Edible films and coatings: Structures, active functions and trends in their use

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ELSEVIER
TRENDS IN FOOD SCIENCE & TECHNOLOGY
Imaging technologies for fish inspection
Cheeses with reduced sodium – functionality, health benefits and sensory properties
Edible films and coatings – structures, functions and use
Modelling analysis of complex food systems
Bioactive and possible health benefits of edible brown seaweeds
Food hygiene in aseptic processing and packaging
Iodine sequestration by amylose to combat iodine deficiency
Expanded 'Fermi Solution' for estimating relationship between product spoilage or deterioration and number of consumer complaints

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Trends in Food Science & Technology 22 (2011) 292–303

TRENDS IN FOOD SCIENCE & TECHNOLOGY

Review

Edible films and coatings: Structures, active functions and trends in their use

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Edible films and coatings are thin layers of edible materials applied on food products that play an important role on their conservation, distribution and marketing. Some of their functions are to protect the product from mechanical damage, physical, chemical and microbiological activities. Their use in food applications and especially highly perishable products such as horticultural ones, is based on some particular properties such as cost, availability, functional attributes, mechanical properties (flexibility, tension), optical properties (brightness and opacity), the barrier effect against gases flow, structural resistance to water and microorganisms and sensory acceptability. In this piece of work, the lastest advances on their composition (polymers to be used in the structural matrix), including nanoparticles addition, and properties have been reviewed, as well as the trends in the research about their different applications, including oil consumption reduction in deep-fat fried products, their use in combination with bioactive compounds that bring foodstuff additional functions and shelf life extension of highly perishable products.

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Introduction

An edible coating (EC) is a thin layer of edible material formed as a coating on a food product, while an edible film (EF) is a preformed, thin layer, made of edible material, which once formed can be placed on or between food components (McHugh, 2000). The main difference between these food systems is that the EC are applied in liquid form on the food, usually by immersing the product in a solution-generating substance formed by the structural matrix (carbohydrate, protein, lipid or multicomponent mixture), and EF are first molded as solid sheets, which are then applied as a wrapping on the food product.

The envelope (packaging, wrapping or coating) plays an important role on the conservation, distribution and marketing of foodstuff. Some of its functions are to protect the product from mechanical damage, physical, chemical and microbiological activities. Some studies have recognized the importance of assessing the preformed matrix of edible films in order to quantify various parameters such as mechanical, optical and antimicrobial properties, since this envelope creates a modified atmosphere (MA) restricting the transfer of gases (O_2, CO_2) and also becoming a barrier for the transfer of aromatic compounds (Miller & Krochta, 1997).

Standard packaging technologies can be improved by the incorporation of EC or EF solutions. In a study about maize starch EC mixed with glycerol as a plasticizer and applied to Brussels sprouts (*Brassica oleracea L.* var. Gemmifera), the buds were treated with the solution, stored in polystyrene trays and covered with polyvinyl chloride (PVC) film, preserving the quality parameters regarding different factors such as weight loss, firmness, surface color of the food, commercial acceptability and nutritional quality, because the ascorbic acid content, total flavonoids and antioxidant activity remained constant during 42 days of storage at a temperature of 0 °C (Viña *et al.*, 2007).

The use of EC or EF in food applications and especially highly perishable products such as horticultural ones, is conditioned by the achievement of diverse characteristics-such as cost, availability, functional attributes, mechanical properties (flexibility, tension), optical properties (brightness and opacity), the barrier effect against gases flow, structural resistance to water and microorganisms and sensory acceptability. These characteristics are influenced by parameters such as the kind of material implemented as structural matrix (composition, molecular weight distribution), the conditions under which films are preformed (type of solvent, pH, components concentration and temperature) and the type and concentration of additives (plasticizers, cross-linking agents, antimicrobials, antioxidants or emulsifiers) (Guilbert, Gontard, & Gorris, 1996; Rojas-Grau, Soliva-Fortuny, & Martín-Belloso, 2009a).

In this review, recent trends in edible films and coatings are summarized, with emphasis on applications to the horticultural chain and their effects on fresh and minimally processed products. In addition, some biopolymers implemented in the development of new EC and EF have been reviewed, stating the importance of their optimization regarding various parameters such as mechanical properties, microbiological stability, wettability and their ability to be associated with compounds with nutraceutical properties and with various additives that improve sensory attributes in processed fruits and vegetables.

Structural matrix: carbohydrates, proteins and lipids

Edible coatings and films are usually classified according to their structural material. In this way, films and coatings are based on proteins, lipids, polysaccharides or composite. For example, a composite film may consist of lipids and hydrocolloids combined to form a bilayer or a cluster (Krochta, Baldwin, & Nisperos-Carriedo, 1994). In some recent studies the production of edible and biodegradable films by combining various polysaccharides, proteins and lipids is considered with the aim of taking advantage of the properties of each compound and the synergy between them. The mechanical and barrier properties of these films not only depend on the compounds used in the polymer matrix, but also on their compatibility (Altenhofen, Krause, & Guenter, 2009). Table 1 summarizes the main compounds used in EF and EC structural matrices, whose applications will be explained in this section.

Table 1. Summary of different compounds used in EF and EC.

