

Edible Food Packaging with Natural Hydrocolloids and Active Agents

Ahmet Yemenicioğlu



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Preface

Due to the health concerns related to chemical food preservatives as well as environmental problems associated with fossil plastics, edible packaging with natural hydrocolloids and active agents (active edible packaging) has become increasingly popular as a food preservation method. In fact, the active edible packaging is now an interdisciplinary research field attracting interest not only from food and material scientists, but also from agronomists, microbiologists, biologists, pharmacists, and nutrition scientists. Researchers with different backgrounds have been collaborating to discover and characterize novel functions, synergistic interactions, and delivery methods of active agents, and to develop innovative and more applicable antimicrobial, antioxidant, flavor-release and bioactive edible packaging. The industrial interest in active edible packaging (especially coatings) has also been increasing continuously. Thus, to better understand the developments in the field, one should know the current content of active edible packaging (*see Chapter 1*) and factors fueling this rapidly-emerging preservation method. First of all, active edible packaging is the most sustainable packaging method since its main ingredients and functional components could be formed by natural hydrocolloids and active agents extracted mostly from agro-industrial wastes. Thus, this emerging packaging method provides an excellent opportunity for utilization of wastes into value-added products. In this book, detailed information has been provided about sources, extraction methods, the major characteristics of natural hydrocolloids (*Chapter 2*), and their ability to form different types of edible packaging (*Chapter 3*). Moreover, main sources and characteristics of natural active compounds have also been discussed in detail, and their potential as components of edible packaging has been analyzed (*Chapter 4*). The other major reason for increased interest in this field is that recent scientific developments, such as better understanding of synergistic interactions of active agents, and application of nanoencapsulation and controlled release technologies in their delivery, have enabled more effective use of natural phenolic compounds in active edible packaging. Developments in encapsulation technologies have also boosted the application of active packaging incorporated with probiotics, nutrients, and bioactive agents (bioactive packaging). These scientific developments and many other strategies used to enhance performance of active edible packaging have been discussed in the book in an easily comprehensible manner (*Chapter 5*). The book also contains basic proved methods of testing antimicrobial and antioxidant properties of edible packaging (*Chapter 6*), and over one hundred recent examples of active edible packaging applications (*Chapter 7*).

The examples have been selected carefully among the most applicable and up-to-date ones, covering a wide range of food, such as whole or minimally processed fresh fruits, vegetables, and mushrooms, nuts and seeds, raw and processed beef, pork, lamb, chicken and fish, dairy products, and bakery products, and dough food. As understood from its title and contents, this book is based on the use of natural hydrocolloids and active agents in edible packaging. Therefore, it lacks or contains minimum essential information about edible packaging of chemically-modified natural hydrocolloids, and chemical food additives. The information in this book will be of great interest, not only to researchers in academia and industry already working in the field, but also undergraduate or graduate students who are planning to start research or write a thesis in this field.

