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Review

Galactomannans use in the development of edible films/coatings for food applications

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Innovations constantly appear in food packaging, always aiming at creating a more efficient quality preservation system while improving foods' attractiveness and marketability.

The utilization of renewable sources for packaging materials, such as hydrocolloids from biological origin, is one the main trends of the industry. Edible films/coatings have been considered as one of the potential technologies that can be used to increase the storability of foods and to improve the existent packaging technology, helping to ensure the microbial safety and the preservation of food from the influence of external factors.

In view of these recent developments, the main objective of this review is to provide information concerning the utilization of galactomannans in the production of edible films/coatings. The most important features of these polysaccharides are discussed, namely: their structure and applications; physical, chemical, thermal and mechanical properties of galactomannan-based films/coatings; transport properties (in particular those related to moisture, oxygen, carbon dioxide exchange through the films/coatings); incorporation of active compounds (e.g. natural antimicrobials and/or antioxidants) and applications in food products.

It is viewed that in a near future tailored edible packaging based on polysaccharides can be applied to selected foods, partially replacing non-biodegradable/non-edible plastics.

Introduction

Packaging based on conventional synthetic materials has led to serious ecological problems due to their non-biodegradability. In the last decade there has been a growing interest in the development of thermoplastic materials from biodegradable biopolymers, particularly those derived from renewable resources (Petersen *et al.*, 1999). In this context, biopolymers can be an alternative source for packaging development. The main problems of biopolymers are their higher costs and less optimal physical and processing properties when compared with synthetic plastics. In addition, there have not been sufficient incentives for downstream processors to incorporate the biodegradable materials into their products (Siracusa, Rocculi, Romani, & Rosa, 2008). To date, biodegradable packaging has attracted great attention, and numerous research projects are under way in this field. One important reason for this attention is the marketing of environmentally friendly packaging materials. Furthermore, the use of biodegradable packaging materials has the greatest potential in countries where landfill is the main waste management tool (Farris, Schaich, Liu, Piergiovanni, & Yam, 2009; Mahalik & Nambiar, 2010; Petersen *et al.*, 1999).

Edible packaging material must meet requirements related with their transport properties (mainly water vapor, carbon dioxide and oxygen permeabilities), mechanical properties (especially their resistance to stretching and rupture), optical properties (mainly related with their opacity and color) and flavor (in most cases, flavorless coatings are needed). Also, the possibility of incorporation of active agents must also be considered.

The use of edible films/coatings based on natural polymers and food grade additives have been constantly increasing in the food industry. The films/coatings can be produced with a great variety of products such as polysaccharides, proteins, lipids, resins, with or without the

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addition of other components (e.g. plasticizers and surfactants). Polysaccharides that have been used to form films/coatings include starch and starch derivatives, cellulose derivatives, alginates, carrageenan, various plant and microbial gums, chitosan and pectinates (Lin & Zhao, 2007; Rinaudo, 2008). Their hydrophilic properties provide a good barrier to carbon dioxide and oxygen under certain conditions but a poor barrier to water vapor and deficient mechanical properties (Guilbert, 1986; Park, 1999). Galactomannans, natural polysaccharides commonly used in food industry, mostly as stabilizer, thickener and emulsifier, are one of the alternative materials that can be used for the production of edible films/coatings based on their edibility and biodegradability.

Edible films/coatings are gaining importance as components of biodegradable packaging and as an alternative to reduce the deleterious effects imposed by minimal processing e.g. on fresh-cut fruits (Vargas, Pastor, Chiralt, McClements, & González-Martínez, 2008). The semipermeable barrier provided by edible films/coatings is aimed at extending shelf-life by reducing moisture and solute migration, gas exchange, respiration and oxidative reaction rates, as well as suppressing physiological disorders on fresh-cut fruits. Edible films/coatings may also serve as carriers of food additives such as anti-browning and antimicrobial agents, colorants, flavors, nutrients and spices (Baldwin, Nisperos, Chen, & Hagenmaier, 1996; Cagri, Uspunol, & Ryser, 2004; Cutter & Sumner, 2002; Han & Gennadios, 2005; Kester & Fennema, 1986).