Compounds	Reference
Carboxymethylcellulose, casein	Ponce *et al.*, 2008
Casein derivates with beeswax and fatty acids	Fabra *et al.*, 2009
Locust bean gum, guar gum, ethyl cellulose	Shrestha *et al.*, 2003
Mesquite gum	Bosquez-Molina *et al.*, 2010
Gelatin with glycerol, sorbitol and sucrose	Arvanitoyannis *et al.*, 1997 Sobral *et al.*, 2001
Gelatin-casein cross-linked with transglutaminase	Chambi & Grosso, 2006
Pectin	Maftoonazad *et al.*, 2007
Cassava starch	Kechichian *et al.*, 2010
Pre-gelatinized maize starch	Pagella *et al.*, 2002
Wheat gluten	Tanada-Palmu & Grosso, 2005
Sodium alginate and pectin cross-linked with $CaCl_2$	Altenhofen *et al.*, 2009
HPMC with fatty acids	Jiménez *et al.*, 2010
Beeswax	Morillon *et al.*, 2002
Carnauba wax	Shellhammer & Krochta, 1997
Chitosan	Romanazzi *et al.*, 2002 No *et al.*, 2002 Devlieghere *et al.*, 2004 Martínez-Camacho *et al.*, 2010 Aider, 2010
Chitosan-gelatin	Arvanitoyannis *et al.*, 1997
Maize starch-chitosan-glycerin	Liu *et al.*, 2009
HPMC-tea tree essential oil	Sánchez-González *et al.*, 2010
Cashew gum	Carneiro-da-Cunha *et al.*, 2009 Souza *et al.*, 2010
Galactomannans	Cerqueira *et al.*, 2009a
Galactomannans-collagen-glycerol	Lima *et al.*, 2010

The optimization of edible films composition is in one of the most important steps of the research in this field, since they must be formulated according to the properties of the fruits and vegetables to which they have to be applied (Rojas-Grau *et al.*, 2009a). Thus, it is very important to characterize and test different coating solutions on fresh and minimally processed food, since each one of them has different quality attributes to be maintained and enhanced during the storage time (Oms-Oliu, Soliva-Fortuny, & Martin-Belloso, 2008a).

Hydrocolloids (proteins and polysaccharides) are the most widely investigated biopolymers in the field of EC and EF. Some of these are: carboxymethylcellulose, casein (Ponce, Roura, del Valle, & Moreira, 2008) and its derivatives (Fabra, Jiménez, Atarés, Talens, & Chiralt, 2009), locust bean gum, guar gum, ethyl cellulose (Shrestha, Arcot, & Paterson, 2003), mesquite gum (Bosquez-Molina, Tomás, & Rodríguez-Huezo, 2010), gelatin supplemented with glycerol, sorbitol and sucrose as plasticizers (Arvanitoyannis, Psomiadou, Nakayama, Aiba, & Yamamoto, 1997; Sobral, Menegalli, Hubinger, & Roques, 2001), composite EF of gelatin-casein cross-linked with transglutaminase (Chambi & Grosso, 2006), pectin (Maftoonazad, Ramaswamy, Moalemiyan, & Kushalappa, 2007), cassava starch with natural antimicrobial compounds (Kechichian, Ditchfield, Veiga-Santos, & Tadini, 2010), pre-gelatinized standard maize starch (Pagella, Spigno, & De Faveri, 2002), wheat gluten (Tanada-Palmu & Grosso, 2005) and mixtures of sodium alginate and pectin, with the addition of $CaCl_2$ as a crosslinker material affecting mechanical properties, water solubility, moisture content, film thickness and its ability to contain calcium (Altenhofen *et al.*, 2009).

In the same way, multicomponent or composite EF have been optimized attending to its mechanical properties and transparency, looking for consumers acceptability and for the ability to withstand mechanical stress and handling during the transport. In the pursuit of these aims, the design of response surface methodology has been implemented, in order to determine the optimal mix of components that allows to take advantage of the features of the added substances (Ozdemir & Floros, 2008). However, when lipids are added for improving moisture barrier properties, other features such as transparency can be affected. As an example, hydroxypropylmethylcellulose (HPMC) has been used in combination with fatty acids to obtain composite films with lower water vapor permeability (WVP) and less transparency in comparison with the same film without lipids (Jiménez, Fabra, Talens, & Chiralt, 2010).

Polysaccharides and proteins are great materials for the formation of EC and EF, as they show excellent mechanical and structural properties, but they have a poor barrier capacity against moisture transfer. This problem is not found in lipids due to their hydrophobic properties, especially those with high melting points such as beeswax and carnauba wax (Morillon, Debeaufort, Bond, Capelle, & Volley, 2002; Shellhammer & Krochta, 1997).

To overcome the poor mechanical strength of lipid compounds, they can be used in combination with hydrophilic materials by means of the formation of an emulsion or through lamination with an hydrocolloid film lipid layer. The efficiency of an edible film against moisture transfer cannot be simply improved with the addition of hydrophobic materials in the formulation, unless the formation of a homogeneous and continuous lipid layer inside the hydrocolloid matrix is achieved (Karbowiak, Debeaufort, & Voilley, 2007; Martin-Polo, Mauguin, & Voilley, 1992). In this way, it has been found that fatty acids can form stable layers in sodium caseinate or HPMC matrices, whose properties depend on their chain length: the lower the chain lenght, the greater the layers (Fabra *et al*., 2009; Jiménez *et al*., 2010).

Emulsion-based films are less efficient in controlling water transfer than bilayer films, as a homogeneous distribution of lipids is not achieved. However, they exhibit good mechanical strength and require a simple process for their manufacture and application, whereas multilayer films require a complex set of operations that depend on the number of coatings. It has been proved, in emulsion-based films, that the smaller the particle size or lipid globules and the more homogeneously distributed, the lower WVP (Debeaufort & Voilley, 1995; McHugh & Krochta, 1994; Pérez-Gago & Krochta, 2001). However, its permeability to water vapor can be similar to the values presented by the films based on proteins or polysaccharides (Morillon *et al*., 2002).

