Whey protein	Cinnamon, cumin, and thyme EOs	Beef	91
Chitosan	Basil and thymus EOs	Pork	115
Agar	Green tea extract and probiotic strains	Fish	116
Gelatin fish	Laurel leaf EO	Trout	117
Methyl cellulose	Nanoemulsions of clove and oregano EOs	Sliced bread	85
Whey protein	Oregano and clove EOs	Poultry	118
Barley bran protein/gelatin	Grapefruit seed extract	Salmon	119
Soy-based protein	Oregano or thyme EO	Ground beef patties	120

homogenization in water. The incorporation of these emulsions into methyl cellulose matrices plasticized with PEG showed that the droplet size reduction provided higher antimicrobial properties. In addition, lower EO contents could be used if they were encapsulated to retain the same antimicrobial efficiency. However, some negative effects on the mechanical properties were observed with the addition of EO emulsions. Some increase in the antimicrobial stability with a decrease in the cinnamon EO release rate was observed for gelatin films with nanoliposomes.[86]

Casting is widely used for the direct incorporation of EOs into film-forming solutions with low temperatures used for the homogenization and drying steps. For example, cinnamon EO increased the antimicrobial activity of whey-protein concentrate (WPC) as well as the water vapor permeability and water solubility.[74] However, the addition of emulsifiers is necessary to help EO distribution in the film-forming solutions.[71,76,78,80] Other authors incorporated lignin to gelatin matrices by casting, resulting in films with excellent antioxidant and light barrier properties to prevent the ultraviolet-induced lipid oxidation in certain food applications.[88] They did not use emulsifiers, showing some microphase separation between gelatin and lignin. Maniglia *et al.*[53] developed turmeric flour films with antioxidant activity by casting, using the turmeric dye solvent extraction residue. However, air bubbles are frequently observed as a consequence of the homogenization step, and vacuum[89–91] or ultrasound[78,79] devices are used to remove them, avoiding the presence of microholes in the film structures.

After homogenization, film-forming dispersions are cast on leveled dishes and allowed to dry under controlled conditions. The high difference in the surface energies between the film and the dish surface is an important factor for easy peeling after casting.[67] Therefore, depending on the polymer, different materials, such as polystyrene (PS),[76,82] polyethylene,[73] polycarbonate,[81] methacrylate,[79,86] and glass,[71,74,91] have been used to obtain films by casting. Other surfaces, such as stainless steel, silicone,[69] and polytetrafluoroethylene (Teflon),[92,93] have also been considered for their high inertness.

A great variation in drying temperatures and times has been reported, with times varying between 5 and 72 h and temperatures varying between 20 and 45°C; the values are significantly lower for edible films with active volatile compounds to prevent their evaporation (Table II).

Blending

The blending of different macromolecules by either a direct method or associated with codrying processes leads to edible films with good control of their final properties. The most recent studies on blending different matrices to yield edible films have focused on the combination of polysaccharides with proteins, taking advantage of their synergistic effects.[87,89,90,96,97] Direct blending consists of an initial preparation stage of individual polymer solutions with subsequent homogenization and casting. It shows some advantages because the codrying method requires strict conditions, such as a narrow concentration range and good compatibility.[89] Table III summarizes the production of active edible films by blending.

Chitosan is one of the most used biopolymers in the production of active edible films.[98] The direct mixing of chitosan with protein concentrate solutions significantly changes the film structure and leads to the reinforcement of the chitosan matrices; this improves their barrier, antimicrobial, and antioxidant properties.[96] Edible films of chitosan with gelatin[90,97] or methyl cellulose[92] have been developed to improve the mechanical and barrier properties and to obtain antimicrobial activity against Gram-positive bacteria. In addition, chitosan/gelatin films have exhibited antioxidant activity as monitored by β-carotene bleaching and 2,2-diphenyl-1-picrylhydrazyl (DPPH) radical scavenging.[97]

Composite edible films formed by the blending of different polysaccharides[92,100,101] or proteins[99] have been recently reported. For example, an active compound (resveratrol) was efficiently incorporated into chitosan and methyl cellulose blends; it provided antioxidant activity with physical changes that did not affect negatively their handling and appearance.[92] The incorporation of clove EO into gelatin/chicken feather protein (CFP) blended films improved their antimicrobial and

antioxidant properties while the mechanical behavior was not affected.[99]

Spraying is a wet-processing method mainly used for coating production, in which a film-forming solution is sprayed onto the polymer surface and droplets are cast and dried. The solvent mainly evaporates after it leaves the nozzle of the sprayer, and this allows for short drying times.[3] Recently, Jindal *et al.*[102] used this method for the production of pectin–chitosan crosslinked films.

Dry Processing

It is known that materials with thermoplastic behavior can be processed into films by the application of different thermal/mechanical processing techniques. Therefore, it is essential to study the rheological properties and the effect of additives (plasticizers, emulsifiers, active agents) on the thermoplastic behavior of film-forming materials to select adequate processing parameters.

The most common dry-processing methods for edible films include extrusion, injection, blow molding, and heat pressing. Recent technical developments, in particular in twin-screw extrusion, have increased the application of this technique for edible film production,[103] where one or two rotating screws induce pressure and high temperature to disrupt the polymer granules and mix the film components. This technique shows some advantages, including the absence of solvents, the possibility of handling high-viscosity polymers and multiple injections, a broad range of processing conditions (0–500 atm and 70–500°C), and the control of the residence time and the mixing degree.[103] Other processing techniques, such as injection molding, film-blown die,[104–106] and thermopressing,[68] are combined with extrusion to produce the final edible films.

