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Entry

Active Edible Packaging

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Definition: Active edible packaging is a food packaging made of comestible bioproducts and active compounds that interacts with the food. The bioproducts, usually biopolymers, must be recognized as safe and with characteristics to be consumed by humans—comestible—and not toxic and capable of carrying an active compound, like anti-browning agents, colorants, flavors, nutrients, antimicrobial and/or antioxidant compounds, in order to extend the product shelf-life, reduce contamination and maintain or even enhance the nutritional value.

Keywords: edible packaging; active packaging; biopolymers; active compounds

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1. History

Food packaging intends to pack and protect food from environmental, chemical and physical damage during transport, distribution, storage and retailing up to its final consumer. In recent years, concerns have arisen regarding conventional packaging, specifically plastic and its derivates, due to the impact it has on the environment when disposed of and due to its non-renewable character. To avoid this problem, researchers are developing new packaging systems that are produced from renewable raw materials and that are biodegradable, such as active edible packages; therefore, they present low environmental impact [1–4].

Edible packaging is used with foods to prolong shelf-life and can be consumed with food. Edible packaging can be used in the form of thin film and applied to the food surface or as a coating, which is a thin layer formed directly on the food surface. Both provide a barrier to moisture, oxygen and others from and to the food, and can be a carrier of active compounds [2,5–8].

The use of edible packaging goes back to the 12th century where, in China, wax was used in citrus fruits to prevent moisture loss and to promote a shiny surface. This type of packaging can be produced from bioproducts, e.g., polysaccharides, proteins, lipids or biocomposites, that are biodegradable, biocompatible and recyclable and of renewable origin.

Active packaging incorporates components, and it can release or absorb substances into or from the packaged food, or the environment surrounding the food, intending to extend the shelf-life or to maintain or improve the condition of packaged food [9]. The active compounds can be anti-browning agents, colorants, flavors, nutrients, spices, antimicrobial or antioxidant compounds, and they can have a natural or synthetic origin. Butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT), propyl gallate (PG) and tertbutyl hydroquinone (TBHQ) are common synthetic compounds used in active food packaging. Nonetheless, more recently, the interest has been towards compounds of natural origin because some of the synthetic antioxidants have been associated with adverse health effects. Compounds of natural origin commonly used include essential oils, extracts of plants and spices, and probiotics. In addition, studies are being carried out on the use of extracts obtained from food by-products, such as fruit and vegetable by-products, as a source of antioxidant compounds, or other bioactive compounds, in the preparation of active edible packaging [2,10–13], thus contributing to the circular and bio-economy.

In the European Union, active edible packaging is regulated by several regulations. As a material or article intended to come into contact with food, it is regulated by Regulations No. 1935/2004 and its amendments, and Regulation No. 2023/2006, regarding its good manufacturing practice. According the European Union, food additives are "any substance not normally consumed as a food in itself and not normally used as a characteristic ingredient of food, whether or not it has nutritive value, the intentional addition of which to food for a technological purpose in the manufacture, processing, preparation, treatment, packaging, transport or storage of such food results, or may be reasonably expected to result, in it or its by-products becoming directly or indirectly a component of such foods". Therefore, the active compounds added to food packaging are considered food additives, specifically indirect food additives as they are not added directly to the food. In this sense, active edible packaging is also regulated by Regulation (EC) No 1333/2008 of the European Parliament and of the Council of 16 December 2008 on food additives and Regulation (EC) No 1334/2008 of the European Parliament and of the Council of 16 December 2008 (on flavorings and certain food ingredients with flavoring properties for use in and on foods). Finally, as an active packaging it is regulated by Regulation No. 450/2009 (active and intelligent materials) [9,14–17].

Additionally, in the great majority of the cases, in order to obtain a package with the best mechanical properties, it is necessary to add a plasticizer, such as sorbitol, glycerol, polyethylene glycol, monoglycerides or glucose. Still, taking into account that the packaging is edible, all the materials from preparation to obtaining the packaging must be able to come into contact with food and fit for consumption [6,18,19].