The functionality and performance of edible films/coatings mainly depends on their wettability, barrier and mechanical properties, which in turn depend on film composition, its formation process and the method of application on the product. Recently, wettability has been used to evaluate the efficiency of the coating of a food surface (Casariego *et al*., 2008; Park, 1999; Ribeiro, Vicente, Teixeira, & Miranda, 2007). A coating solution must wet and spread on the surface of the food product, and upon drying will form a film that should have the adequate properties and durability. The coating process thus involves wetting of the food product by the coating solution, and the possible penetration of the solution into them food's skin (Hershko, Klein, & Nussinovitch, 1996). In the last years, a great number of works have evaluated the effects of different factors in water vapor permeability (*WVP*), oxygen and carbon dioxide permeability (O_2P and CO_2P, respectively) and mechanical properties of polysaccharides films (Bangyekan, Aht-Ong, & Srikulkit, 2006; Bergo & Sobral, 2007; Carneiro-da-Cunha *et al*., 2009; Casariego *et al*., 2009; Chen, Kuo, & Lai, 2009).

Normally, plasticizers are added to films/coatings in order to improve their physical properties (Bergo & Sobral, 2007). They help to decrease brittleness and improve flexibility, through reducing the intermolecular forces and increasing the mobility of polymeric chains (Rivero, García, & Pinnoti, 2010; Sothornvit & Krochta, 2001). Lipids, due to their hydrophobic behavior, are often added to polysaccharide films aiming at decreasing their hydrophilicity and consequently, decreasing the *WVP*. The incorporation of active substances such as antibacterial, antifungal and antioxidant is one of the emerging utilizations of edible films/coatings; leading, in some cases, to changes in the physicochemical properties of edible films/coatings (Lee, 2005).

This paper reviews the most recent and known literature dealing with galactomannan-based films/coatings and exposes some of the strategies that can be employed for the manipulation of their properties, their utilization for the incorporation of antioxidant and antimicrobial compounds and their application in the food industry.

Galactomannans

Galactomannans are heterogeneous polysaccharides composed by a β-(1−4)-D-mannan backbone with a single D-galactose branch linked α-(1−6) (Fig. 1). They differ from each other by the mannose/galactose (M/G) ratio. These gums are mostly obtained from the endosperm of dicotyledonous seeds of numerous plants, particularly the *Leguminosae*. The endosperm has several functions: it serves as food reserve for germinating seeds and it retains water, preventing the complete drying of the seeds (Gidley & Reid, 2006; Srivastava & Kapoor, 2005).

The general procedure to obtain galactomannans from seeds combines extraction and purification processes. Briefly, the seed hull is removed from the seeds and the germ is separated from the endosperm. The most used separation procedures are: filtration, sifting and in some cases (e.g. lab scale) they are separated manually. The endosperm is dissolved in water (at temperatures that can range between 20 and 120 °C), followed by a precipitation step using alcohol (in a ratio water:alcohol that can range between 1:1 and 1:3). No effects of the alcohol on galactomannans' structure have been reported (Cerqueira, Pinheiro *et al*., 2009; Cunha, Vieira, de Paula & Feitosa, 2009; Dakia, Blecker, Robert, Wathelet, & Paquot, 2008; Vendruscolo *et al*., 2009).

The great advantage of galactomannans is their ability to form very viscous solutions at relatively low concentrations that are only slightly affected by pH, ionic strength and heat processing (Sittikijyothin, Torres, & Gonçalves, 2005). Galactomannans' viscosity tends to remain constant over a broad pH range (1−10.5), mainly due the neutral character of their molecules, and also is not expected that their properties will change with ionic strength. However some degradation may occur under highly acidic and alkaline conditions at high temperatures. The viscosity and the stability of the galactomannan solutions upon storage depend on the time and temperature used during the film-forming solution preparation. Heating the galactomannans at temperatures above 60 °C tends to provide a high initial viscosity but leads to an inferior stability (in terms of time-dependant changes in viscosity). The most convenient temperature depends on the galactomannan source. For