Ahmet Yemenicioğlu

I dedicate this book to

My wife Ayla and my daughter Feride Lila

for their endless support and understanding during my long writing sessions



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CHAPTER

1

Introduction to Active Edible Packaging

1.1 The history of edible packaging

Some people might think that the application of ‘edible film’ or ‘edible coating’ in food preservation is one of the new methods discovered in the 20th century. However, the small and large intestines of cattle and sheep have been used as natural casing material for sausages since ancient times. The earliest record about meat-stuffed casings (sausages) was discovered in almost 4,000-year-old Sumerian tablets found in Mesopotamia (Eckholm, 1985). The ripening of some cheese types in sacks made of lamb skin in Balkans and Turkey has also been applied since ancient times (Kalit *et al.*, 2010). However, the first record related to use of edible coatings in fruits is not too old as this is attributed to the Chinese, who formulated and applied edible wax coatings for preservation of oranges in the 12th century (Hardenburg, 1967). Moreover, it is also thought that the *yuba*, a proteic film formed at the surface of boiled soy milk, was discovered by Japanese in the 15th century as the first self-standing edible film applied for wrapping of food (Umaraw and Verma, 2017). In the 16th century, the coating of meat with fat (larding) in England is also an example of food preservation with edible coating (Bertuzzi and Slavutsky, 2016). After that, the use of gelatin coating in meats (Havard and Harmony, 1869) and the familiar process called ‘fruit waxing’ (Hoffman, 1916) were patented in the USA in the second half of 19th and at the beginning of 20th centuries, respectively. Then, the collagen casings were manufactured in Germany in the mid-1920s (Naga *et al.*, 1996). What about active edible packaging? Has this method been discovered recently by the modern food scientists? Yes, it is true that some patents exist related to incorporation of antifungal food preservatives into edible pectin and pectate films in 1950s (Owens and Schultz, 1952), but systematic studies to incorporate food preservatives into packaging had been accelerated in the 1980s and 1990s (Torres *et al.*, 1985; Guilbert *et al.*, 1996). Examples related to the use of active edible packaging in ancient times are scarce. However, it should be kept in mind that the ancient process of smoking applied to sausages causes accumulation of antimicrobial smoke components (e.g. acids, phenol, carbonyl) within the casing and on the sausage surface. Moreover, wrapping minced meat or rice and seasonings in grape leaves (causes grape leaf aromas and bioactive polyphenols to release into the filling during cooking) has been a traditional dish in Greece since ancient times (Cosme *et al.*, 2017). These historical knowledge mean

that some elementary applications of edible films have been laying around us for a very long time, while active edible packaging is an emerging new method that has boosted the global interest of using edible films in food preservation.

1.2 Definition of edible packaging

In general, ‘edible packaging’ is defined as a continuous protective matrix made up of polysaccharides, proteins or lipids (used alone or in combination), and applied to respiring or non-respiring food as a self-standing film (used as film wrap, casing or pouch) or coating that acts as a barrier against moisture, gas, flavor, aroma or oil transfer (Kester and Fennema, 1986; Guilbert *et al.*, 1986; Guilbert *et al.*, 1996; Miller and Krochta, 1997; Park, 1999). However, rapid developments in the field showed that edible food packaging is more than film, pouch, coating and casing. For example, different emerging packaging, such as antimicrobial stickers, gel-based pads, and electrospun mats produced by using edible materials could also be considered edible packaging (Tracz *et al.*, 2018; Boyacı and Yemenicioğlu, 2020; Kavur and Yemenicioğlu, 2020; Sameen *et al.*, 2021). All film-forming components and functional additives of edible films must be food-grade non-toxic substances selected considering related national or international regulations, and foods packed with these edible films must be labeled properly for their potential allergenic constituents (see Chapter 2 for different allergens in natural hydrocolloids) (Rojas-Graü *et al.*, 2009). Moreover, it should also be noted that edible films refer to films having thickness less than 254 µm while thicker films are defined as sheets (Janjarasskul and Krochta, 2010).

1.3 Definition of active packaging and active edible packaging concepts

The major definitions related to active packaging, according to European Commission Regulation No. 450/2009, are seen in Table 1.1. Active edible packaging takes some specific names, such as antimicrobial, antioxidant, flavor-release or bioactive edible packaging depending on the functionality of active component(s) used in their production. An antimicrobial or antioxidant edible packaging is manufactured mostly by incorporating (rarely impregnating) antimicrobial or antioxidant substance(s) into edible packaging or by using an inherently antimicrobial or antioxidant hydrocolloid in manufacturing of edible packaging (Appendini and Hotchkiss, 2002; Shendurje *et al.*, 2018). The other active packaging concepts are flavor-release packaging and bioactive packaging that are conducted by incorporating flavor substance(s) (Marcuzzo, 2010) and health-promoting substance(s) (or food-grade probiotic microorganism) into edible packaging, respectively (Lopez-Rubio *et al.*, 2006).

