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# Films and Edible Coatings in the Development of Biodegradable Packaging: Sustainable and Eco-friendly Alternatives

## *Filmes e Revestimentos Comestíveis no Desenvolvimento de Embalagens Biodegradáveis: Alternativas Sustentáveis e Ambientalmente Corretas*

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Packaging is fundamental to food industry because its main purpose is to protect food from external contamination. However, since most traditional packaging material are not made from renewable and biodegradable sources, they cause major environmental problems. Packaging made from synthetic polymers is inexpensive due to several facts, such as easily available raw material and their low cost, but its useful life is short, a fact that leads to inadequate accumulation and discard which impact on the environment. Development of biodegradable packaging made from renewable sources is an excellent alternative, mainly regarding discard. This study aimed at compiling the main biotechnological studies of different types of films and edible coatings that have already been published. Innovative material may replace packaging made from synthetic plastic. Therefore, alternative means must be made available so that industries can invest in them. Findings in the literature led to the discussion of the development of some types of films made from biopolymers, additives and natural bioactive compounds. Researchers bet on films and edible coatings not only because their physico-chemical characteristics are effective to protect food but also because their development is easy. Finally, another strength is that the raw material needed to develop them, such as starch, is highly available.

**Keywords:** Biopolymers; casting; sustainable development; starch; food chemistry.

## 1. Introduction

Packaging is likely to be the most important method of food preservation. It protects, preserves and provides information on the product, besides enabling it to be widely distributed and sold.[1-2] Its characteristics depend on the food product that it protects. Different raw material, such as paper, cardboard, metal, glass and plastic, have been used to manufacture packaging.[3] However, old methods of traditional preservation are responsible for large amounts of urban solid waste (USW). Data issued by the Ministry of Environment and Natural Resources showed that tons of packaging – mostly made from plastic, such as polystyrene (PS), polyethylene (PE), polypropylene (PP) and polyethylene terephthalate (PET) – have been discarded in the environment.[4]

The world's attention should be called to this problem since environmentalists have stated that, despite campaigns created by several worldwide specialized institutions, such as Departments of Environment, bad management of plastic waste has been the rule, i. e., only 5% has been recycled.[5]

Food industries acknowledge the need for alternative and sustainable methods, and some are even interested in something innovative. A lot of them have reported that most packaging consists of a mixture of material that has distinct characteristics. As a result, material recovery, selection, cleaning and reprocessing make recycling a complicated and costly task.[2] Therefore, renewable raw material have been deeply explored in order to mitigate problems caused by environmental pollution. Packaging based on synthetic polymers has been replaced with alternative biodegradable packaging (based on biopolymers).[6]

Biodegradable packaging not only protects products but also has relatively easier production, recycling and degradation.[1] Most biodegradable packaging usually uses natural and eco-friendly polymeric material. They basically aim at preserving quality and useful life of minimally processed products, such as fruit and vegetables. [7-8]

Therefore, this paper aims at compiling literature findings on the general and update view of the application of films and edible coatings to different types of food. Special attention was given to the following compounds: biopolymers, additives, incorporation of essential oils and bioactive plant extracts and development techniques. Different studies carried out by researchers in this promising area were shown. In addition, the main advantages of alternative biodegradable and eco-friendly packaging were described.

## 2. Material and Methods

The methodology of this study consisted in a brief compilation of some previously published papers whose focus was biodegradable packaging. The following descriptors were used: films, edible coatings, main raw material, application, importance to durability and maintenance of the standard condition of products (in transport or useful shelf life) and natural products (essential oils and plant extracts) incorporated into the development of this type of innovative material. Data were collected in several databases, such as Google Scholar, Web of Science, ScienceDirect, SciELO - Scientific Electronic Library Online and Portal de Periódicos da CAPES. Selected papers which introduced the topic relevantly were cited in the References.

## 3. Results and Discussion

### 3.1. Main characteristics of films and edible coatings

A film or edible coating is any material whose thickness is below 0.3 mm[9], formed by a combination of biopolymers and different additives dispersed in an aqueous medium.[10-11] Some authors use both terms edible films and coating interchangeably even though others believe that they differ because of techniques used for incorporating different types of food products that are expected to be inserted into the composition.[12] Edible coating is directly integrated into food whereas edible film is previously manufactured and then adhered to the product.[12-13] Anyway, in both cases, they are considered rigid matrices in terms of thickness and have similar characteristics.[6-14]