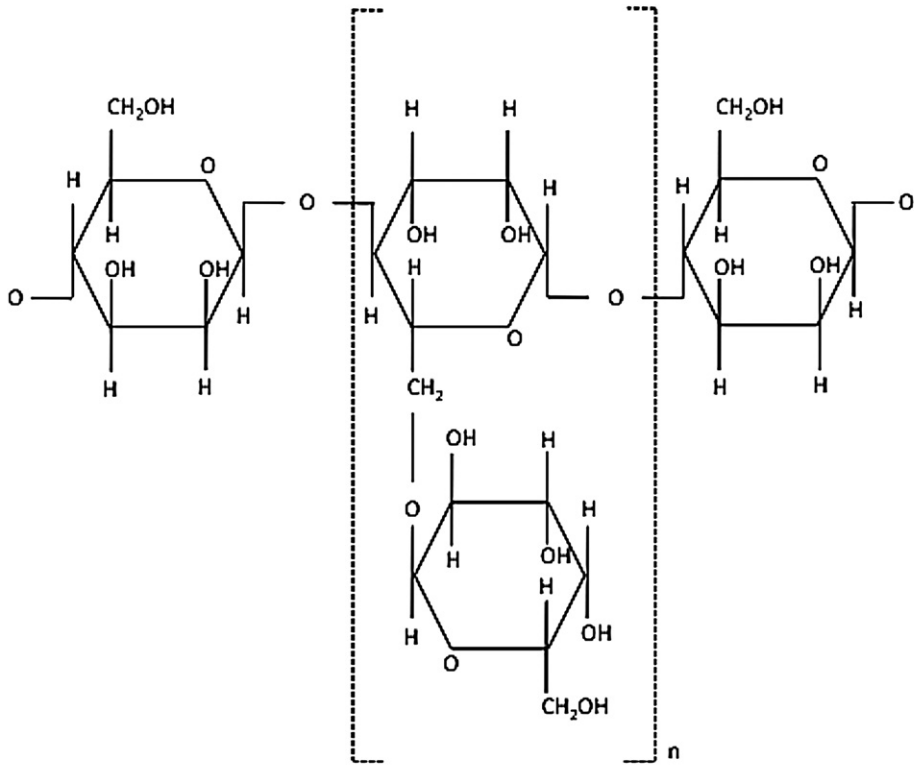

Fig. 1. General molecular structure of galactomannan.

example, locust bean gum must be heated at 80 °C for 20–30 min to guarantee its full dispersion, while the optimal conditions to disperse guar gum involve heating at 25–40 °C for 2 h (Srivastava & Kapoor, 2005).

Galactomannan solutions usually exhibit a non-Newtonian behavior, in which the viscosity decreases with the increase of shear rate (Garti, Madar, Aserin, & Sternheim, 1997). The degree of substitution in galactomannans profoundly affects their solution properties (Izydorczyk, Cui, & Wang, 2005; Kök, Hill, & Mitchell, 1999; Neukom, 1989).

The characterization of physicochemical properties of galactomannans can be performed using different techniques (e.g. gas chromatography, high-pressure anion exchange chromatography, size exclusion chromatography, ^{13}C NMR spectroscopy, capillary viscometry, shear and extensional rheology). The most important parameters that define the nature of a galactomannan are: mannose and galactose (M/G) ratio, average molecular weight, fine structure, and intrinsic viscosity. Mannose and galactose ratio, the degree of substitution and the degree of polymerization have been reported to affect edible films properties (Mikkonen *et al*., 2007). No effect of the average molecular weight as been reported for galactomannan films. The monomeric sugars content and M/G ratio are generally determined by gas chromatography or by high-pressure anion exchange chromatography after partial or total hydrolysis catalyzed by acid. The molecular weight distributions can be determined by size exclusion chromatography. The galactose distribution along the mannan chain can be characterized by ^{13}C NMR spectroscopy, or by enzymatic methods with β-D-mannanase that specifically degrades the non-substituted regions of galactomannans (Cerqueira, Pinheiro *et al*., 2009; Cunha *et al*., 2009; Dakia *et al*., 2008; Vendruscolo *et al*., 2009). Intrinsic viscosity can be determined using a capillary viscometer, by application of Huggins' and Kramer's equations (Cerqueira, Pinheiro *et al*., 2009; Sittikijyothin *et al*., 2005). Furthermore, the evaluation of the rheological behavior plays an important role in the characterization of galactomannan solutions, since these are often used to modify textural attributes (Marcotte, Taherian, Trigui, & Ramaswamy, 2001). This characterization can be performed through shear (steady and dynamic conditions) and extensional rheology (Bourbon *et al*., 2010).

The three major galactomannans of commercial importance in food and non-food industries are guar gum (GG, *Cyamopsis tetragonolobo*, M/G ratio: 2:1), tara gum (TG, *Caesalpinia spinosa*, M/G ratio: 3:1) and locust bean gum (LBG, *Ceratonia siliqua*, M/G ratio: 3.5:1) (Dakia *et al*., 2008; Gidley & Reid, 2006). However, the industry trends demand the introduction of alternative sources of seed gums and therefore it is important to search for such alternative renewable sources (Joshi & Kapoor, 2003). In some works galactomannans have been used in binary mixtures with other polysaccharides such as xanthan gum, agar and k-carrageenan, to form gels with new properties (Bresolin, Milas, Rinaudo, Reicher, & Ganter, 1999; Fernandes, Gonçalves, & Doublier, 1991; Pinheiro *et al*., 2011; Vendruscolo, Andreazza, Ganter, Ferrero, &

Bresolin, 2005). Some of the most used galactomannans and their alternative sources are presented in Table 1.