Finally, since the ideal packaging depends on the food, studies are being carried out to determine the best combination of polymers and active compounds for each group of food, so that the best edible active packaging can be obtained. Still, it is necessary to optimize the packaging preparation process so that it is fast, efficient and low cost.

2. Current Knowledge and Applications

2.1. Polymers

The materials used to produce an edible active packaging must be fit for human consumption, non-toxic and capable of transporting active compounds. The polymers normally used are polysaccharides, proteins, lipids or composites.

Polysaccharides, complex carbohydrates, are wildly used in the preparation of edible packaging. Polysaccharides occur naturally in nature and can have different origins: animal (e.g., chitin and chitosan), plant (e.g., starch and pectin), marine (e.g., alginate) and microbial (e.g., xanthan gum and pullulan). Polysaccharides can originate films and coatings that are colorless, tasteless, have an oil-free appearance and are good oxygen and carbon dioxide barriers, but are poor barrier to water vapor [6,20,21].

Proteins are macromolecules made up of more than 100 amino acid residues linked by peptide bonds. Proteins can be divided into fibrous and globular proteins. Fibrous protein is water-insoluble and has an animal origin, whereas globular proteins are soluble in water, acids and basic solutions and have a plant origin. Different proteins, such as whey protein, casein, gelatin, collagen, soy protein, wheat gluten and corn zein, are used in the preparation of edible food packaging. They can originate films and coatings that are hydrophilic and with good oxygen barriers but poor mechanical strength [6,20,22].

Lipids originate from animals, insects or plants, and they are naturally hydrophobic polymers. The most commonly used lipids in edible packaging are natural waxes, acetylated monoglycerides and resins. Lipids can provide films and coatings with good barrier properties to moisture but poor barrier properties against oxygen and carbon dioxide. In addition, lipids originate brittle and thicker films and coatings with poor mechanical properties [6,20,22,23]. To overcome the disadvantages of each polymer, they can be combined to obtain films or coatings with better barrier and mechanical properties. The combination of different polymers results in composite films and coatings. Several combinations can be made, such as polysaccharides and proteins, polysaccharides and lipids, protein and lipids, a combination of two different polysaccharides or the combination of natural and synthetic polymers, among other options. Lipids are frequently added to polysaccharide and protein films and coatings to improve their water barrier properties. Then again, to overcome the poor mechanical properties of lipid polymers, polysaccharides and proteins can also be added [6,20,22,24,25]. Some examples of composite films and coatings are corn starch and carboxymethyl cellulose [26], whey protein isolate and zein [27], starch, gellan and thyme essential oil [28], chitosan and alginate [29] and others. The weaker mechanical properties of these biopolymers or biocomposites can also be overpassed with the introduction of nanofillers, e.g., montmorillonite or nanocellulose, that reinforce the polymeric structure [30–32].

2.2. Antioxidants Compounds

According to Regulation No. 1333/2008, antioxidants are "substances which prolong the shelf-life of foods by protecting them against deterioration caused by oxidation, such as fat rancidity and color changes" [14]. This activity can be present in the chosen polymer or in the added compounds to the food and/or packaging. The main active compounds reported to be used in food packaging are phenolic compounds (flavonoids, tannins, and phenolic acids), which present powerful antioxidant and antimicrobial activities. The antioxidant activities of these compounds are related to their redox properties. Different mechanisms have been proposed to explain such activities, like transition-metal-chelating activity, free-radical scavenging activity and/or singlet-oxygen-quenching capacity. In addition, their ability in retarding food lipid oxidation has been shown; therefore, they can extend food shelf-life. In addition, phenolic compounds have various health benefits, as they are anti-inflammatory, antihypertensive, antioxidative and antiaging. These compounds naturally occur in plants as well as fruits and vegetables [33–38]. Some herbs and spices, like rosemary (*Rosmarinus officinalis* L., syn Salvia rosmarinus Spenn.), marjoram (*Origanum majorana* L.), basil (*Ocimum basilicum* L.), oregano (*Origanum vulgare* L.), sage (*Salvia officinalis* L.), savory (*Satureja hortensis* L.) and thyme (*Thymus vulgaris* L.), have been widely studied for this purpose [33–36,39]. Teas, from the Theaceae family, are also rich in phenolic compounds and widely used for this purpose. The main phenolic compounds found in tea leaves are catechins, flavonols, flavones, anthocyanins, hydrolysable tannins and phenolic acids. For instance, the antioxidant activity of green tea (*Camellia sinensis* L.) is due to the high amounts of catechins, whereas epicatechin, epicatechin gallate and catechin have the highest antioxidant activity. The catechins in green tea are also responsible for its antimicrobial activity [33,34,39–42].