Among polysaccharides, bioactive compounds such as chitosan and its derivatives show a great number of applications focused on active coating systems, in view of the increasing concern about the production of poorly biodegradable plastic materials. Chitosan has a vast potential that can be applied in the food industry because of its particular physicochemical properties such as biodegradability, biocompatibility with human tissues, null toxicity and especially its antimicrobial and antifungal properties (Aider, 2010). In addition to research based on its antimicrobial properties, some aspects such as mechanical and thermal properties and permeability to gases (O_2, CO_2) have been quantified, revealing that chitosan-gelatin films plasticized with water and polyols suffer an increase in permeability as the amount of plasticizers in their formulation is increased (Arvanitoyannis *et al*., 1997).

Chitosan is a polysaccharide obtained by deacetylation of chitin, which is extracted from the exoskeleton of crustaceans and fungal cell walls. It has been extensively used in films and coatings due to its ability to inhibit the growth of various bacteria and fungal pathogens (Romanazzi, Nigro, Ippolito, Di Venere, & Salerno, 2002). Chitosan has also been studied in combination with other biopolymers. Films composed of maize starch-chitosan plasticized with glycerin have shown improved mechanical properties (such as elongation at break) and water vapor permeability in contrast to membranes developed with only one of these structural components, as a result of interactions between the hydroxyl groups of starch and the amino groups of chitosan. Its antibacterial activity has been proved observing inhibition zones by disk diffusion on agar containing *Escherichia coli* O157:H7 (Liu, Qin, He, & Song, 2009). New research and recent reviews on the use of chitosan gather some information on the effect of the deacetylation degree on its antimicrobial activity, its use in active coating and its interaction with other components of the treated food products (Aider, 2010; Devlieghere, Vermeulen, & Debevere, 2004; Martínez-Camacho *et al*., 2010; No, Park, Lee, & Meyers, 2002). Besides, EF have been formulated by mixing chitosan with essential oils. Sánchez-González, González-Martínez, Chiralt, and Cháfer (2010) found that chitosan-tea tree essential oil based films were effective against *Listeria monocytogenes*.

Other very interesting hydrocolloids are the gum exuded from the cashew tree (*Anacardium occidentale L*), known as cashew gum, and galactomannans. First edible films based on cashew gum have been evaluated, testing its mechanical properties, wettability, surface tension, opacity, tensile strength, elongation at break and water vapor permeability, in order to obtain biopolymer structures able to generate edible coatings applied to minimally processed fruits. In addition, properties such as wettability and surface tension were quantified by using it as a coating on Golden apples. As a result, it was found that concentrations below 1.5% w/v create fragile films; the addition of Tween80 reduced cohesive forces and therefore decreased surface tension, increasing wettability of the coating solution and thereby improving the compatibility of the EC with the fruit surface (Carneiro-da-Cunha *et al*., 2009). Edible films based on cashew gum were also tested in mango (*Mangifera indica* var. Tommy Atkins) with the aim of determining its effect on the shelf life of refrigerated fresh product. It was determined that it acts as a barrier to mass transport, reducing weight loss as a result of respiration processes (Souza *et al*., 2010).

Galactomannans are hydrocolloids that deserve some interest due to their contribution to strengthen matrix structures. They are stored as reserve polysaccharides and extracted from seeds. Their polymeric structure is mainly influenced by the proportion of mannose/galactose units and the distribution of galactose residues in the main chain

(Cerqueira *et al*., 2009a). *Adenanthera pavonina* and *Caesalpinia pulcherrima*, two plants belonging to the legume family, were recently used to develop coatings from new sources of galactomannans (Lima *et al*., 2010).

In an exhaustive study carried out by Lima *et al*. (2010), different proportions of galactomannans, collagen and glycerol were prepared and tested in order to design possible mixtures with a high degree of wettability, this is having the ability to be easily adhered and homogeneously distributed in mango and apple fruits. With the assayed products and conditions (the films were maintained at 20 °C and 50% relative humidity), it was determined that the best mixes for mango and apple are: 0.5% of galactomannan from *A. pavonina*, 1.5% collagen and 1.5% glycerol, or 0.5% of galactomannan from *A. pavonina*, 1.5% collagen without the addition of glycerol. A lower use of O_2 (28%) and a lower production of CO_2 (11.0%) was achieved in coated mango compared to the control samples (without coating). In apples, the production and consumption of O_2 and CO_2 was approximately 50% lower in the presence of the coating. These results suggest that the galactomannan-based coatings can reduce gas-transfer and thus become useful tools to extend the shelf life of these fruits.

Edible films and coatings and their role as active packages

The development of coatings based on polysaccharides has brought a significant increase in their applications and in the amount of products that can be treated, extending the shelf life of fruits and vegetables due to the selective permeability of these polymers to O_2 and CO_2. Table 2 summarizes some of these compounds and their effects. These polysaccharide-based coatings can be used to modify the internal atmosphere of fruits, delaying senescence (Rojas-Grau *et al*., 2009a). Edible coatings create a passive modified atmosphere, which can influence various changes in fresh and minimally processed foodstuff in some areas such as: antioxidant properties, color, firmness, sensory quality, microbial growth inhibition, ethylene production and volatile compounds as a result of anaerobic processes (Oms-Oliu, Soliva-Fortuny, & Martin-Belloso, 2008b).

The effectiveness of an edible coating to protect fruits and vegetables depends on the control of wettability (Cerqueira, Lima, Teixeira, Moreira, & Vicente, 2009b), on the film ability to maintain the functionality of some compounds (plasticizers, antimicrobials, antioxidants) within the matrix, as the loss of these molecules affects the thickness of the film (Park, 1999), and the solubility in water as it is necessary to avoid the dissolution of the coating (Ozdemir & Floros, 2008).