M/G ratio varies considerably depending on the galactomannan source and typically ranges between 1.1 and 5.0 (Table 1) (Srivastava & Kapoor, 2005) Variations in the galactomannans' structure, particularly in M/G ratio and fine structure, cause significant changes in the solubility, viscosity and in the interactions between galactomannans and other polysaccharides. Usually, galactomannans with higher galactose content (such as GG) are ready soluble in water and exhibit a lower tendency to form gels as a result of synergistic interactions, when compared with galactomannans with a lower M/G ratio (such as LBG). The higher solubility of GG has been attributed to the presence of a higher number of side chains, which keep the main mannose chains far enough to prevent effective intermolecular interactions. On the other hand, galactomannans with fewer side chains (higher M/G ratio) can interact with other polysaccharides due to their long blocks of unsubstituted mannose units (Srivastava & Kapoor, 2005).

Galactomannans can often be used in different forms for human consumption. Featuring different physicochemical properties, galactomannans are a versatile material used for many applications: they are excellent stiffeners and stabilizers of emulsions, and the absence of toxicity allows their use in the textile, pharmaceutical, biomedical, cosmetics and food industries (Baveja, Rao, Arora, Mathur, & Vinayah, 1991; Krishnaiah, Karthikeyan, Gouri Sankar, & Satyanarayana, 2002; Varshosaz, Tavakoli, & Eram, 2006; Vendruscolo *et al*., 2009; Vieira, Mendes, Gallão, & De Brito, 2007). Particularly in the food industry the main applications of galactomannans are in dairy products, fruit-based water gels, powdered products, bakery, dietary products, coffee whiteners, baby milk formulations, seasonings, sauces and soups, tinned meats and frozen and cured meat foods. This broad range of applications reflects a great number of different functional characteristics including high solution viscosity, stabilization of frozen systems and mixed gel formation with other polysaccharides and proteins (Gidley & Reid, 2006).

Recently, some works showed the possibility of using galactomannans in the formation of films and coatings (Cerqueira, Lima, Souza *et al*., 2009; Cerqueira, Lima, Teixeira, Moreira, & Vicente, 2009; Martins, Cerqueira, Souza, Avides, & Vicente, 2010; Mikkonen *et al*., 2007).

Galactomannan-based films/coatings

The literature on the characterization and application of galactomannans as films and/or coatings is very limited when compared with that available for other polysaccharides. However, very recent works brought new perspectives about the properties and the utilization of edible films/coatings from galactomannan sources. This was mostly due to the specific characteristics of galactomannans that form very viscous solutions at relatively low concentrations, and only need water in their preparation (Cerqueira, Lima, Souza, 2009a; Cerqueira, Lima, Teixeira, 2009b; Cerqueira, Sousa-Gallagher et al., 2010; Conforti & Totty, 2007; Lima *et al*., 2010; Martins *et al*., 2010; Rojas-Argudo, del Río, & Pérez-Gago, 2009).

One of the first works that characterized the *WVP* of galactomannan films studied the influence of increasing concentrations of polyethylene glycol (PEG) with different molecular weights (Aydinli & Tutas, 2000). The results showed that for LBG films with the addition of PEG with a molecular weight between 200 and 600 Da, the *WVP* values increased for higher concentrations of plasticizer. However, when PEG 1000 was used, the *WVP* values decreased when higher concentrations of plasticizer were added. The different behavior observed between the films containing PEG 1000 and PEG with lower molecular weight was explained by the solid state of PEG 1000 at room temperature. Moreover, PEG 200 presents the lowest values of *WVP*, which has been attributed to its lower hydrophobic character when compared with that of PEG of higher molecular weight. In 2004, the same authors studied

Table 1. Commercial and alternative sources of galactomannans, their mannose/galactose (M/G) ratio and possible applications.