The main characteristics that films and edible coatings exhibit are: (i) protection against ultraviolet light[15]; (ii) solute transport (e. g., salts, additives and pigments), water vapor, organic vapor (e. g., aromas and solvents) and gases (e. g., oxygen, carbon dioxide, nitrogen and ethylene) between food and the atmosphere[15-16]; (iii) barrier against mechanical damage (e. g., smashes and cuts)[12]; and (iv) increase in the useful life of products.[16] Another possibility is to incorporate different types of products into films and coatings to make them even more functional. Some of them are: (v) bioactive compounds (e. g., antioxidants)[17]; (vi) compounds that exert antimicrobial effect on bacterial reproduction and fungal contamination (e. g., the use of silver nanoparticles[18-19]; (vii) healthy microorganisms (e. g., probiotics) that bestow benefits to consumers[20]; and (viii) essential oils and bioactive plant extracts.[21-22]

Biopolymeric material used for manufacturing biopackaging have been differently incorporated due to their glycosidic (i. e., polysaccharides), protein or lipidic nature. Two main types of formulation are emulsion (based on lipids) and colloidal systems (based on polysaccharides or proteins).[23]

Emulsions are systems that consist of immiscible liquid or semi-liquid compounds, such as an oil and an aqueous phase, which may be fused by means of an emulsifying agent. This agent usually has a hydrophilic region and a hydrophobic one which show affinity for polar and nonpolar compounds.[18] In film and edible coating formulation, preferred systems are the ones that are thermodynamically stable and can mainly dissolve lipophilic antimicrobial components (e. g., plant essential oils) and bioactive components (e. g., fatty acids, carotenoids, antioxidants, phytosterols or quinones and plant extracts).[18]

The main basis of colloidal systems is polymeric since they are composed of polysaccharides or proteins dissolved in an aqueous phase[23]. They form a dense matrix that can protect active components (e. g., antioxidant and antimicrobial agents)[24] and enable their controlled release into the matrix.[25] Due to the hydrophilic nature of polysaccharides and proteins, colloidal systems are mainly used for developing films and edible coatings since they can transport and protect several molecules that act as additives (essential oils and plant extracts)[26] and probiotics (lactic acid bacteria).[20]

### 3.2. Different types of films and edible coatings

This subitem aims at showing the diversity of films and edible coatings that have already been formulated and evaluated to be applied to some type of food.

The first film under investigation is cassava starch with nanocellulose as its support and with yerba mate (*Ilex paraguariensis* Saint Hílare) extract as its antioxidant additive.[27] Conceptually, cellulose nanocrystals are crystalline domains of cellulosic sources that result from acid hydrolysis and exhibit characteristics of high rigidity and crystallinity, besides large nanometric size.[27] When these nanoparticles are isolated in a solution, they have been incorporated and evaluated as reinforcing material of polymeric matrices, due to their potential to improve mechanical, barrier, thermal, optical and dielectric properties; when incorporated into matrices from renewable sources, they result in totally biodegradable nanobiocomposites.[27] An antioxidant additive that may be added to this type of polymeric matrix is yerba mate extract, whose chemical composition includes phenolic compounds and flavonoids, known as quercetin and rutin, that bestow its antioxidant potential.[27]

Applicability of the incorporation of the three ingredients – cassava starch plasticized with the addition of glycerol and yerba mate extract – has been intensely discussed. Since production of this antioxidant film was found to be viable in laboratories, it may be applied as packaging to avoid oxidation of lipid products.[27]

There are also films whose preparation and characterization are based on cassava starch incorporating acerola pulp.[28] The authors called them biofilms[28], but we do not agree with this term, since a biofilm is understood as a bacterial agglomerate.[29] Acerola pulp was incorporated into the polymeric matrix composed of cassava starch by casting. The resulting film exhibited good thickness, solubility in water and effective fruit preservation, thus, delaying ripening and extending shelf time of pears.[28]

Fernandes *et al.* (2015)[30] made biodegradable films based on protein concentrate of irradiated milk serum and stated that this type of film has high potential to be used as packaging because of its excellent mechanical and optical properties. Protein of milk serum is a good alternative to produce biodegradable films, which become translucent and yellowish. Other strengths of this type of film are that they represent a good barrier against moisture loss that affects apples and have good solubility in water.[30]

Bean starch is another alternative that may be capable of composing a polymeric matrix. In the literature, a study showed the development and characterization of films based on starch made from black-eyed pea (*Vigna unguiculata* (L.) Wap).[31] Its authors introduced some important information, i. e., that films made from starch alone are quite inflexible and brittle, have high hygroscopy and low adequacy to industrial processing. These characteristics may be improved with the incorporation of additives into the polymeric matrix. Regarding rigidity, improvement may take place after the addition of plasticizers which also favor mechanical properties of films. Plasticizers may also increase polymer flexibility and, consequently, its adequacy to mechanical-industrial processing.[31]