Although some EF have been successfully applied to fresh products, other applications adversely affected quality. The modification of the internal atmosphere through the use of edible coatings can increase disorders associated with a high concentration of CO_2 or low O_2 (Ben-Yehoshua, 1969). In fresh-cut melon coated with gellan gum a growing increase of phenolic compounds was quantified in response to stress generated by excessive change in the atmosphere of the minimally processed fruit during storage. Although the generation of these substances (phenols) contributed to the antioxidant power, sensory properties such as odor, color and flavor were affected. Translucent tissue was also observed, which appeared to be a symptom of senescence (Oms-Oliu *et al*., 2008a). When a gas barrier is created, an increase in the presence of some volatiles associated with anaerobic conditions can be induced. This is the case of ethanol and acetaldehyde, which were detected after two weeks of storage in apple slices treated with alginate and gellan gum EC. The production of these substances is related to anaerobic fermentation, to a decrease of sensory quality and especially to the loss of minimally processed fruit flavors (Rojas-Grau, Tapia, & Martín-Belloso, 2008). Therefore, it is clear that the control of gas permeability should be a priority in the development and study of active coatings (Parra, Tadini, Ponce, & Lugão, 2004).

Edible films and coatings with antimicrobial properties have innovated the concept of active packaging, being developed to reduce, inhibit or stop the growth of

Table 2. Summary of different components of EF and EC used as active packages.

Components	Effect	Reference
Gellan gum	Increase of phenolics	Ben-Yehoshua, 1969
Alginate and gellan gum	Gas permeability modification	Rojas-Grau *et al*., 2008
Sorbic acid, benzoic acid, sodium benzoate, citric acid	Antimicrobial	Quintavalla & Vicini, 2002
Potassium sorbate	Antimicrobial	Ozdemir & Floros, 2008
Nicines, pediocin	Antimicrobial	Sebti & Coma, 2002
Natamycin in a chitosan matrix	Antimicrobial	Durango *et al*., 2006 Ribeiro *et al*., 2007 Fajardo *et al*., 2010 Maqbool *et al*., 2010
Tea tree essential oil in HPMC matrix	Antimicrobial	Sánchez-González *et al*., 2009
Chitosan	Antimicrobial	El Ghaouth *et al*., 1992 Coma *et al*., 2002 Ponce *et al*., 2008 Kyu Kyu *et al*., 2007 Maqbool *et al*., 2010
Chitosan	Shelf life extension	Lazaridou & Biliaderis, 2002 Geraldine *et al*., 2008 Márquez *et al*., 2009
Chitosan-oleic acid	Shelf life extension	Vargas *et al*., 2006
Chitosan	Tissue firmness conservation	El Gaouth *et al*., 1997
Chitosan	Respiration rate reduction	Li & Yu, 2000
Chitosan	Fungistatic	Martínez-Camacho *et al*., 2010
Essential oils	Antimicrobial and antioxidant	Atarés *et al*., 2010 Sánchez-González *et al*., 2010

microorganisms on food surfaces (Appendini & Hotchkiss, 2002). In most fresh or processed products microbial contamination is found with the highest intensity on their surface. Therefore, an effective system to control the growth of that biota is required (Padgett, Han, & Dawson, 1998). Traditionally, antimicrobial agents are directly added to foods, but their activity can be inhibited by different components of these products, decreasing its efficiency. In such cases, the implementation of films or coatings can be more efficient that antimicrobial additives used in the foodstuff, since they can migrate selectively and gradually from the wrapping compounds to the surface of the food (Ouattara, Simard, Piette, Bégin, & Holley, 2000).

Antimicrobial EC and EF have been shown to be an efficient alternative in the control of food contamination. Spoilage and pathogens can be reduced by incorporating antimicrobial agents into edible films and coatings (Sorrentino, Gorrasi, & Vittoria, 2007). Some of these compounds included into EF and EC are sorbic acid, benzoic acid, sodium benzoate, citric acid (Quintavalla & Vicini, 2002), potassium sorbate (Ozdemir & Floros, 2008), and bacteriocins such as nicin or pediocin (Sebti & Coma, 2002), or even natamycin in a chitosan EC, which had the ability to release the compound and synergistically prevent the growth of molds and yeasts (Durango, Soares, & Andrade, 2006; Fajardo *et al*., 2010; Maqbool, Ali, Ramachandran, Smith, & Alderson, 2010; Ribeiro, Vicente, Teixeira, & Miranda, 2007). Additionally, hydrophobic compounds such as tea tree essential oil in HPMC based films have also been used (Sánchez-González, Vargas, González-Martínez, Chiralt, & Cháfer, 2009).

As it has been already mentioned, chitosan is a polysaccharide that has been used in films and coatings due to its ability to inhibit the growth of various microbial pathogens. In some fungi, chitosan can cause alterations in membrane function, through its strong interaction with the electronegative surface charge, leading to permeability changes, metabolic disturbances and even death (Fang, Li, & Shih, 1994). According to Muzzarelli *et al*. (1990), the antimicrobial activity of chitosan against bacteria could be due to the polycation nature of the molecule, which enables interaction and formation of polyelectrolyte complexes with acidic polymers produced in the surface of the bacterial cell (as lipopolysaccharides or teichoic acid). Chitosan-based coatings and films tested on *L. monocytogenes* showed inhibitory effect on the growth of this bacteria (Coma *et al*., 2002; Ponce *et al*., 2008). Other studies have shown that chitosan-based coatings have the potential to increase the shelf life of fruits and vegetables by inhibiting the growth of microorganisms, reducing ethylene production, increasing the concentration of carbon dioxide and reducing oxygen levels (Geraldine, Ferreira, Alvarenga, & Almeida, 2008; Lazaridou & Biliaderis, 2002; Márquez, Cartagena, & Pérez-Gago, 2009). Furthermore, chitosan-oleic acid based coatings are able to increase significantly the shelf life of cold-stored strawberries as it have been studied by Vargas, Albors, Chiralt, and González-Martínez (2006).