Species	M/G ratio	Applications	Reference
Ceratonia siliqua (locust bean gum)	3.9:1	Thixotropic, binder, stabilizer, lubricator	(Prado, Kim, Özen & Mauer, 2005)
Caesalpinia pulcherrima	2.9:1		(Cerqueira, Pinheiro *et al*., 2009)
Gleditsia triacanthos	2.8:1	Stabilizer, thickener, emulsifier	(Bourbon *et al*., 2010; Cerqueira, Pinheiro *et al*., 2009; Sciarini, Maldonado, Ribotta, Pérez & León, 2009)
Cesalpinia spinosum (tara gum)	2:1		(Prado *et al*., 2005)
Dimorphandra gardneriana Tul	1.8:1		(Cunha *et al*., 2009)
Cyamopsis tetragonolobus (guar gum)	1.8:1	Stabilizer, thickener, emulsifier, firming agent	(Prado *et al*., 2005)
Adenanthera pavonina	1.4:1		(Cerqueira, Pinheiro *et al*., 2009)
Prosopis pallida (mesquite gum)	1.4:1	Stabilizer, thickener,	(Chaires-Martínez, Salazar-Montoya & Ramos-Ramírez, 2008)
Mimosa scabrella	1.3:1		(Vendruscolo *et al*., 2009)
Trigonella foenum graecum (fenugreek gum)	1:1		(Prado *et al*., 2005)

Table 2. Comparison between the values of water vapor permeability (*WVP*), oxygen permeability (O_2P), tensile strength (*TS*) and elongation-at-break (*EB*) values for galactomannan-based films and those for common commercial films.

Films	*WVP* × 10^{-11} (g Pa^{-1} s^{-1} m^{-1})	O_2P × 10^{-15} (g Pa^{-1} s^{-1} m^{-1})	*TS* (MPa)	*EB* (%)	Reference
LBG:PEG200 (0.7:0.3)	1.78		≈28	≈10.2	(Aydinli & Tutas, 2000; Aydinli *et al.*, 2004)
LBG:PEG200 (0.7:1.2)	2.12		≈17	≈1.8	(Aydinli & Tutas, 2000; Aydinli *et al.*, 2004)
LBG (1%):Gly (0.2%)			∼25	∼25	(Mikkonen *et al.*, 2007)
LBG (1%):Gly(0.6%)			∼20	∼70	(Mikkonen *et al.*, 2007)
GG (1%):Gly (0.2%)			∼13	∼5	(Mikkonen *et al.*, 2007)
GG (1%):Gly (0.6%)			∼6	∼55	(Mikkonen *et al.*, 2007)
LBG (0.7):PEG200 (0.3)	3.1				(Bozdemir and Tutas 2003)
LBG (0.7):PEG200 (0.3): beeswax (0.1)	1.7				(Bozdemir and Tutas 2003)
CP:Gly (0.5:1.0)	5.25	0.97			(Cerqueira, Lima, Teixeira, 2009)
CP:Gly (0.5:2.0)	7.70	1.10			(Cerqueira, Lima, Teixeira, 2009)
High-density polyethylene	<0.1	1.6–16.5	10–100	>100	(Han & Gennadios, 2005)
Cellophane	1.2–11.5	0.2–1.6	>100	10–100	(Han & Gennadios, 2005)

the mechanical and the light transmittance of the same films, but without using PEG 1000. They observed that *TS* and *EB* decreased for higher concentrations of PEG. Also, the luminous transmittance and total light transmittance decreased for higher PEG concentrations (Table 2) (Aydinli, Tutas, & Bozdemir, 2004).

Bozdemir and Tutas (2003) studied the *WVP* of edible films made with LBG and various plasticizers (glycerol, propylene glycol and PEG 200). They showed that films containing PEG 200 and sorbitol had the lowest *WVP* values and films containing glycerol have the highest *WVP* values. These results were explained by the higher water affinity of glycerol, when compared to PEG 200 and sorbitol (Table 2) (Bozdemir & Tutas, 2003).

In 2006, Barkalow *et al.* presented a patent where the utilization of a low viscosity hydrolyzed vegetable gum as a film forming solution was evaluated. These authors concluded that the invention presents an advantage to reduce the gumminess and off-flavors (Barkalow, Zyck, & Soto, 2006) of the products where it would be applied.

Mikkonen *et al*. (2007) also used galactomannans as an alternative for films formation. Mechanical and thermal properties of different film formulations from GG and LBG with glycerol and sorbitol were tested. In addition, they applied an enzymatic treatment to GG, thus obtaining structurally modified galactomannans. With this work they have shown that films from galactomannans with lower galactose content exhibited higher *EB* and *TS* values (in some cases with *TS* and *EB* in the range of high-density polyethylene and cellophane, respectively). From thermal measurements these authors showed that films plasticized with sorbitol present higher T_g values when compared with films plasticized with glycerol. Moreover, it was shown that those galactomannan films are in rubbery state at room temperature ($T_g < 0$ °C) (Table 2) (Mikkonen *et al*., 2007).