1.3.1 Antimicrobial packaging

Antimicrobial packaging improves food safety by inhibiting contaminated pathogenic bacteria and/or prolonging food’s shelf-life by suppressing growth of

Table 1.1: Major definitions related to active packaging according to European Commission Regulation No. 450/2009

Term/Concept	Definition
Active materials and articles	Materials and articles that are intended to extend the shelf-life or to maintain or improve the condition of packaged food; they are designed to deliberately incorporate components that would release or absorb substances into or from the packaged food or the environment surrounding the food.
Releasing active materials and articles	Active materials and articles designed to deliberately incorporate components that would release substances into or on to the packaged food or the environment surrounding the food.
Released active substances	Substances intended to be released from releasing active materials and articles into or on to the packaged food or the environment surrounding the food and fulfilling a purpose in the food.
Active component	An individual substance or a combination of individual substances which cause the active function of a material or article, including the products of an <i>in situ</i> reaction of those substances; it does not include the passive parts, such as the material they are added to or incorporated into.

spoilage flora using minimum amount of antimicrobial agents (Appendini and Hotchkiss, 2002). Antimicrobial agents, such as antimicrobial enzymes, bacteriocins, phenolic compounds, essential oils, etc. are loaded into edible packaging by different methods, such as incorporation, impregnation, coating or immobilization (Table 1.2). However, the most frequently used method is incorporation of antimicrobials into self-standing films or coatings. The hydrophilic antimicrobials are solubilized directly in film- or coating-forming solutions while hydrophobic ones are mostly homogenized with the aid of film-forming hydrocolloid and an emulsifier to obtain emulsion-based films or coatings. The nanoencapsulation of hydrophobic antimicrobials before incorporation is also a frequently used technique to disperse hydrophobic antimicrobial substances homogeneously within film- or coating-forming solutions. However, it is important to note that encapsulation is a key process for both hydrophilic and hydrophobic antimicrobials since this process also improves stability of antimicrobials and helps in sustaining their release rates. The antimicrobials can also be coated on to the surface of self-standing films by spreading, spraying or brushing, or they could be impregnated by dipping films into antimicrobial solution. The antimicrobials loaded into packaging by incorporation, impregnation or coating generally release on to packaged food's surface (also headspace of external food package, if volatile) at a certain rate, thus, showing antimicrobial activity both on the food surface and below food surface, depending on their capacity to diffuse into depths of food without losing their antimicrobial activity. In contrast, antimicrobials immobilized on to films or coatings can be used to obtain an antimicrobial effect only on the food surface (Lian *et al.*, 2012). Finally, inherently antimicrobial hydrocolloids are used in development of antimicrobial films and coatings. For example, chitosan is a unique antimicrobial hydrocolloid to be used as a self-standing film, casing, and monolayer or layer-by-layer coating

Table 1.2: Methods used to transform edible packaging into active edible packaging

Description	Name of method	Applications
Solubilization/dispersion/emulsification of free or encapsulated active agent(s) in the edible packaging forming solution.	Incorporation	Self-standing films, coatings, casings, pouches, gel-based pads, electrospun nanofiber mats
Dipping of packaging into solution of active agent(s).	Impregnation	Self-standing films, casings
Spreading/spraying/brushing of active agent(s) solution on to edible packaging surface.	Coating	Self-standing films, coatings, casings
Creating charge – charge interaction, covalent cross-linking, hydrogen bonding, etc. between active agent(s) and hydrocolloid that form the packaging.	Immobilization	Self-standing films, coatings, casings
Use of inherently antimicrobial (e.g. chitosan) or antioxidant (e.g. milk proteins) hydrocolloids in development of packaging.	-	Self-standing films, coatings, casings

(when combined with a negatively-charged hydrocolloid). However, films and coatings of inherently antimicrobial hydrocolloids show antimicrobial effect only on the food contact surface.