This hydrocolloid (chitosan) has the ability to slow the growth of certain microorganisms that are deleterious in fruit postharvest such as *Fusarium* spp., *Colletotrichum musae* and *Lasiodiplodia theobromae* in banana (*Musa acuminate* L. Var. Kluai Hom Thong) (Kyu Kyu, Jitareerat, Kanlayanarat, & Sangchote, 2007; Maqbool *et al*., 2010), or *Botrytis cinerea* on pepper (*Capsicum annuum* L. Var. Bellboy). In this case, mold suffered cell damage in invading hyphae and reduced the production of polygalacturonase, which has an effect in maintaining the firmness of the tissues (El Gaouth, Arul, Wilson, & Benhamou, 1997). Its activity has also been reported in fruits of peach (*Prunus persica* L. Batsch.), reducing the respiration rate represented in the production of CO_2 and maintaining the firmness of the fruit covered until the end of 12 days of storage at a temperature of 23 °C (Li & Yu, 2000). Moreover, El Ghaouth, Ponnampalam, Castaigne, and Arul (1992) showed that the coatings with chitosan content between 1% and 2% reduced the incidence of deterioration in tomato mainly caused by *Botrytis cinerea*. In addition, some studies suggest that chitosan shows fungistatic activity even if it is used inside a preformed film matrix. Some factors such as storage temperature and changes in the mechanical and barrier properties influenced by additives and other types of antimicrobials can promote antimicrobial effect of these films (Martínez-Camacho *et al*., 2010).

Nowadays edible films have different applications, and their use is expected to be expanded with the development of Active Coating Systems. This second generation of coating materials can use chemicals, enzymes or microorganisms that prevent, for example, microbial growth or lipids oxidation in coated food products. In this sense essential oils, in combination with structural polymers, can be a promising source since different pieces of work have constituted the evidence of their effectiveness as antimicrobial and antioxidant compounds (Atarés, Bonilla, & Chiralt, 2010; Sánchez-González *et al*., 2010). Coatings of second generation may contain nutrients or other bioactive compounds that have a positive effect on health, especially due to the application of new microencapsulation or nanoencapsulation techniques. In this way, coating materials would act as carriers of these bioactive compounds to be transported to target sites such as the intestine without losing its activity, being within a matrix during its passage through the gastrointestinal tract (Korhonen, 2005).

Effect of edible films and coatings in food browning and polyphenol oxidase activity

In food products, not only microbiological stability plays an indispensable role in its quality, but also sensory aspects are essential to ensure that the application of emerging technologies such as edible films and coatings become successful (Rojas-Grau, Oms-Oliu, Soliva-Fortuny, & Martín-Belloso, 2009b). Thus, color is one of the most important parameters that must be controlled, and enzymatic browning is the main process that modifies it. Polyphenol oxidase (PPO) is the main enzyme responsible for these changes in vegetable tissues that contain phenolic or polyphenolic molecules. It catalyzes the *o*-hydroxylation of monophenols to *o*-diphenols (monooxygenase or cresolase activity) and the subsequent oxidation of*o*-diphenols to o-quinones (diphenolase or catecholase activity). Later polymerization of these compounds leads to the formation of an heterogeneous group of melanins (Falguera, Gatius, Pagan, & Ibarz, 2010).

Some researchers have proved the effectiveness of edible films and coatings on the control of browning processes and polyphenol oxidase activity. Vangnai, Wongs-Aree, Nimitkeatkai, and Kanlayanarat (2006) applied chitosan coatings on "Daw" longan (*Dimocarpus longan* Lour.) fruits, finding that these treatments reduced increasing activities of PPO during the 20 days of storage at 4 °C, slightly reducing pericarp browning. Chitosan coatings were also used by Eissa (2008), who found that they delayed discoloration associated with reduced enzyme activity of PPO and other enzymes, and had a good effect on the evolution of colour characteristics and parameters of fresh-cut mushroom during storage at 4 °C. Ponce *et al.* (2008) applied chitosan films enriched with olive and rosemary oleoresins on pumpkin (*Cucurbita moschata* Duch) slices, which showed a clear antioxidant effect by slowing the action of polyphenol oxidase (PPO) and peroxidase (POD) within five days of storage. In addition, these edible coatings showed no deleterious effects on the sensory acceptability of the pumpkin juice.

Hui-Min, To, Li-Ping, and Hai-Ying (2009) investigated the effects of three kinds of edible coatings (carrageenan, carboxymethyl cellulose (CMC) and sodium alginate) and their combinations on browning parameters of fresh-cut peach (*Prunus persica*) fruits during storage at 5 °C. Sodium alginate coating and the various composite ones reduced the declines of Hunter L^* value and the increases of Hunter a^* and Hunter b^* values, inhibited PPO activity and reduced the browning degree of peach fruits.

Zhang, Xiao, Luo, Peng, and Salokhe (2004) applied combinations of an ozone water treatment and different coatings on minimally processed cucumber (*Cucumis sativus* L.). The study showed that a concentration of 4.2 mg m^{-3} ozone and a composite coating made of polyvinyl alcohol 134 (1%), chitosan (1%), lithium chloride (0.5%), glacial acetic acid (2.5%) and sodium benzoate (0.05%) inhibits respiration, chlorophyll breakdown and polyphenol oxidase activity.

Edible films as a matrix of nanobiocomposites

Different methods to improve the properties of biopolymer-based films such as lipids or antimicrobial components addition have been mentioned. In addition, a novel technique based on the use of very small particles has become remarkable in food developments recently.

