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Edible films and coatings containing bioactives

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Edible films and coatings are promising systems to be used as active ingredient carriers. As they can be consider both a packaging and a food component, they have to fulfill specific requirements: good sensory attributes, high barrier and mechanical properties, biochemical, physicochemical, and microbial stability, safety, non-polluting nature, simple technology and low raw material and processing cost. The addition of bioactive compounds and additives contributes to achieve these requirements to preserve the quality, safety and sensory properties of foods. Research efforts are focused in attempting to optimize the composition of film and coatings materials in order to be well-processed and effective in real, pragmatic, everyday food systems. This short review attempts to summarize the current state of knowledge on this subject, including general aspects, new trends and some successful applications.

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Edible films and coatings

Edible coatings have been used for centuries to protect foods and prevent moisture loss. These practices were accepted long before their associated chemistries were understood [1]. Some records indicate that citrus fruits were waxed already in China in the 12th century, and later lard or fats were used to prolong shelf life of meat products in England. In the early to mid-twentieth century, coatings were used to prevent water loss and add shine to fruits and vegetables, as casings for sausages, and as some sort of sugary coating on confectionaries, including chocolate. And since these simple early attempts the applications of food coatings have been refined [2•].

When a packaging material such as a film, a thin layer or a coating, is an integral part of a food and may be eaten with the food, it qualifies as 'edible' packaging. Coatings are either applied to or formed directly on foods, while films are self-supporting structures that can be used to wrap food products. They can be located either on the food surface or between different components of a food product (see Figure 1) [3].

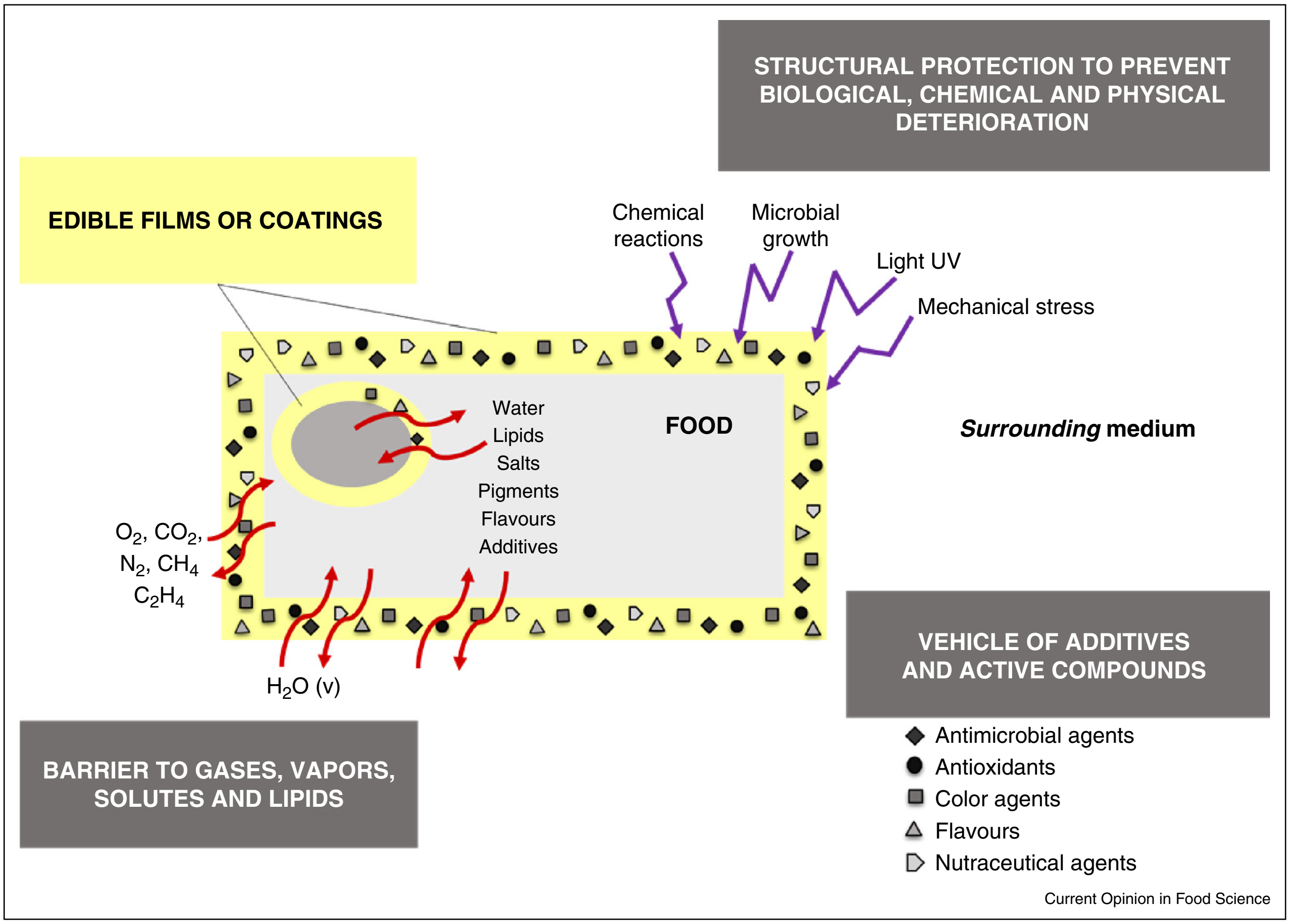
Edible films and coatings should be produced mainly from edible biopolymers and food-grade additives. They should be safe under intended use conditions and processed and applied in accordance with good manufacturing practices. In the USA, food-safe materials should have the Food and Drug Administration's approval or at least acquire GRAS status [4].