In 2009, Cerqueira *et al*. evaluated novel sources of galactomannans (*Caesalpinia pulcherrima* and *Adenanthera pavonina*) to be used as coatings applied on tropical fruits. These authors described a methodology to optimize the composition of edible coatings taking into account parameters such as wettability, permeability to gases and mechanical properties. The results showed that the increase of plasticizer (glycerol) concentration in films made with galactomannans of either species lead to higher values of *WVP* and O_2P. On the other hand, the values of CO_2P decreased for higher values of glycerol.

Interaction of galactomannan films/coatings with other materials

In the latest years, a great number of works evaluated the utilization of different materials and the blending with other types of biopolymers to enhance the properties of polysaccharide-based films/coatings (Cerqueira, Souza, Martins, & Vicente, 2010). Different materials can be used in order to taylor polysaccharide-based films/coatings properties.

Lipids are one of the most used materials aiming at decreasing the hydrophilicity of polysaccharide-based films/coatings leading to a decrease of the water sensitivity of this kind of materials, thus improving properties as *WVP* and water solubility. Furthermore, transport (of oxygen and carbon dioxide) and mechanical properties of lipid-based films/coatings can be improved through blending with polysaccharides. E.g. waxes have been used as hydrophobic components to improve the water barrier properties of edible films (Pérez-Gago & Krochta, 2001).

Chen and Nussinovitch (2001) evaluated the *WVP*, O_2P, CO_2P and the roughness of wax films (carnauba wax or shellac) blended with LBG and GG and compared their properties with those of commercial coatings (polyethylene and shellac). The *WVP* values of those films did not show statistically significant differences, however the incorporation of GG and LBG in wax coatings decreased the permeability to oxygen and carbon dioxide (Chen & Nussinovitch, 2001). The same authors studied the effects of introducing LBG and GG in traditional wax formulations when applied in citrus fruits and showed that the LBG-wax coating produced the juice with the best taste (Chen & Nussinovitch, 2000).

In another work (Bozdemir and Tutas 2003), LBG was blended with two lipids (stearoptene and beeswax) and the *WVP* was measured, in order to evaluate if this kind of hydrophobic compounds could be used to improve the films' transport properties. Results showed that films with stearoptene and beeswax present lower *WVP* values when compared with control films. Moreover, it was shown that if PEG 200 and sorbitol were used as plasticizers, LBG films with stearoptene would present lower *WVP* values than LBG films with beeswax; the authors explained these results based on the higher hydrophilicity of beeswax.

Lima *et al*. (2010) successfully blended collagen with two galactomannans from different species (*A. pavonina* and *C. pulcherrima*). The compositions of films/coatings with different proportions of galactomannan, collagen and glycerol were optimized based on films' wettability, transport and mechanical properties; these films were subsequently used to coat mangoes and apples (Lima *et al*., 2010).

Recently, Martins, Cerqueira, Bourbon, Pinheiro, and Vicente (2011) studied the effect of different LBG and κ-carrageenan ratios on film properties. Edible films composed by different mixtures of κ-carrageenan and LBG were developed and their physicochemical properties were evaluated. The films composed by 40/60% of κ-carrageenan/LBG showed a synergistic effect presenting enhanced water vapor barrier and mechanical properties when compared with the other samples. Mixtures of different materials besides allowing functionality also brings cost savings because the total amount of hydrocolloids used is lower, showing that these mixtures could be the best choice to be applied on food systems.

Galactomannan films/coatings with antimicrobial and antioxidants

Besides their protective effects, edible films/coatings can also act as carriers for a broad range of food additives, including antioxidants, anti-browning agents, antimicrobials, colorants and flavors. As a result, wrapped or coated foods' shelf-life could be extended, the risk of pathogen growth on food surfaces could be reduced and the sensory quality enhanced (Vargas *et al*., 2008). The interactions with the food or the food environment make edible films/coatings an excellent alternative or complement to traditional food packaging. Several studies were carried out addressing the antimicrobial and antioxidant properties of some compounds such as plant extracts (oregano, rosemary), enzymes (lysozyme), bacteriocins (nisin) and salts (potassium sorbate) (Cagri *et al*., 2004; Gómez-Estaca, Bravo, Gómez-Guillén, Alemán, & Montero, 2009; Pranoto, Salokhe, & Rakshit, 2005).