Nowadays nanotechnology is applied with great results in many research areas. One of these fields of application is polymer research. A nanoparticle is an ultrafine particle in the nanometer size order (Hosokawa, Nogi, Makio, & Yokoyama, 2008), which is able to form nanobiocomposite films when it is combined with natural polymers. The research and development of nanobiocomposite materials for food applications is expected to grow with the advent of new polymeric materials with inorganic nanoparticles, although it is not widely widespread yet (Restuccia *et al.*, 2010; Sorrentino *et al.*, 2007). Some of the applications associated with nanotechnology include improved taste, color, flavor, texture and consistency of foodstuffs, increased absorption and bioavailability of food or food ingredients (nutrients), and the development of new food-packaging materials with improved mechanical, barrier and antimicrobial properties (Restuccia *et al.*, 2010).

Traditionally, mineral fillers such as clay, silica and talc have been incorporated in film preparation in the range of 10–50% w/w in order to reduce its cost or to improve its performance in some way (Rhim & Ng, 2007). Thus, the most important nanoparticles that have been used to provide enhanced properties to edible films are clays. According to Rhim and Ng (2007), the nanometer-size dispersion of polymer-clay nanocomposites exhibit the large-scale improvement in the mechanical and physical properties compared with pure polymer or conventional composites. Both proteins (Shotornvit, Rhim, & Hong, 2009) and polysaccharides (Casariego *et al.*, 2009; Tang, Alavi, & Herald, 2008) have given rise to films in combination with nano-clay particles. However, other nanoparticles such as tripolyphosphate-chitosan (De Moura *et al.*, 2009), microcrystalline cellulose (Bilbao-Sáinz, Avena-Bustillos, Wood, Williams, & McHugh, 2010) and silicon dioxide (Tang, Xiong, Tang, & Zou, 2009) have also been added to biopolymers to obtain films. These nanoparticles are able to improve moisture barrier properties (Casariego *et al.*, 2009; De Moura *et al.*, 2009; Shotornvit *et al.*, 2009) and restrict microbial growth (Shotornvit *et al.*, 2009). In this way, Rhim, Hong, Park, and Perry (2006) found that the use of nanoparticles has a potential application in the development of natural biopolymer-based biodegradable packaging materials. In this study different nanoparticles improved the physical properties of chitosan-based films as well as showed promising antimicrobial activity. Regarding optical properties, these were more or less affected depending on the nano-clay type as it has been observed in isolated whey protein based films (Shotornvit *et al.*, 2009). In addition, nanoparticles have also been added to conventional polymers such as EVOH (Cabedo, Giménez, Lagarón, Gavara, & Saura, 2004) or PP/HDPE (Chiu, Yen, & Lee, 2010).

Trends in the use of edible films and coatings

The properties that have been reviewed have given edible films and coatings several uses. Nowadays, some of the research lines involving these active envelopes include oil consumption reduction in deep-fat fried products, transport of bioactive compounds and shelf life extension of highly perishable products.

Oil consumption reduction in deep-fat fried products

Deep-fat frying is a widely used method in the preparation of tasty food with an attractive appearance. The tenderness and humidity of the inner part of these products combined with a porous crunchy crust provides an increase in palatability that is responsible for their great acceptance. However, fried foods have a significant fat content, reaching, in some cases, 1/3 of the total weight of the product. The development of more acceptable products for consumers, who are increasingly more conscious and concerned about their health, has led to the need to reduce oil incorporation during the frying process (Freitas *et al.*, 2009). Some hydrocolloids with thermal gelation or thickening properties, such as proteins and carbohydrates, have been tested on the migration of oil and water (Debeaufort & Voilley, 1997; Williams & Mittal, 1999). Various coating options are being studied for the reduction of oil incorporation during frying, such as alginate, cellulose and its derivatives, soy protein isolate, whey protein, albumin, corn, gluten and pectin (Albert & Mittal, 2002; Khalil, 1999; Mallikarjunan *et al.*, 1997; Mellema, 2003; Salvador, Sanz, & Fiszman, 2005).

Research with mashed potato spheres coated with zein, hydroxypropylmethylcellulose (HPMC) and methylcellulose (MC) has reported a decrease in food moisture of 14.9, 21.9 and 31.1% and in fat consumption of 59.0, 61.4, and 83.6%, respectively (Mallikarjuna, Chinnan, Balasubramaniam, & Phillips, 1997). Other studies have also shown that MC films have better barrier properties against fat absorption than hydroxypropylcellulose (HPC) and gellan gum ones (Williams & Mittal, 1999).

García, Ferrero, Bértola, Martino, and Zaritzky (2002) used MC and HPMC in the formulation of coatings applied to potato chips (0.7 $\times$ 0.7 $\times$ 5.0 cm) and wheat flour discs (3.7 cm diameter $\times$ 0.3 cm high), which were submerged in the coating suspension for 10 s and fried immediately. The most effective coatings were 1% MC and 0.75% sorbitol for wheat flour discs and 1% MC and 0.5% sorbitol for potato chips, reducing the oil consumption to 35.2% and 40.6% respectively. The use of coatings did not have a significant impact on the sensory quality, according to the group of panelists.

Albert and Mittal (2002) carried out an extense piece of work comparing eleven hydrocolloid materials including gelatine, gellan gum, k-carrageenan-konjac-blend, locust bean gum, methyl cellulose (MC), microcrystalline cellulose, three types of pectin, sodium caseinate, soy protein isolate (SPI), vital wheat gluten and whey protein isolate (WPI), as well as some composite films made of different combinations of these compounds. Two of them, SPI/MC and SPI/WPI composite coatings, provided the highest index *reduction in fat uptake/decrease of water loss* value, and reduced the fat uptake up to 99.8%.