These materials enhance the quality of food products by protecting them from physical, chemical and biological deterioration, which results in an extended shelf life and improved safety. The most common functions of edible films and coatings are described in Figure 1. They should act as barriers against gas or vapors, oils and solutes, provide structural protection to prevent mechanical damage during transportation, handling and display, and protect food against oxidation, microbial growth and other chemical reactions. They also could be used to enhance the visual and tactile features of food products and to carry active substances such as antioxidants, antimicrobial, nutraceuticals, colors and flavors [5].

Edible films and coatings also are biodegradable, which means that can be completely degraded by microorganisms in a composting process ultimately to only carbon dioxide, water, methane, and some biomass residues [6]. The challenge to biodegradable films and coatings for food packaging and other uses is that they must serve its function safely and effectively during the required time and only after the intended functional use has ended biodegradation should proceed. Their use in specific applications — especially those of short service life, such as food packaging or agriculture uses — could contribute to resolve the environmental problems caused by the accumulation of nonrenewable and non-biodegradable synthetic materials (mainly derived from petroleum), widely used [7•].

So, as they can be considered both a packaging and a food component, edible films and coatings have to fulfill some specific requirements: (i) good sensory attributes, (ii) high barrier and mechanical properties, (iii) biochemical,

Figure 1



Main functions of edible films and coatings.