2.3. Antimicrobial Compounds

According to Regulation No. 1333/2008, antimicrobials, or preservatives, are "substances which prolong the shelf-life of foods by protecting them against deterioration caused by micro-organisms and/or which protect against growth of pathogenic microorganisms" [14]. Several polymers have antimicrobial activity on their own, without the need to add an active compound. This is the case of chitosan and whey protein. Chitosan can inhibit bacteria (*Listeria monocytogenes*, *Bacillus cereus*, *Staphylococcus aureus*, *Escherichia coli*, *Salmonella typhimurium*), yeast and mold (*Candida lambica*, *Botrytis cinerea*, *Rhizoctonia solani*, *Phomopsis asparagi*, *Fusarium oxysporum*) growth. Chitosan antimicrobial activity is associated with the reaction of its positively charged amino groups with microbial cell membranes that are negatively charged. As a consequence, cell membranes become more permeable, and intracellular material can leak. In addition, chitosan can chelate the metals in the vicinity of bacteria, stopping the movement of essential nutrients [43]. Nonetheless, the antimicrobial activities depend on concentration, degree of acetylation, pH, the test organism, cationic nature, and the period of exposure [43–46]. Whey protein is a by-product of the dairy industry and holds high biological activities. Similar to chitosan, the exact antimicrobial mechanism is still unknown, but it depends on the ionic environment, pH, lipid presence, and pre-heat and heat treatments [47–50].

Essential oils are secondary metabolites from plant materials such as seeds, flowers, leaves, herbs, buds, stem, bark and fruit by-products. They are known for their antimicrobial activity, besides other properties (antioxidant, analgesic, anti-diabetic, anti-tumor, anti-inflammatory and insecticidal). There are a wide range of essential oils originated from different plants, like rosemary (*Rosmarinus officinalis* L., *syn Salvia rosmarinus* Spenn.), basil (*Ocimum basilicum* L.), oregano (*Origanum vulgare* L.), cinnamon (*Cinnamomum zeylanicum Blume*), lemon (*Citrus limon* L.) and garlic (*Allium sativum*). Essential oils have presented antimicrobial activities against a wide range of microorganisms (*Staphylococcus aureus*, *Escherichia coli*, *Streptococcus* spp., *Bacillus cereus*, *Enterococcus faecalis, Klebsiella pneumoniae*). Different factors can affect the antimicrobial activities of essential oils. The main factors are species, chemical composition, method of analysis, method of essential oil extraction and tested microorganisms [34,51–54]. The incorporation in the biopolymers of nanometal oxides is also an interesting option as it contributes to improve the antimicrobial and UV-blocking properties of the packaging [55,56].