Singthong and Thonkaew (2009) investigated the influence of sodium alginate, carboxyl methyl cellulose (CMC) and pectin on the oil absorption in banana chips. The uncoated control sample had an oil consumption of 40 g/100 g sample, while lower values were obtained for banana chips blanched in 0.5% $CaCl_2$ and treated with a coating matrix of 1% pectin or 1% CMC. Using these two coatings oil consumption was reduced to 22.89 and 22.90 g/100 g of sample, respectively.

Freitas *et al.* (2009) investigated the effect of edible coatings from pectin, whey protein and soy protein isolate in the deep-fat frying of preformed products made of cassava flour and cassava puree. Whey protein coating was the most effective one regarding fat absorption due to its thickness, achieveing a 27% reduction.

Otherwise, EC can improve the crispness of fried products by reducing the moisture diffusion between fish meat and the crust during reheating in a microwave as it have been observed by Chen *et al.* (2008). This experience was based in the thermal gelation of HPMC which occurs at high temperature.

Transport of bioactive compounds

Consumers require fresh and minimally processed foods that are exempt from chemically synthesized substances, and look for those enriched with natural substances that bring health benefits and maintain nutritional and sensory characteristics (Falguera, Pagan, & Ibarz, 2011). Therefore, in recent times the efforts of researchers have been focused on searching for new naturally occurring substances that act as possible alternative sources of antioxidants and antimicrobials (Ponce *et al.*, 2008).

Rojas-Grau, Tapia, Rodríguez, Carmona, and Martin-Belloso (2007) proved the ability of edible coatings based on sodium alginate and gellan gum to transport N-acetylcysteine and glutathione as antibrowning agents, besides the positive effect of the addition of vegetable oils in these edible coatings to increase resistance to water vapor transport in minimally processed fruits of Fuji apple. Moreover, it was also stated that the coatings were able to keep the vegetable oil enriched with essential fatty acids ($\omega3$ and $\omega6$) encapsulated.

Biodegradable films based on cassava starch (*Manihot esculenta* Crantz) have been characterized from some points of view, including mechanical properties, the effect of various plasticizers such as glycerol and polyethylene glycol and cross-linkers as glutaraldehyde or $CaCl_2$ on water vapor transmission, and their possible use in the food industry because this hydrocolloid is abundant and cheap (Parra *et al.*, 2004; Ribeiro *et al.*, 2007). At present, studies have been guided to the ability of these films to transport natural antimicrobial agents such as chitosan (Vásconez, Flores, Campos, Alvarado, & Gerschenson, 2009).

As it has been already introduced, sensory aspects are very important in the evaluation of films and coatings applications. In order to slow changes in flavor during food storage, the encapsulation of aromatic compounds has been implemented as a possible strategy to reduce the effect of degrading reactions such as oxidation. Marcuzzo, Sensidoni, Debeaufort, and Voilley (2010) encapsulated 10 different aromatic compounds in carrageenan films, including ethyl acetate, ethyl butirate, ethyl isobutyrate, ethyl hexanoate, ethyl octanoate, 2-pentanone, 2-heptanone, 2-octanone, 2-nonanone and 1-hexanol. Carrageenan films were appropriate to conduct these experiments because they show high affinity for polar volatile compounds. These EF may achieve the aim of gradually releasing aroma compounds and thereby maintain the sensory characteristics such as aroma and taste for certain periods of time.Furthermore, Hambleton, Debeaufort, Bonnotte, and Voilley (2009) proved that matrices made of other polysaccharides such as alginate are able to protect an encapsulated aroma compound (n-hexanal), due to its low oxygen permeability.

On the one hand, according to the mentioned studies it can be concluded that polysaccharide matrices are able to encapsulate aroma compounds in order to maintain the organoleptic quality in food systems. On the other hand, proteins have been less studied as protective polymers for aroma components, maybe due to its minor effectivity for this purpose. In this sense, Monedero *et al*. (2010) found that it was necessary to add beeswax to improve the capacity of soy protein isolate based films to retain n-hexanal.

The transport and release of various active compounds (antioxidants, flavorings, antibrowning and antimicrobial compounds, vitamins or enzymes) is one of the most important aspects within the features of edible films and coatings. Nowadays, trends in research consider the use of nanotechnology solutions, previously reviewed, using encapsulated nanoparticles of functional and bioactive compounds, which can be released from the matrix that contain them in a controlled pace (Rojas-Grau *et al*., 2009a).

Shelf life extension of highly perishable products

One of the most important uses of edible films and coatings is focused on the shelf life extension of horticultural products. Consequently, there are many pieces of work investigating the application of different coatings on different foodstuff, some of which are reviewed in this section.

Ribeiro *et al*. (2007) studied the ability of edible coatings based on polysaccharides (starch, carrageenan and chitosan) to extend the shelf life of strawberry fruits (*Fragaria ananasa* cv. Camarosa) and its possible industrial application. The best wettability was achieved with combinations of 2.0% starch and 2.0% sorbitol, 0.3% carrageenan, 0.75% glycerol and 0.02% Tween 80 or 1.0% chitosan and 0.1% Tween 80. The oxygen permeability of carrageenan films was approximately 40.0% of the value obtained with starch ones. The values of the fruit firmness loss were the lowest ones in carrageenan films with added calcium chloride. The minimum mass loss was achieved in edible coatings based on carrageenan and chitosan with added calcium chloride. The lower microbial growth rate was observed in strawberries coated with chitosan and calcium chloride.