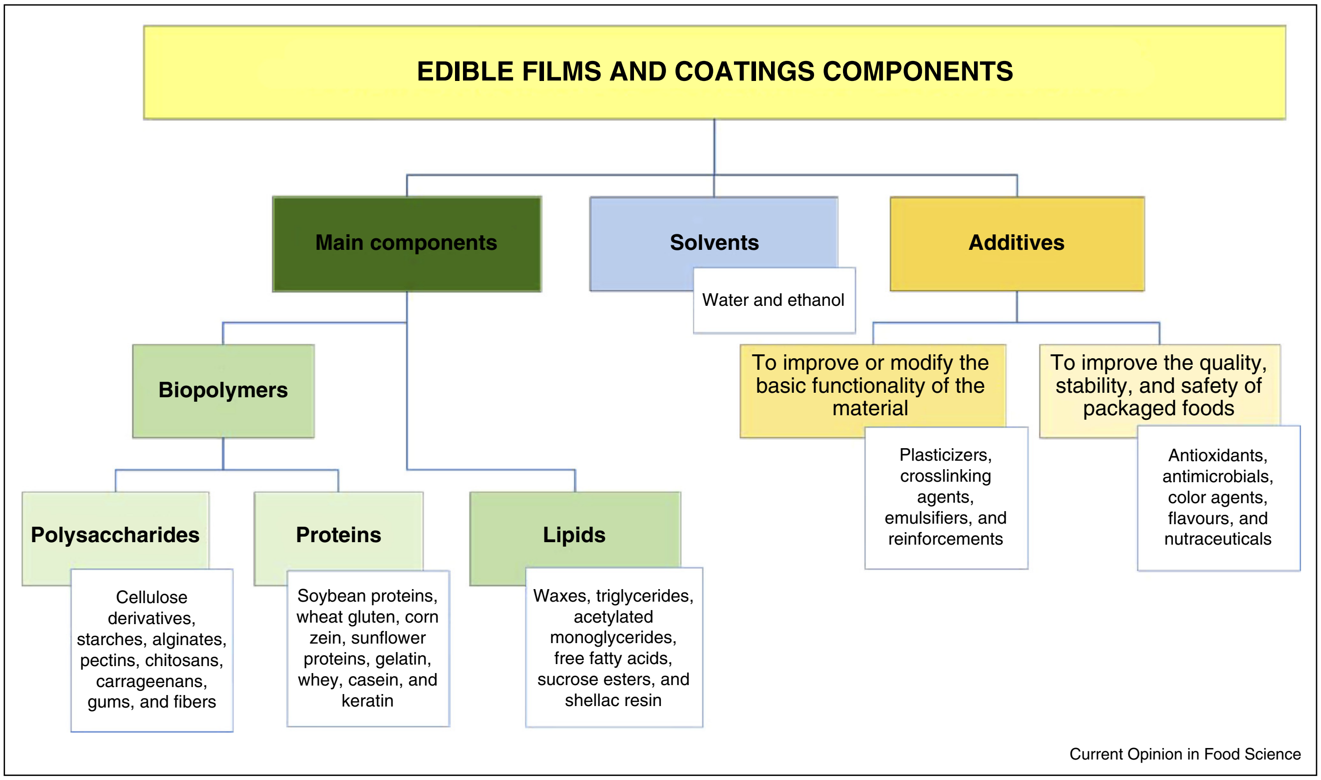
physicochemical, and microbial stability, (iv) non-toxicity and safety, (v) non-polluting nature, (vi) simple technology and (vii) low raw material and processing cost [8,9].

Edible films and coatings formulation and processing

Figure 2 summarizes edible films and coatings composition. Their main components are biopolymers and lipids, or their mixtures. The physical and chemical characteristics of these components are very different, therefore they would influence the functionality of the materials formed. Biopolymers include proteins, from vegetal or animal origin like soybean proteins, wheat gluten, corn zein, sunflower proteins, gelatin, whey, casein and keratin; and polysaccharides, such as cellulose derivatives, starches, alginates, pectins, chitosans, carrageenans, gums and fibers [10,11]. Both protein and polysaccharides films generally exhibit excellent barrier properties against oxygen, lipids, and aromas, moderate mechanical properties but high water vapor permeabilities. Edible lipids include beeswax, candelilla wax, carnauba wax, triglycerides (e.g. milk fat fractions), acetylated monoglycerides, free fatty acids, fatty alcohols, sucrose esters and edible terpene resins such as shellac resin [9,10]. Even though they are not polymers, they form cohesive biomaterials since they can be molded taking into account its characteristics phase transitions temperatures [12]. Lipid and resin materials may provide desirable gloss and an effective barrier for water loss. Composite films can be created by mixing two or more of these components, yielding one homogeneous film layer, or a multi-layer film, in order to achieved the best properties of each component and minimize their disadvantages [13]. Thus the advantage of the good water barrier properties of lipids coatings and the ability to form cohesive films with good gas permeability properties and no greasy texture of polysaccharides or proteins can be combined to form edible emulsion or bilayer coatings [14].