Other antimicrobial compounds that can be used are certain bioactive peptides and protein hydrolysates. These compounds present different functional properties, including antimicrobial properties. Antimicrobial peptides are characterized by having low molecular weight, a highly cationic character and an amphipathic structure. Their mechanism of action is based on the peptides' electrostatic interaction with the negatively charged cell membrane of microorganisms, causing its disruption [57–59]. Their antimicrobial action can be also explained though its iron chelating activity [58]. Antimicrobial peptides are very effective against several microorganisms, so they have great potential to be incorporated into active packaging. Antimicrobial peptides are generally obtained from natural sources, such as bacteria, fungi, amphibians and mammals, or by enzymatic hydrolysis of food proteins, but they can also be chemically synthesized. Antimicrobial peptides produced by bacteria originate bacteriocins like nisin [57,58]. Nisin is a bacteriocin synthesized by *Lactococcus lactis* subsp. Lactis and has been used as a food additive for decades. It is identified as Generally Recognized as Safe (GRAS) in the United States, and in the European Union it is permitted as a food additive (E 234), mostly in dairy products and canned food. Nisin presents antimicrobial activity mostly against Gram-positive bacteria, yeasts and molds [57,60,61]. Natamycin, or piramicin, is fungicide produced by aerobic fermentation of *Streptomyces natalensis*. It has also been identified as a GRAS in the United States, and in the European Union it is allowed as a food additive (E 235). Natamycin is commonly used as a surface treatment for cheese and dry sausage, against the proliferation of yeast and molds [60,62,63].

2.4. Other Compounds

In addition to the active compounds already mentioned, there are other compounds that can be added to active edible packaging in order to improve barrier and mechanical properties, protect against UV radiation, and others. Among these compounds are nanomaterials such as nanocellulose, Montmorillonite (MMT), nanometal oxides (e.g., silver nanoparticle) and inorganic nanoparticles (e.g., silica). Due to their nanoscale size, these compounds are able to interact with the polymer at the atomic, molecular or macromolecular level, consequently modifying the properties of the film/coating. The incorporation of these nanomaterials creates a more tortuous path that slows the permeation of gases through the polymer and improving its barrier properties. In addition, these nanomaterials can act as a delivery system of active compounds or they can interact with the permeant species (water vapor, O_2), or other species released from the food or from the outer environment, adding activity to the films. Nonetheless, more studies are needed to evaluate the safety of these compounds, whether they migrate to the food and the interaction they may have with the food [25,64–68].

Cellulose is the most common polysaccharide present in nature and can be transformed into nanocellulose by breaking down the cellulose fibers. Regarding the origin and shape, nanocellulose can be divided into cellulose nanocrystals (CNC), cellulose nanofibers (CNF) and bacterial nanocellulose (BNC). CNC have a crystalline structure leading to a more rigid mechanical properties, whereas CNF have both crystalline and amorphous structure, and consequently present more flexible properties [64,67,69]. The impact that nanocellulose has on the film/coating depends on its concentration and interaction with the polymer matrix. Nevertheless, both CNC and CNF are able to increase the thickness of the packaging as well as the opacity of the film, and effectively protect the food against UV radiation. Depending on its concentration, the incorporation of CNC can reduce the gas and water vapor permeability. On the other hand, CNF provides good gas barrier properties, but it is unstable in relation to water vapor permeability. Both improve the mechanical properties of films and may enhance the antioxidant and antimicrobial properties of the active compound of the packaging [65,67]. Montmorillonite is one of the most common used nano clays in food packaging. MMT is composed of one silica tetrahedral sheet sandwiched with two edge-shared octahedral sheets of aluminum hydroxide. One of the main advantages of MMT is that it is a stable compound, which can be easily found in nature, and with low cost. Similar to nanocellulose, MMT is frequently added to the film matrix to improve its properties, at low concentrations. MMT can be modified through the organophilization process, where there is an exchange of cations, so that its application to polymers is more compatible. The addition of MMT to the film matrix affects the optical properties, creating a more opaque film. MMT can enhance the mechanical properties as well as the thermal properties of the packaging [32,54,70–72].