The selection of active agents to be incorporated should be limited to food-grade substances since they have to be consumed along with the edible films or coatings. Additionally, when incorporating some compounds in edible films and coatings it is important to evaluate their impact on film/coating functionality because its presence may affect films/coatings' basic functional properties (e.g. water vapor and gas barrier properties or solute transport properties). The influence of an ingredient on films/coatings functionality depends on its concentration, stability, chemical structure, degree of dispersion in the film/coating and degree of interaction with the polymer (Suppakul, Miltz, Sonneveld, & Bigger, 2003). Different methods can be used to incorporate active agents in films/coatings. The choice for a particular method will depend on the properties of the active agents used and their interaction with the galactomannan matrix. In a great number of cases the dispersion in the film forming solution is the methodology adopted for a successful incorporation into edible films/coatings matrix. In the case of hydrophobic active agents, their incorporation into edible films/coatings matrices can be achieved with the help of a surfactant that will promote the formation of micelles, where the active agent is entrapped. Also available is the option for high energy methods (e.g. homogenization by high speed blender) (Cerqueira, Souza, Martins, Teixeira, & Vicente, 2010; Fabra, Hambleton, Talens, Debeaufort, & Chiralt, 2011; Seydim & Sarikus, 2006).

Significant research has been performed to assess the antimicrobial and antioxidant properties of compounds when added to polysaccharide-based films/coatings. Trinetta, Floros, and Cutter (2010) demonstrated that pullulan films incorporating sackacin A had antimicrobial activity against *Listeria monocytogenes* when applied on the surface of turkey breast (Trinetta *et al*., 2010). (Seol, Lim, Jang, Jo, & Lee, 2009) reported that κ -carrageenan-based films containing ovotransferrin combined with EDTA showed an inhibitory effect against total aerobic bacteria and *E. coli* growing on chicken breast during storage at 5 °C.

However, the works in which galactomannan films/coatings are used as a vehicle for antimicrobial and antioxidant compounds are very scarce.

Martins *et al*. (2010) used galactomannan from *Gleditsia triacanthos* incorporating nisin to decrease the growth of *L. monocytogenes* on Ricotta cheese stored at 4 °C. The incorporation of nisin into galactomannan films led to an increase of their opacity, CO_2P, TS and EB and to a decrease of WVP and O_2P. Galactomannan-based edible coatings, when combined with nisin, showed good performance in reducing surface post-contamination of cheese products during storage. After 7 days of storage, the count of *L. monocytogenes* on cheese samples treated with coating containing nisin was 2.2 log CFU g^{-1} lower than those in cheese samples without coating.

As previously stated, galactomannan films/coatings could also carry antioxidants compounds. The galactomannan from *G. triacanthos* was used to entrap antioxidant extracts (obtained during the extraction of the galactomannan itself). The film with antioxidant extracts showed to have *in vitro* antioxidant capacity. It was also observed that the incorporation of the extracts led to an increase of

WVP and to changes in the color of the films (Cerqueira *et al.*, 2010).

Applications

Edible films/coatings can be useful as barriers to gases; if their composition is changed in order to provide the appropriate mechanical properties they can also be useful for food protection, reducing bruising and breakage and thus improving food integrity. Applications of galactomannan edible films/coatings are discussed below.

Chen and Nussinovitch (2000) studied the effects of the introduction of LBG and GG in traditional wax formulations when applied in citrus fruits. They showed that despite the observed changes in the O_2P and CO_2P of the films, when GG and LBG were added to wax formulations the internal oxygen and carbon dioxide concentrations did not present significant differences, as expected (Chen & Nussinovitch, 2000). The same authors studied the effects of coatings application on two easy-peeler citrus. They compared the performance of the commercial waxes and the inclusion of the GG and LBG in the coating formulation and no differences were reported in terms of weight loss. Moreover, the citrus with the LBG-wax coating produced the juice with the best taste (Chen & Nussinovitch, 2001).

The shelf-life quality of freshly harvested apples (cv. Golden Delicious) coated with three individually developed lipid/hydrocolloid coatings in which LBG was one of the constituents was assessed throughout refrigerated storage. The application of the coatings resulted in a lower internal oxygen concentration and in a lower firmness of the fruit when compared with the non-coated apples group. Sensory analyses showed that coated apples maintained consistent quality in firmness, crispness and juiciness throughout the storage period (Conforti & Totty, 2007).

Recently, 'Fortune' mandarins were coated with LBG-based coatings in order to extend their shelf-life, improve the external appearance and avoid flavor degradation of the fruit. Two studies were performed: in the first one, three experimental LBG–lipid edible coatings were tested and compared with a commercial wax and with the uncoated control. Among the experimental coatings, the coating with beeswax and glycerol and without carnauba wax and olein was the best for controlling weight loss and improving gloss. The second experiment was designed to optimize the performance of the best coating chosen from the first experiment with two modifications: decreasing emulsion solids content and increasing plasticizer content. Both coating modifications decreased ethanol levels in the juice compared with the unmodified coatings. From these, the coating modified by increasing glycerol content showed the best performance in controlling weight loss, improving gloss and reducing ethanol content (Rojas-Argudo *et al.*, 2009).