1.3.2 Antioxidant packaging

This type of packaging is applied mainly by using antioxidant loaded self-standing films or coatings to inhibit lipid oxidation or enzymatic browning. Recently, there has been increased interest in using antioxidant loaded nanofiber mats for packaging (Vilchez *et al.*, 2020). In general, antioxidants, such as phenolic compounds, essential oils, carotenoids, tocopherols, ascorbic acid and derivatives, proteins and peptides, etc. are loaded into edible packaging, using similar methods described for antimicrobial packaging. However, the antioxidant compounds, capable of inhibiting lipid oxidation or enzymatic browning, are released almost always from food-contact packaging on to the food surface. Rarely some inherently antioxidant hydrocolloids (e.g. milk proteins) are employed to obtain antioxidant films or coatings effective locally on the food surface (Shendurse *et al.*, 2018). However, performances of such inherently antioxidant coatings cannot be compared with those loaded with free antioxidant agents.

1.3.3 Flavor-release packaging

This active packaging concept involves incorporation of desired flavor compounds into edible films and coatings (Marcuzzo, 2010). The application of flavor-release packaging is useful in enhancing or maintaining the desired flavor attributes of

food during storage. However, to obtain maximum benefits from flavor-release packaging, it is essential to conduct encapsulation of incorporated flavor compounds, using suitable encapsulant and encapsulation methods. In some cases, the flavor compound could be encapsulated during film making by forming an emulsion between film-forming hydrocolloid and lipids (Marcuzzo, 2010).

1.3.4 Bioactive packaging

Active packaging is named bioactive packaging when its unique role is to enhance food impact on the consumer's health (Lopez-Rubio *et al.*, 2006). In bioactive edible packaging concept, probiotics and prebiotics or bioactive substances, such as phytochemicals, vitamins, marine oils, etc. are maintained on the food surface (within edible coating) or delivered on to the food surface (for solid food) or into food (for beverages). However, such a packaging must be designed considering the stability of bioactive agents during processing and storage of packaged food, and ensuring their bioavailability after consumption. In order to stabilize and improve their bioavailability, the bioactive compounds employed in packaging are treated by different technological methods (e.g. enzymatic modification, nanoencapsulation, co-encapsulation, dissolution by ultrasonication, etc.). Moreover, some compositional and structural modifications might be conducted in the food (if possible) to maximize bioavailability of delivered bioactive agents. The bioavailability of a bioactive agent is determined by limitations in its bioaccessibility (e.g. liberation from food matrix, solubilization in intestinal fluids and interactions, and insoluble complex formation), absorption (e.g. mucus layer transport, bilayer permeability, tight junction transport, active transporters, efflux transporters), and transformation (e.g. chemical degradation and metabolism) (McClements *et al.*, 2015). The definitions of basic terms used in nutrition science help to understand the challenges of bioactive packaging (Table 1.3).

1.4 Main materials used for development of active edible packaging

Proteins and polysaccharides (hydrocolloids) are the main materials used for development of active edible films and coatings. The self-standing edible films and coatings are produced mainly from (1) a single hydrocolloid (a protein or polysaccharide), (2) mixture of different hydrocolloids (blends or composites), or (3) mixture of hydrocolloids with lipids (emulsions or composites). The nanofiber mats, considered active edible packaging materials of future, are also manufactured from electrospinnable hydrocolloids (e.g. zein, gluten, gelatin, alginate) (Zhang *et al.*, 2006; Akman *et al.*, 2019; Karim *et al.*, 2020; Nie *et al.*, 2008; Fang *et al.*, 2011). However, although the lipids can be used to develop edible coatings, they cannot be utilized in manufacturing self-standing edible films due to their poor mechanical properties. Moreover, hydrophilic natural active compounds are not soluble in lipids and cannot be delivered on to the food surface with pure lipid coatings. In contrast, the majority of hydrocolloid-based packaging materials are compatible with the hydrophilic antimicrobial and antioxidant agents, and they could be utilized into

Table 1.3: Definitions of important terms necessary to understand principles of bioactive packaging