Several sources, such as rice, corn, cassava and potato, have been used for supplying the demand for starch. Bean is a source that has been little explored in this field. Black-eyed pea exhibits 19.92% and 41.36% of starch when it is ripe and green, respectively. In both phases, amylopectin (13.33% and 27.01%) outweighed amylose (5.51% and 15.63%).[31] Films produced with the use of black-eyed pea starch exhibit excellent characteristics, such as continuity, uniformity, manageability and transparence, with neither bubbles nor cracks on their surfaces.[31]

Since physico-chemical properties of films and edible coatings are extremely important, several researchers have tested different types of raw material to develop quality films. It is worth mentioning the plasticizing and antiplasticizing effects of glycerol and sorbitol in biodegradable films made from cassava starch.[32] Casting showed that, at the end of film preparation, storing conditions and contents of glycerol and sorbitol under study were important factors to determine plasticizing and antiplasticizing effects of glycerol and sorbitol. When values of water activity were low, films with low contents of plasticizers were less hydrophilic and flexible that non-plasticized films, a fact that suggests the antiplasticizing effect of glycerol and sorbitol. The comparison between glycerol and sorbitol showed that the former led to more effective plasticization which made films become more hydrophilic (due to increase in their capacity to interact with water) and more flexible.[32]

In addition, Sueiro *et al.* (2016) contributed to this scientific filed by describing preparation of biodegradable films made from cassava starch enriched with pullulan and bacterial cellulose.[33] The authors highlighted that hydrocolloids represent a new frontier to be explored in the search for highly efficient biodegradable material. One of the hydrocolloids is pullulan, a fungal $\alpha$-glucan produced by *Aureobasidium pullulan* and composed of repetitive subunits of maltotriose and a small number of maltotetraoses linked by $\alpha$ bonds ($1\rightarrow6$), which is compatible with starch.[33] Pullulan is easily soluble in water and produces colorless, odorless and atoxic paste and films. Studies of pullulan have shown that this biopolymer may be used as a vehicle of bioactive compounds, edible films and packaging of food and pharmaceuticals. Although it has many advantages, its high cost has prevented it from being broadly used by industries. On the other hand, several researchers have tried to decrease costs by using mixtures with other polysaccharides, such as alginate, chitosan, cellulose derivatives and starch.[33]

Another biopolymer that has been deeply studied is bacterial cellulose, which is produced by the bacterium *Gluconacetobacter xylinus* and has peculiar characteristics that make it differ from plant cellulose. Bacterial cellulose is pure, i. e., it is lignin-free and hemicellulose-free, has hydrophilic nature and exhibits higher crystallinity than most other sources of this biopolymer. These properties, together with its nanometric three-dimensional structure, enable it to have broad application.[33]

### 3.3. Starch as a promising alternative to replace synthetic polymers

It is estimated that there are about 121 PE producers that yield 70 million ton/year, on average, worldwide; it reached 88 million ton in 2010 with the significant contribution of developed and developing countries.[34] In Brazil, considering the production of rigid plastic and flexible films, 16.5% is recycled; it represents 200 thousand ton/year. The largest limitation of recycling is the diversity of resins, a fact that leads to difficulties in separating and reusing them. Difficulty in recycling most available synthetic packaging has triggered national and international studies to improve and/or develop biodegradable material that can be used in packaging.[34]

Biodegradation is a natural and complex process in which organic compounds are converted into simple mineralized compounds by biochemical mechanisms and then redistributed in the environment by element cycling, such as the cycles of carbon, nitrogen and sulphur.[34] In short, biodegradation of a polymer is an intrinsic process in which microorganisms and their enzymes consume it as a source of nutrients under normal conditions of moisture, temperature and pressure. The polymers that are best adapted to complete biodegradation are the natural ones, the ones that are hydrolysable to $CO_2$ and $H_2O$, or to $CH_4$, and synthetic polymers whose structures are similar to natural ones.[34]

Starch may be obtained from several plant sources, such as cereals, roots and tubers, besides fruit and vegetables. However, commercial starch is extracted from cereals, roots and tubers. Starch is the storage polysaccharide in plants, found in the form of granules which have a certain level of molecular organization. This fact bestows them partially crystalline or semicrystalline nature; crystallinity ranges from 20 to 45%.[35]