There is currently a special interest in the use of biomaterials extracted from renewable agricultural resources or

Figure 2



Edible films and coatings compositions.

industrial by-products to this end, due to their greater availability, lower cost and as an alternative to provide more added value to these byproducts. Starch and cellulose obtained from different vegetables, chitosan from crustacean, carrageenan and protein extracted from seaweed, whey protein from the dairy industry, gelatin from slaughterhouses and tanneries, soybean and sunflower proteins from oilcakes and keratin from feathers comply with the premises mentioned previously [15•].

Film forming materials, can be hydrophilic or hydrophobic, but in order to maintained edibility, only water or ethanol could be used as solvent during processing. Other components can also be added into the materials matrix to enhance its functionality. This additives include: (i) those that can improve or modify the basic functionality of the material, such as plasticizers (specially polyols-like glycerol, propylene glycol, sorbitol, sucrose, polyethylene glycol-, fatty acids, and monoglycerides) [4], cross-linking agents (such as tranglutaminase or genipin for proteins and citric or tannic acid for polysaccharides) [16,17], and reinforcements (such as fibers and nanoreinforcements derived from polysaccharides like cellulose nanofibers, starch nanocrystals, and chitosan nanowhiskers) for improving mechanical properties [18–21], and emulsifiers for stabilizing composite coatings and improving its adhesion (such as tweens, spans, fatty acid salts and lecithins) [22,23] and (ii) those that intend to improve the quality, stability, and safety of packaged foods, such as antioxidants, antimicrobial compounds, nutraceuticals, flavors, and/or color agents [24,25••,26•].

Despite the film forming process, whether it is a wet or dry casting, an edible film is essentially a dried and extensively interacting polymer network with all film forming components into the resulting three dimensional gel structure. Wet processes are based on the solubilization or dispersion of all ingredients and subsequent drying to form film structure [27]. Dry processes do not use solvents, which is an environmental advantage. They are based on the thermoplastics properties of film forming ingredients, since in this processes heat is applied to temperatures above the melting point of the materials to cause them to flow. Conventional processing techniques of synthetic polymers, such as compression and injection molding and extrusion are being studied to process biopolymer and composite (and nanocomposite) based materials [28–31]. Coatings can be formed directly on the food surface by dipping the product into, brushing or spraying it with the film forming dispersion or by covering the food surface with a film previously formed [2•].

Active films and coatings for food packaging

Traditionally, the package should act purely as a container and a barrier of food systems to isolate it from the exterior, remaining inert toward the packaged food. On the last two decades food packaging technology has been continuously evolving in response to growing challenges from the modern society [32••]. These challenges include safer and healthier food, longer shelf life, convenience, global markets, legislation, authenticity, food waste and environmental concerns [33••]. Active packaging is one of the most dynamic technologies used to preserve food. It interacts positively with product and environment to extend the shelf life of food or to improve its safety or sensory properties, while maintaining its quality [34,35]. These packages base their activity on intrinsic properties of the polymer or on the properties of specific additives that are incorporated into the packaging systems [8,34].

Some biopolymers such as chitosan have antibacterial and antifungal activity *per se*, that can be attributed to changes in cell permeability caused by interactions between the amine groups of chitosan and the electronegative charges on the cell surface [36,37]. The intentional grafting of an active group or the introduction of an active monomer inside the polymer chain, can also lead to polymers with new functionalities [8].

An active agent can be incorporated inside or onto the surface of packaging materials, and also in particular elements associated with the packaging such as sachets, labels or bottle caps [8,38•]; instead of applying them directly on foods, in order to attain the desired goal with lower concentrations, thus limiting unwanted flavors and odors to the food [39]. These active compounds are incorporated to be released into or absorb substances from the packaged food or its surrounding environment, or to perform changes in the composition or the organoleptic characteristics of the food; on condition that these changes should comply with the provisions foreseen in the community or food legislations [2•]. Currently active packaging systems involve: oxygen, carbon dioxide, ethylene, and flavors scavenging/adsorbers, moisture absorption, carbon dioxide, sulfur dioxide, ethanol and flavors generation, and finally antimicrobial and antioxidant systems [5,39–44]. Thus, the development of active packaging materials has expanded attempting to provide answers to specific problems and to look for new market opportunities.