Term	Definitions	Reference
Bioavailability	The rate and extent to which the active ingredient or active moiety is absorbed from a drug product and becomes available at the site of action.	FDA (2002)
	The fraction of ingested nutrient that is available for utilization in normal physiologic functions and for storage.	Parada and Aguilera (2007)
Bioaccessibility	The fraction of an ingested biocomponent that becomes accessible for absorption through the epithelial layer of the gastrointestinal tract (GIT).	Dima <i>et al.</i> (2020)
	Fraction that is released from food matrix and is available for intestinal absorption.	Parada and Aguilera (2007)
Bioactivity	The ability of a compound to exhibit a biological effect (e.g. antioxidant effect, antimicrobial effect, anti-inflammatory effect, etc.).	Dima <i>et al.</i> (2020)
Potency of a bioactive compound	The concentration or quantity of a biocomponent necessary to produce the corresponding biological effect.	Dima <i>et al.</i> (2020)

emulsion-based packaging to accommodate hydrophobic active agents. Therefore, this book focuses on utilization of natural hydrocolloids in development of active edible packaging materials. It is accepted that only materials having the ability to form self-standing packaging could be the future alternatives to fossil plastic films. On the other hand, the lipids have important roles in improving the moisture barrier, swelling, and sustained release properties of hydrocolloid-based films and coatings. Moreover, most of the novel delivery systems, such as nanoemulsions, solid lipid nanoparticles, nanoliposomes, nanomicelles, etc. are based on lipids. Thus, the lipids are considered essential components used to improve functional and active properties of hydrocolloid-based packaging.

1.5 Edible packaging of different natural hydrocolloids

Hydrocolloids from plant, animal, algal (seaweed), and microbial sources could be used for development of active edible packaging, such as coatings, casings, self-standing films or nanofiber mats (Table 1.4). However, extensive efforts have been spent on employing proteins and polysaccharides obtained from animal and plant sources found worldwide as agro-industrial byproducts and wastes. The major polysaccharide-based hydrocolloids obtained from plants are cellulose, starch, and pectin. The cellulose obtained from wood or non-wood sources is the

Table 1.4: Abilities of major natural hydrocolloids to form edible packaging

Hydrocolloids	Coating	Extruded casing	Solution- cast film	Compression molded film	Extruded film (D or B) ^a	Nanofiber mat
Plant-origin polysaccharides						
Starch	X ^b	-	X	X	X (D)	-
Pectin	X	X	X	X	X (D)	-
Plant-origin proteins						
Zein	X	-	X	X	X (B)	X
Gluten	X	-	X	X	X (B)	X
Soy protein	X	X	X	X	X (D)	-
Animal-origin proteins						
Gelatin	X	-	X	X	X (B)	X
Collagen	X	X	X	X	X (B)	X
Na-caseinate	X	-	X	X	X (B)	-
Whey protein	X	-	X	X	-	-
Animal-origin polysaccharides						
Chitosan	X	X	X	X	-	-
Algal-origin polysaccharides						
Alginate	X	X	X	X	-	X
Carrageenans	X	-	X	-	-	-
Microbial-origin polysaccharides						
Pullulan	X	-	X	-	-	X
Xanthan	X	-	X	-	-	-

^a Slit-die (D) or blown (B) extruded film.^b The symbol "X" shows ability of hydrocolloid to form the indicated packaging type.

most abundant hydrocolloid on earth, but insolubility of natural cellulose prevents its direct use in development of edible packaging. The derivatives of cellulose obtained by chemical modification, such as cellulose ethers (e.g. carboxymethyl cellulose, methylcellulose, hydroxypropyl methylcellulose, hydroxyethyl cellulose, etc.) could be used to obtain solution-cast self-standing edible films and coatings. However, most cellulose ethers cannot be utilized alone by classical polymer processing methods because of their high viscosity between their glass transition temperature (Tg) and denaturation temperature (Tg) (Meena *et al.*, 2016). Moreover, electrospinning of cellulose ethers needs solubilization in organic solvents or use of electrospinnable carrier polymers (Frenot *et al.*, 2007). In contrast, native starches obtained from different sources, such as corn, wheat, rice, potato, or tapioca could be used effectively in development of edible packaging materials, such as coatings and solution-cast self-standing films. The self-standing edible films of starch can also be developed by compression molding and slit-die extrusion, but the development