The reported works show how different coatings constituents can affect the effectiveness of the coatings to extend the shelf-life of foods. However, if a great number of different compositions have to be tested, the number of combinations can increase and great variability or different results can be obtained.

In the last years, galactomannans have been evaluated as coatings for fruit and cheese using wettability as the evaluation parameter to select the best coatings. In 2009, Cerqueira *et al.* studied the wettability of sixteen different formulations of galactomannan solutions in a semi-hard cheese. The three coatings with the best wettability values were chosen for the subsequent evaluation of the transport properties. The film/coating with the lowest *WVP* and O_2P values (1.5% of galactomannan, 2.0% of glycerol and 0.5% of oil) was finally chosen to be applied on cheese. Results showed that this coating formulation could be used to decrease gas transfer rates, weight loss and color changes in cheese (Cerqueira, Lima, Souza *et al.*, 2009; Cerqueira, Sousa-Gallagher *et al.*, 2010). *G. triacanthos* galactomannan with and without nisin was used as coating and tested against *L. monocytogenes* on Ricotta cheese at 4 °C. The best formulation was selected based on the wettability of the coatings on cheese samples. Results showed that the cheese coated with nisin-added galactomannan film was the treatment that revealed the best results in terms of microbial growth delay during 28 days. Also weight loss and moisture content decreased when those coatings were applied. These results demonstrated that novel galactomannan-based edible coatings, when combined with nisin, may provide consumer-friendly alternatives to reduce post-contamination on cheese products during storage (Martins *et al.*, 2010).

The same method was used for the evaluation of galactomannan-based coating formulations on fruits. Cerqueira *et al.* (2009) evaluated nine formulations of two galactomannans species (*C. pulcherrima* and *A. pavonina*) as coatings for five tropical fruits. The formulations that exhibited the best wettability values were cast and dried and their transport and mechanical properties were evaluated. The selected coatings were: for acerola–0.5% of *A. pavonina* galactomannan and 1.0% of glycerol; cajá– 1.0% of *A. Pavonina* galactomannan and 1.0% of glycerol; mango and pitanga–1.5% of *A. pavonina* galactomannan and 1.0% of glycerol; and seriguela–0.5% of *C. pulcherrima* galactomannan and 1.5% of glycerol. In a subsequent work (Lima *et al.*, 2010), the same galactomannans were blended with collagen and their application on apple and mango was evaluated, following a similar methodology. Results showed that the application of the coatings lead to a decrease of gas transfer rates of the fruits. For mangoes, a coating of *A. pavonina* galactomannan (0.5%), collagen (1.5%) and glycerol (1.5%) decreased oxygen consumption and carbon dioxide production in 28% and 11%, respectively. For apple, the oxygen consumption and carbon dioxide production decreased for both gases by approximately 50%, with the utilization of a coating of *C. pulcherrima* galactomannan (0.5%) and collagen (1.5%).

Final remarks

The aim of this review was to provide an insight on the relevant work that is being developed in the area of novel galactomannan-based biodegradable packaging materials and to explore the potential use of those alternative materials by the food industry. The mechanical, barrier and rheological properties of galactomannans films/coatings may be used to improve the stability, safety, and quality of food products.

Nowadays, a great discussion exists about the potential applications of edible films/coatings on food products. The general trend is to find the correct combination between the food product and the edible film/coating, which will ensure the success of the technology (e.g. despite of the high *WVP* values generally found for edible coatings, they can be used successfully to reduce the oxygen and carbon dioxide exchange rates of some food products, as these materials present lower oxygen and carbon dioxide permeability values when compared with synthetic materials). Nevertheless, further research is needed to taylor galactomannan-based films/coatings for each specific application, especially if they are to serve as vehicles of bioactive compounds (e.g. antimicrobials and antioxidants).

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Abbreviations

M/G	mannose to galactose ratio
O_2P	oxygen permeability
CO_2P	carbon dioxide permeability
WVP	water vapor permeability
TS	tensile strength
EB	elongation-at-break
T_g	glass transition temperature
GG	guar gum (*Cyamopsis tetragonolobo*)
TG	tara gum (*Caesalpinia spinosa*)
LBG	locust bean gum (*Ceratonia siliqua*)
CP	*Caesalpinia pulcherrima*
PEG	polyethylene glycol
Gly	glycerol
O_2	oxygen
CO_2	carbon dioxide

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