2.5. Preparation of Edible Active Packaging

There are several processes for preparing edible active packaging, which can be divided into wet and dry processes. For the wet processes, there are casting, dipping, spraying and spreading techniques. For the dry processes, there are thermopressing/thermoforming and extrusion. The main difference between dry and wet processes is that the wet processes require a drying step, unlike the dry processes. Films are normally prepared through casting, extrusion and thermopressing/thermoforming. Whereas coatings are prepared through dipping, spraying and spreading [5,12,73–76]. Casting is generally the method of choice in the preparation of edible active packaging. The casting technique is well established on a laboratory scale, but it is not viable on an industrial level. This technique requires the dissolution of the selected biopolymers in a specific film solution, along with the chosen active compounds and plasticizer. Water and/or ethanol are normally the selected solvents for edible packaging. Afterwards, the pH is adjusted, and the solution is heated above the melting temperature until complete homogenization. Finally, the solution is spread on a flat surface to dry. Different drying processes can be applied, such as air drying, hot surface drying, infrared and microwave techniques. Finally, the obtained film after the drying step is used to wrap the food sample. The major setback of the casting technique is its inability to be applied on an industrial scale, making production difficult in large sizes and with long drying process [5,12,73–75,77]. The dipping, spraying and spreading techniques follow the same steps as the casting technique up until cooling the solution. For the dipping technique, the food sample is dipped in the solution, after it is cooled down, and then the liquid excess is drained and is allowed to air dry. The dipping technique is a low-cost procedure and easy to apply, but there may be microorganism growth in the dipping tank, in addition to generating a lot of waste [73,77–79]. The spraying technique is also a very common technique that allows a uniform coating around the food sample as it is easy to control the process. In this case, the solution is sprayed with nozzles on the food sample, which is on a rotating platform at a controlled speed [73,77,78,80]. Finally, the spreading, or brushing, technique consists of spreading the solution on the surface of the food sample with the help of a brush or a spatula. It is normally applied to small foods, like fruits and vegetables, and originates homogeneous coatings. Finally, these wet processes can be applied in a layer-by-layer mode, where layers with different polymeric matrices are applied sequentially. The layer-by-layer technique aims to produce food packaging with better polymeric properties, but it requires more production time as it is necessary to dry one layer before adding another [6,73,78,81].

Regarding the dry processes, the extrusion technique is the industry's preferred technique as it originates food packaging with enhanced mechanical and optical properties in a very short period, with low energy consumption. Although extrusion is a low-cost technique, it requires a high initial investment with the equipment and the maintenance thereof. This technique involves minimum solvent contents, besides the plasticizers, the possibility of using viscous polymers and easy control of working temperature and pressure. Nonetheless, some polymers may not be used with this technique due to its moisture limitation. The extrusion technique can also be applied with multiple layers (co-extrusion) to improve the properties of the films [73,74,77]. Another dry process is the thermopressing/thermoforming technique; it is a method normally used for the production of utensils, namely made of plastic. Similar to extrusion, it involves high temperatures and pressure responsible for the formation of the film. It is normally used on an industrial scale and has been recently applied for the production of bioactive films [73].

Lately, new processes have surfaced in the making of edible active packaging. Among them, there is electrospinning and controlled-release active packaging. Electrospinning is a simple technique used to produce nanoscale fibers from various polymer materials using a high-voltage electric field. The electrospinning technique has several advantages associated with it. For instance, it is a low-cost technique that produces films with high porosity, ultrafine structures, tailored morphology and high surface-to-volume ratio. The process temperature is controlled, allowing its application to temperature-sensitive compounds, thus preserving its characteristics. Still, it is possible to effectively encapsulate the bioactive compounds. However, electrospinning has low productivity, which limits large-scale production. The characteristics of the polymers may also limit this technique due to their poor mechanical properties and good barrier properties [74,82,83]. Controlled-release active packaging is a package that allows the release of the active compounds from the package to the food in a controlled way in order to extend the shelf-life of the product. The main aim of controlled-release active packaging is to prolong the release of the active compound throughout transport and storage. One of the main conditions to be controlled is the speed of release of the active compound, as a slow release can result in the deterioration of the food, while a quick release can result in the loss of the active compounds due to its degradation. Therefore, it is important to define when the release of the active compound will start, how it will be made and the amount that will be released. Nonetheless, it is also necessary to take into account the choice of the polymer, the active compound, their concentration, the processing method and the food product intended to be packaged in order the controlled-release active packaging is effective in maintaining the quality of the food and extending its shelf-life [74,84].