The nature of the active agents may be diverse, including organic acids, enzymes, bacteriocins, fungicides, natural extracts, ions, ethanol, polyphenols, protein hydrolyzates, etc. Nowadays there is a renewed interest in the use of natural additives, instead of those of synthetic origin — sometimes associated with certain health risks [45,46]. Even some agroindustrial waste or by-products, such as those derived from fruit and vegetable processing, or wine, beer, dairy and meat industries, provide practical and economic sources of potent active compounds, such as essential oils, extracts, polyphenols, anthocyanins, pigments, peptides, etc., that could replace synthetic preservatives [47,48].

Moreover, the processing technique of the packaging material should be selected considering the characteristics of the active agents, especially their heat resistance and their action mechanism, to avoid the damage on these compounds during processing and ensure the package activity.

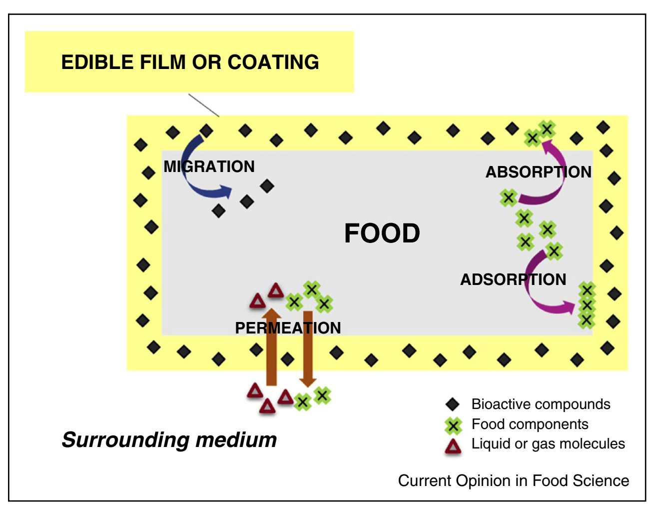
Eventually some additives, able to sense, detect, or record internal or external changes in the product's environment were used to develop 'intelligent packaging'. Their function is to inform the manufacturer, retailer and consumer on the present properties of food or to record aspects of its history. Sometimes intelligent packaging can be used to check the effectiveness and integrity of active packaging systems [34,49].

Mass transfer processes involved in active films and coatings

The interactions that can occur between the food product, the film or coating used as packaging and the surrounding environment are governed by different mass transfer processes: migration, adsorption, absorption and permeation [5]. Figure 3 depicts these processes graphically.

Through migration process, the bioactive compounds included in the polymer matrix can be transferred to the product or headspace and there pursue their protective action (antioxidants, antimicrobials, etc.) [5]. However,

Figure 3



Mass transfer processes involved in active films and coatings.

they could also modify the organoleptic and nutritional characteristics of the product (dyes, peptides, vitamins, etc.). It is worth noting that the migration of these components — initially present in the film or coating — should not present a risk of toxicity or alter the properties of the resulting polymeric materials.

In the adsorption process, components of food or headspace are transferred and incorporated into the surface of the film or coating, while in the absorption process these components are finally incorporated within the polymer matrix [6]. In both cases, the package may act as scavengers of O_2, CO_2, H_2O, and C_2H_4, which for example have an important role in the ripening and senescence of vegetables. But by these processes, some important food components such as vitamins, minerals, and other nutrients can also be sequestered. As a result, the incorporation of these new compounds in the packaging material would probably modify its properties and functionality.

Permeation involves the diffusion of liquid or gas molecules through the polymer matrix [5]. In this process, different types of components, such as H_2O, O_2, CO_2, C_2H_4, flavors, etc., can be transferred from the environment to the food, or vice versa; leading to moisture gain or loss, changes in texture and color, development or inactivation of microorganisms, loss of taste, odor occurrence, lipid autoxidation and enzymatic browning reactions, and the establishment of modified atmospheres inside the packaging, among others effects.