of blown extruded starch films is problematic due to their highly brittle nature (Thunwall *et al.*, 2006; Shanks and Kong, 2012). The development of electrospun mats from starch is also not practical since this needs solubilization in solvents that should be removed after nanofiber production (Kong and Ziegler, 2014). The high or low methyl ester pectins obtained mainly from citrus peels are also used in manufacturing edible coatings and solution-cast self-standing edible films. The low methoxyl pectin can be effectively cross-linked with CaCl_2 to obtain insoluble solution-cast self-standing films or insoluble coatings fixed at the food surface. The pectin could also be thermosplasticized and utilized in extruded sausage casings (Liu *et al.*, 2005; Liu *et al.*, 2007) or compression molded films (Gouveia *et al.*, 2019), but data about applicability of blown extruded pure pectin films are scarce. Pectin is also among the hydrocolloids that need the presence of electrospinnable carrier polymers to obtain nanofibers (Cui *et al.*, 2016).

Chitosan is the only animal-origin polysaccharide used extensively in active edible packaging. Due to its unique inherent antimicrobial properties, chitosan has become one of the most important coating materials. Moreover, the recent application of chitosan as an antimicrobial sausage casing might trigger its more extensive use by the food industry (Adzaly *et al.*, 2016). Chitosan is also used in development of self-standing films by solution-casting and compression molding, but it cannot be used in manufacture of extruded films since it is not a thermoplastic material (Van den Broek *et al.*, 2015). The high viscosity of aqueous chitosan solution interferes with its electrospinning. Therefore, electrospinning of chitosan also needs solubilization in hazardous acids (e.g. trifluoroacetic acid) with addition of toxic solvents (e.g. dichloromethane) (Ohkawa *et al.*, 2004).

The corn zein, wheat gluten, and soy proteins are major plant-origin proteins used in development of edible packaging. Zein and gluten are highly functional proteins that can be used in development of different kinds of packaging (solution-cast, extruded or compression molded self-standing films, or coatings and electrospun nanofiber mats) (Gontard and Guilbert, 1998; Padgett *et al.*, 1998; Wang and Padua, 2003; Tanada-Palmu and Grosso, 2005; Akman *et al.*, 2019; Karim *et al.*, 2020). Gluten is also one of the rare hydrocolloids to be used in development of three-dimensional materials by injection molding (Gontard and Guilbert, 1998). The soy proteins is also used in development of edible coatings or self-standing films by solution-casting and compression molding (Stuchell and Krochta, 1994; Ogale *et al.*, 2000). The thermoplasticized soy proteins were also used for development of extruded casings and self-standing films, but extruded soy protein films have limited applicability due to their poor mechanical and moisture barrier properties (Naga *et al.*, 1996; Zhang *et al.*, 2001; Koshy *et al.*, 2015). The soy proteins cannot be used to develop electrospun nanofibers without the presence of other carrier proteins (Xu *et al.*, 2012). However, the gels obtained from soy proteins and their composites and emulsions are used in delivery of bioactive compounds, such as vitamins and phenolic compounds (Hu *et al.*, 2015; Ding and Yao, 2013; Brito-Oliveira *et al.*, 2017; Marinea *et al.*, 2021).

The animal-origin proteins, such as gelatin, collagen, sodium caseinate, and whey proteins are the most extensively used proteins by the food industry as functional and nutritional ingredients. Therefore, the utilization of these proteins