2.6. Application of Edible Active Packaging

Several studies are being made to evaluate the effectiveness of active edible packaging in different food groups. For instance, Vital et al. [85] studied the effects of two alginate-based edible coatings containing, respectively, rosemary and oregano essential oils, on the quality and shelf-life of beef under refrigerated storage. The authors concluded that both coatings were effective against lipid oxidation of the beef, as well as water and color loss, compared to the control, but the oregano coating was more powerful, compared with the rosemary coating. Another study carried out by Botelho et al. [86] evaluated the effect of cinnamon essential oil incorporated in cassava starch on guavas. Botelho and colleagues concluded that the coating prevented weight loss and maintained the green color of the fruit up to the 8th day of storage when compared with control treatment. Castro et al. [33] studied the effectiveness of an edible whey protein concentrate film, incorporated with green tea extract, on retarding the lipid oxidation of fresh salmon. The green tea whey protein film was effective as it decreased the lipid oxidation of fresh salmon, compared with the control, up to 14 days of storage. In the study of Souza et al. [87], results showed that the incorporation of ginger essential oil in chitosan enhanced the film activity, by reducing lipid oxidation and microbiological growth of the wrapped poultry meat. Dhumal et al. [88] successfully incorporated carvacrol and citral essential oils in a sago starch and guar gum edible film. The active film with both carvacrol and citral essential oils presented high antimicrobial activity against *Bacillus cereus* and *Escherichia coli*. An edible active composite coating was tested by Di Pierro et al. [89] on ricotta cheese. The coating was prepared with whey protein and chitosan, and the quality of the coated and the control cheese, stored at 4 °C under modified atmosphere conditions for 30 days, was evaluated. The authors observed no significant difference in the pH of both coated and control cheese. The titratable acidity of the control reached 0.34 milliequivalent/100 g of analyzed sample after 21 days, and remained constant, while the coated cheese reached 0.33 milliequivalent/100 g of analyzed sample only after 30 days. In addition, the coating was effective against mesophilic and psychrotrophic microorganisms, and lactic acid bacteria [89]. Finally, Souza and colleagues [32] studied the physical properties of a film based on chitosan reinforced with different MMT (natural and commercial) and further incorporated with rosemary essential oil. The film incorporated with the natural MMT presented the best reinforcement of the mechanical properties. Therefore, it was further incorporated with rosemary essential oil. In this case, there was also an improvement in the mechanical properties and a reinforcement of the UV radiation block [32].

3. Conclusions and Prospects

The development of active edible packaging is growing. Research studies are capitalizing efforts on the use of natural polymers which, being biodegradable, do not generate residues and have low environmental impact. Moreover, the renewable character of the feedstocks and the possibility to make use of agroindustry wastes (e.g., chitin from the exoskeletons of crustaceans or pectin from wastes of citrus fruits) also contributes to the circular economy and to alleviate the environmental burden related with plastics of fossil origin. In addition, active compounds can be added to these packages, and the most recent studies have increasingly opted for those of natural origin and that can be consumed by humans. Thus, active edible packaging is promising for the near future, having associated several advantages. This type of packaging helps to extend the shelf-life and to prolong or even improve the quality of the food and meet the increased consumer demand for more natural and environmentally friendly products. However, before they can be adopted by the industry on a larger scale, and thus reduce the use of current conventional packaging, some limitations still need to be overcome. Further studies on natural active compounds (including those that can be retrieved from agri-food wastes) and their compatibility with biopolymers are needed, as well as studies on solutions to improve the barrier and mechanical properties of the active packaging. Studies on innovative mechanisms that can encapsulate natural active compounds and that allow a more controlled migration to food also represent an opportunity to improve these packaging systems. In addition, efforts must be made to develop packaging production techniques that are more efficient and cost-effective, and that make it easier to expand these types of edible packaging systems for the industry.

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