Carrot is one of the most popular vegetables, but its marketing is limited by its rapid deterioration during storage, mainly due to physiological changes that reduce its shelf life. The product suffers a loss of firmness, with the production and release of a characteristic odor generated by anaerobic catabolism, due to high respiration rate and microbial spoilage (Barry-Ryan, Pacussi, & O'Beirne, 2000). Durango *et al*. (2006) developed coatings based on yam (*Dioscorea* sp.) starch and chitosan. The maximum antimicrobial activity was obtained in the EC containing 1.5% of chitosan, which was completely effective on the growth of molds and yeasts reducing the count by 2.5 log units in the carrot sticks that were stored for 15 days. Coating with a chitosan concentration of 0.5% controlled the growth of molds and yeasts for the first 5 days of storage. After this time, tested samples generated a count similar to the one of the control sample. Thus, the use of antimicrobial coatings based on chitosan and yam starch significantly inhibited the growth of lactic acid bacteria, total coliforms, psychrotrophic microorganisms, mesophilic aerobes, molds and yeasts. Subsequently, Pastor, Sánchez-González, Cháfer, Chiralt, and González-Martínez (2010) obtained films based on HPMC and ethanolic extract of propolis which are effectively against *Aspergillus niger* and *Penicillium italicum*. These films appeared yellowish, which can restrict their use on different foodstuff but not in carrots or oranges, where their shade would mask films colour.

Maqbool *et al*. (2010) applied edible coatings based on arabic gum, 95% deacetylated chitosan and arabic gum + chitosan composite films on fresh banana fruits, in order to determine their potential in the control of *Colletotrichum musae*. This fungus causes anthracnose, a disease that affects postharvest quality in transport and storage of bananas. *In vivo* tests determined that composite EC formed by 10% arabic gum and 1% chitosan was the best treatment, because it had the lowest disease incidence (16%). In addition, the composite film reduced the percentage of weight loss, retained fruit firmness during and after storage and marketing conditions compared to control samples, minimizing moisture loss. Arabic gum + chitosan edible coating showed a synergistic behavior that allowed maintaining sensory quality and microbiological parameters, without phytotoxic effects on bananas stored for 33 days.

Edible films and coatings: commercial and regulatory aspects

The commercial use of edible films has been limited due to problems related to their poor mechanical and barrier properties when compared to synthetic polymers (Azeredo *et al*., 2009). As it has been explained, several nanocomposites have been developed by adding reinforcing compounds (nanofillers) to biopolymers, improving their properties and enhancing their cost-price-efficiency (Sorrentino *et al*., 2007). However, there are many safety concerns about nanomaterials, as their size may allow them to penetrate into cells and eventually remain in the human organism. While the properties and safety of the materials in their bulk form are usually well known, the nano-sized counterparts frequently exhibit different properties from those found at the macro-scale, and there is limited scientific data about their eventual toxicological effects. So the need for accurate information on the effects of nanomaterials on human health following chronic exposure is imperative before any nanostructured food packaging is available for commercialization.

Anyway, several authors have stated that the use of edible films and coatings is expected to grow, in part due to the growing trend for individualized portion size, which has made packaging-per-unit to increase. In addition, their functions fall entirely into "green-packaging" applications, such as the US EPA suggested plan for improved municipal waste management and reduction (Dangaran, Tomasula, & Qi, 2009). In order to reduce the initial amount of packaging, the EPA suggests designing packaging systems that reduce the amount of environmentally toxic materials used in packaging to make it easier to reuse or compost them. They also suggest packaging that reduces the amount of damage or spoilage to food products, increasing their shelf life. Edible films and coatings fit both criteria.

In Europe, the European framework regulation (2004/1935/EC) authorizes the concept of active packaging with intentional active agents' release (Guillart *et al*., 2009). With the formation of the European Union, legislation of all member states was harmonized in order to create a single market and overcome barriers to trade. So far, the EU legislation on materials in contact with food products has protected the health of consumers by ensuring that no material in contact with foodstuffs can bring about a chemical reaction that would change their composition or organoleptic properties. Regulation 1935/2004/EC repeals this legislation in order to allow packaging to benefit from technological innovation. This was necessary in the EU because all packaging materials (including those that intentionally add substances to food) are subject to all requirements for food-contact materials, including the overall migration limits (OMLs) and specific migration limits (SMLs) (Restuccia *et al*., 2010).

Regarding the compounds that can be incorporated into edible coating formulations, these ingredients are majorly regarded as food additives and are listed within the list of additives for general purposes, although pectins, Acacia and karaya gums, beeswax, polysorbates, fatty acids, and lecithin are mentioned apart for coating applications. The use of these coating forming substances is permitted provided that the 'quantum satis' principle is observed (Rojas-Grau *et al*., 2009a). In addition, the Directive 2008/84/EC introduces specific purity criteria for food additives. Since edible coatings could have ingredients with a functional effect, inclusion of these compounds should be mentioned on the label.

Conclusions

Edible films and coatings applied to fresh, minimally processed and processed fruits and vegetables are effective in extending their shelf life, maintaining their microbiological, sensory and nutritional quality. Some formulations have been specifically tested on their ability to inhibit polyphenol oxidase activity and delay browning reactions. In addition, EF and EC are able to transport substances that bring some benefits not only for food itself but also for the consumer, through the encapsulation of bioactive compounds, developing new products with nutraceutical or functional effect.

The most important properties to be evaluated in an edible coating are its microbiological stability, adhesion, cohesion, wettability, solubility, transparency, mechanical properties, sensory and permeability to water vapor and gases. Knowing these properties, their composition and behavior may be predicted and optimized.

Nowadays, trends in the use of these active envelopes include oil consumption reduction in deep-fat fried products, transport of bioactive compounds and shelf life extension of highly perishable products. Thus, research in this field aims at the characterization of new hydrocolloid films based on non-conventional sources, as well as at the determination of the ability of these compounds to release molecules with specific functions such as vitamins, antioxidants, natural colors, flavors, aromatics and assess the interactions that can provide these molecules with the encapsulation matrix.

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