in development of edible packaging has also been studied extensively. The use of collagen in extruded sausage casings is probably the most important industrial edible film application in the world. The collagen suspensions prepared by acid-swelling/homogenization method could also be employed as coating, and in the manufacture of self-standing films by solution-casting (Wang *et al.*, 2016) and compression molding (Andonegi *et al.*, 2020). The native collagen can be converted into thermoplastic collagen (TC) by partial heat denaturation to obtain blown-extruded films, but these films have limited applicability due to their high sensitivity to moisture and poor mechanical properties in moist environments (Klüver and Meyer, 2013). Electrospinning of aqueous collagen solutions gives nanofibers, but this process transforms collagen into gelatin (Zeugolis *et al.*, 2008). This interferes with the use of collagen in electrospinning since gelatin itself can be electrospun into nanofibers. However, high solubility and mechanical weakness of gelatin are great limitations to obtain its electrospun mats suitable for industrial packaging applications (Zhang *et al.*, 2006). The pure gelatin is not commercially applied for casings, but is extensively used in food-coating applications. The self-standing films of gelatins can be manufactured by solution-casting, compression molding (Krishna *et al.*, 2012), and blown-extrusion (Andreuccetti *et al.*, 2012) methods, but widespread industrial use of these films is also limited due to their poor mechanical and water barrier properties, and high water solubility. Finally, the excellent gel-forming capacity of gelatin can be exploited to obtain active gel-based pads suitable both in absorbing drip-loss and in delivering natural antimicrobials and antioxidants on to food surface (Boyaci and Yemencioğlu, 2020). Sodium caseinate and whey proteins are the other animal-origin proteins to be used frequently in development of edible coatings and self-standing solution-cast edible films. Both these proteins can be used for development of compression-molded edible films, but only sodium caseinate was employed successfully to develop blown extruded edible films (Belyamani *et al.*, 2014a, b). Sodium caseinate and whey proteins are also among the hydrocolloids that cannot be utilized in nanofibers by electrospinning (Sullivan *et al.*, 2014; Tomasula *et al.*, 2016).

The major marine-origin polysaccharides extracted from seaweeds, such as sodium alginate and carrageenans (kappa-, lambda- and iota-), and microbial polysaccharides, such as pullulan and xanthan can be employed in manufacture of edible packaging. Sodium alginate is one of the most extensively used edible coating materials. Since cross-linking of sodium alginate by Ca^{++} atoms causes gelation, it is possible to develop insoluble fixed coatings or self-standing solution-cast edible films of this hydrocolloid. The combination of negatively-charged alginate with positively-charged chitosan also gives layer-by-layer coatings (Poverenov *et al.*, 2014). Moreover, extruded alginate or extruded blends of alginate with gelatin, pea protein, cellulose, or starch are employed to obtain (dry or wet) sausage casings (Liu *et al.*, 2007; Harper *et al.*, 2015; Marcos *et al.*, 2020). The alginate plasticized sufficiently with polyols is also used to obtain compression-molded self-standing films, but the development of extruded alginate films by thermal polymer processing methods is difficult due to its thermal degradation in the molten state (Gao *et al.*, 2017). It is also possible to obtain electrospun alginate nanofibers when this hydrocolloid is mixed with glycerol or its entanglements are enhanced

by CaCl_2 cross-linking (Nie *et al.*, 2008; Fang *et al.*, 2011). Therefore, the alginate is considered as one of the highly functional hydrocolloids suitable for edible packaging.

The carrageenans are also attracting industrial interest as edible coatings (Tavassoli-Kafrani *et al.*, 2016) while the application of pure carrageenans as self-standing films is very limited due to their highly hydrophilic and brittle nature. However, it was reported that the blending of carrageenans with other hydrocolloids or preparing their composites with lipids, nanoclays or nanocellulose might be employed to obtain more applicable edible films than those of pure carrageenans (Sedayu *et al.*, 2019). Moreover, the carrageenans are among the hydrocolloids that cannot be electrospun into nanofibers (Stijnman *et al.*, 2011). Finally, the potential of microbial polysaccharides, such as pullulan and xanthan as water-soluble edible coatings, should be noted (Kandemir *et al.*, 2005; Sharma and Rao, 2015). However, both pullulan and xanthan find limited edible film applications due to their high water solubility. The aqueous solutions of xanthan are not electrospinnable, but pullulan is well known for its ability to form electrospun nanofibers (Stijnman *et al.*, 2011).

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