Belyamani *et al.*[106] reported the production of sodium caseinate (NaCas) films on a large scale with glycerol as a plasticizer and twin-screw extrusion to obtain homogeneous thermoplastic pellets; these were further processed into transparent films with a classical film-blowing machine. These films showed mechanical properties similar to those obtained by solution casting, and they were highly sensitive to glycerol and environmental moisture contents. Colak *et al.*[105] prepared a mixture of NaCas containing lysozyme (1 wt %) and converted in pellets by extrusion (between 65 and 100°C) with the addition of glycerol. These pellets were further blown by an industrial-like blown-film extruder to obtain thermoplastic antimicrobial films. The incorporation of plasticized chitosan to thermoplastic starch in a twin-screw extruder improved the subsequent blown-film processability and properties (mechanical, thermal stability, water absorption, and surface stickiness), although it caused some changes in the color and transparency. The authors observed that the lysozyme stability was mainly dependent on the processing temperature and glycerol content.[104]

A thermocompression efficiency was shown for thermoplastic corn starch with chitosan/chitin with a thermohydraulic press with a previous melt-mixing process with glycerol and water at 140°C. The films were homogeneous with a uniform thickness, good appearance, and antimicrobial activity.[68]

Edible Film Modifications

Lamination is another method for improving film performance; it consists of the formulation of multilayered structures and the combination of properties of different materials into one sheet. Multilayered films usually show a higher toughness and tensile strength than single-layer films. However, this technology is complex and solvent-consuming at high temperature and time; this increases the production costs. In addition, the high differences in the surface energy between layers could result in their separation.[89,107] For example, three-layer films obtained by the heat compression of dialdehyde-crosslinked starch and plasticized gelatin films as outer layers and plasticized gelatin–sodium montmorillonite composite films as an inner layer provided a new biodegradable multilayer material with a compact structure and modulated properties.[107] Individual films were obtained separately in a first step, and layers were stacked together by heat compression. Bioactive trilayer films were also prepared with poly(ε-caprolactone) as external layers and methyl cellulose with encapsulated EO as the internal film.[108,109] Other methods were proposed to improve the properties in active edible films based on chitosan by the application of moderate electric fields during processing.[4,110]

INDUSTRIAL APPLICATIONS OF EDIBLE FILMS

Edible films and coatings offer practical advantages, such as aesthetics, barrier properties, nontoxicity, and low cost.[4] In addition, the high compatibility of edible films with multiple active compounds has resulted in many interesting studies on the development of active food-packaging materials because of their ability to extend food shelf life. Some examples of these applications are summarized in Table IV.

EOs extracted from herbs and spices are the more usual active compounds in these formulations. In fact, the incorporation of natural extracts from plants, spices, and herbs represents a promising approach for the development of bioactive edible films/coatings with improved bioactive, mechanical, and physicochemical properties and applications.[12] For example, Pires *et al.*[121] studied the incorporation of different EOs to protein edible films. They concluded that the addition of citronella, coriander, tarragon, and thyme oils reduced the water-vapor permeability and increased the solubility in water in the resulting films. The amount of protein released from these films upon water solubilization was quite dependent on the EO composition.

Volpe *et al.*[111] studied the effect of lemon EO on carrageenan matrices and reported the efficiency of lemon EO in slowing down the microbial growth and lipid oxidation in trout fillets. In addition, the carrageenan matrix showed good performance in keeping the shiny and fresh aspect of trout beyond 7 days of cold storage. Conservation of fish patties was also studied by Salgado *et al.*[82] by the addition of clove EO to sunflower protein concentrates to obtain edible and biodegradable films with *in vitro* antioxidant and antimicrobial properties. These sunflower protein films contributed to the limitation of the lipid auto-oxidation and to the delay of the growth of total mesophiles when applied to refrigerated sardine patties.

Fernández-Pan *et al.*[112] studied the quality and extension of the shelf life of chicken breast with whey-protein isolate edible coatings with oregano or clove EOs. These insoluble and homogeneous whey-protein isolate coatings formed an imperceptible second skin covering the chicken breast, and they were effective in controlling the release of the antimicrobial compounds onto the food surface throughout the storage period.

Biomedical applications are other future trends for the use of edible films. In fact, the use of biopolymers as effective carriers for drug delivery has been extensively reported, and many reviews have been already published in the last few years.[122] For example, novel hydrogels were prepared by Chetouani *et al.*[123] by the crosslinking of gelatin with oxidized pectin. The reaction of the aldehyde groups of the oxidized pectin with the amino groups of gelatin was responsible for the crosslinking. In comparison with pure gelatin, these edible films exhibited a higher thermal stability and better blood compatibility with potential uses in wound dressings, tissue engineering scaffolds, and other biomedical fields.

CONCLUSIONS

The use of edible films has found a very important niche of applications, including in food packaging, biomedical applications, and others through their good performance as carriers for active compounds. Research in this field has largely increased in the last few years, but some drawbacks are still to be solved to permit their use in massive applications in the packaging of consumer goods, in particular difficulties in processing because most of the current research has been performed with wet methods. Nevertheless, this important bottleneck, which results in difficulties in up-scaling the laboratory results to industrial production, is currently under study, as shown in many of the most recent references in this review because the successful production of these active films on a large scale will be possible soon. The most updated prospect for the use of edible films as a real sustainable alternative to conventional plastic formulations in active food packaging and/or biomedical systems reflects their high possibility of being introduced into the market in the next few years.

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