Regardless of the mechanism governing the transport of the active agents, the transfer rate of these compounds depends on the chemical nature and the crosslinking grade of the polymeric material, the concentration of the active compound, its affinity with the polymer matrix and food; and finally on the environmental conditions, such as relative humidity, temperature and contact time. Therefore, it is important to know how these variables affect the effectiveness of the active packaging.

Currently, there is a pronounced interest in the potential use of nanocomposite systems as carriers of active compounds for food packaging, as it is expected that the presence of nanoreinforcements — well-dispersed in the polymer matrix — could modulate the passage of molecules through the film [50••,51••].

New patents on films and coatings containing bioactives (mini-review)

Indeed, the production of edible films and coatings containing bioactives and their applications on food products have been subject of a great number of scientific publications during the last decade. In particular numerous studies performed all round the world, using different biopolymers and active compounds for different purposes, were described in some patents in the last two years. For example, the preparation of chitosan films activated with green tea polyphenols to prevent food product oxidation was described in CN 103087356A patent [52]. The development of gelatin and glucomannans films with garlic juice — as antimicrobial and flavoring agent — or with red bean powder — as colorant and flavoring agent — to be used in candies and brewing food were reported in CN 103589168A [53] and CN 103589173A [54] patents respectively. Pectin and chitosan films incorporated with tea polyphenols, natamycin and citric acid were proposed to protect fruits and vegetables against microbial growth and enzymatic browning (CN 103719267A patent) [55]. Gelatin and CMC films with potassium sorbate were probed to be effective to extend the shelf-life of bacon in CN 102487988B patent [56]. Edible wrap film based on sodium alginate and chitosan and activated with lactic acid, ascorbic acid and nisin could prolong the shelf-life of packed fresh pork (CN 102977414B patent) [57]. The formation of a film containing bacteria, yeast and its nutrients to be used as a bioactive modifier of packaged food atmosphere was described in EP 2783584A1 patent [58]. The preparation of edible films based on pea or tapioca hydroxypropylated starches containing cola flavoring was described in US 20140272069A1 patent [59]. Arabic gum and starch films including flavoring and sweetening agents were proposed to mask the taste of foods that contain meats or vegetables such as broccoli, cabbage, spinach, etc. (US 20140234509A1) [60]. A confectionery packet or sachet formed with an edible film based on HMPC and CMC and enclosing a mixture of sugar alcohols, flavors and/or colors, anti-bacterial, and nutraceutical or pharmaceutical compounds was reported in US 20140302203A1 patents [61].

Future directions

Edible films and coatings are promising systems to be used as active ingredient carriers. In order to asses a successful application of these materials as food packaging, the mechanisms of deterioration of each food and the mode of action of each package should be understood and related [62,63••].

Studies in real, pragmatic, everyday systems are essential and should be performed in order to prove the true effectiveness of the developed package. Because sometimes the *in vitro* properties of a materials could not be evidenced in food systems, often due to difficulties in the release of the active compounds or their inactivation during processing.

There are great research efforts focused in attempting to optimize the composition of film and coatings materials in order to be well-processed by new and economically feasible processing systems, such as those extensively used by the plastic industry. The use of bioactive compounds and biopolymers derived from agroindustry byproducts or waste would also favor the profitable development of these environmentally friendly materials.

It is worth noting that no single film or coating would be appropriate, or even adequate, for all applications involving foods, agriculture, or nutraceuticals. On the contrary, each material should be compatible with a given concrete application and through that multifaceted specificity demonstrates the inherent diversity of these systems [64••].

Finally, the massive application of these materials would depend on the efficiency of the packaging, its economic and environmental impact, the existing legal restrictions, and the acceptability of these systems by consumers.

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This review gathers updated research reported over the last ten years related to antimicrobial edible films and coatings applied to meat and meat products. The perspective of this technology includes tailoring of coating procedures to meet industry requirements and shelf life increase of meat and meat products to ensure quality and safety without changes in sensory characteristics.