Starch is formed by two types of glucose polymers – amylose and amylopectin – with different structures and functionality. Amylose is a linear polymer composed of D-glucose units linked by $\alpha$-(1$\rightarrow$4) bonds. Degree of polymerization ranges from 200 to 3,000, depending on the source of starch. Amylopectin is a highly branched polymer, with D-glucose units linked by $\alpha$-(1$\rightarrow$4) bonds and branches in $\alpha$-(1$\rightarrow$6).[35] Variations in ratios among these components and in their structures and properties may result in starch granules with very different physico-chemical and functional properties, which may affect their industrial applications.[35]

Due to their semicrystalline nature, starch granules exhibit birefringence when they are observed in a polarized light microscope. The linear part of amylopectin molecules forms double helical structures which are stabilized by hydrogen bonds among hydroxyl groups, originating crystalline regions in granules. The amorphous region is composed of amylose chains and amylopectin ramifications.[36]

Starch application to film production is based on chemical, physical and functional properties of amylase to form gels and on its capacity to form films. Due to their linearity, amylose molecules in solution tend to orient parallel to each other and get close enough to form hydrogen bonds among adjacent polymer hydroxyls. As a result, affinity of the polymer for water decreases and favors development of opaque paste and resistant films.[36]

Even though the main commercial sources of starch are corn, potato, rice, wheat and cassava, both water yam tubers (*Dioscorea alata*) and oat grains (*Avena sativa*) are also promising sources of starch.[35] Water yam starch, by comparison with the previously mentioned starches, has higher mean amylose content, which is interesting to develop films. Regarding oat starch, despite its high variation in amylose and amylopectin contents, due to the different cultivars, its lipid content seems to be promising in film development. Films made from oat starch are more stable to environmental conditions due to its high lipid content, by comparison with starches from other sources.[35]

In order to produce thermo-plastic material based on starch, its semicrystalline granular structure must be disrupted to originate a homogeneous and essentially amorphous polymeric matrix. Both phenomena that enable disruption of the organization of starch granules are gelatinization and fusion. The former is the irreversible transformation of starch granules into viscoelastic paste that results from excess of water and leads to disruption of crystallinity and molecular order of granules by breaking hydrogen bonds which kept their integrity. On the other hand, when starch is heated in little amounts of water, the phenomenon that disrupts its granules is fusion, which requires much higher temperatures than gelatinization to take place.[35-37]

After gelatinization, starch molecules may start to re-associate through hydrogen bonds and favor the development of more ordered structures, which, in favorable conditions, may form a new crystalline structure. This alteration is called re-crystallization, the most important phenomenon that leads to aging of starch films and makes them rigid and brittle. It should be emphasized that storage conditions, in general, may accelerate or delay re-crystallization.[35]

### 3.4. Tape casting and casting

These techniques have been commonly used because they help to improve physico-chemical properties, since they enable other material, such as cellulose fibers, to be incorporated into films.

Tape casting (also known as spreadcasting and knifecoating) has been well-known in ceramic, paper, plastic and paint industries. In the process, slurry is placed in a reservoir which has a leveling blade called doctor blade, whose height may be adjusted by micrometric screws.[38] Slurry is cast in a thin layer onto a surface (polymeric tape), as the result of either the movement of the surface (continuous process) or the movement of the blade (discontinuous process). Spreading velocity depends on slurry characteristics and configuration of the leveling blade, such as height of the spreader and volume of the reservoir. Tape casting is a promising method to increase scale of production of biodegradable films since knowledge developed by ceramic and paper industries can be applied to the new field.[38]

Evaluation of rheological properties of suspensions that form films is important since they are transported and processed. Suspensions that are subject to tape casting should exhibit pseudoplastic behavior, which is shown by suspensions of starch-glycerol-cellulose fibers. Non-Newtonian fluids should be characterized not only by viscosity but also by viscoelasticity. Oscillatory assays, which have often been used for characterizing polymeric material, enable to separate the elastic contribution of the polymer from the viscous one in relation to time or frequency.[38]

Evaluation of variation in viscoelastic storage modules (G') and loss modules (G'') at a frequency of oscillation ($\omega$)

enables to make deductions about the macromolecular organization of a system. In general, in dilute polysaccharide solutions at low frequencies of oscillation, the viscous nature of the system is very high. The higher the polysaccharide concentration or frequency of oscillation, the higher both G' and G"; as a result, the elastic component may outweigh the viscous one.[38]

In film production, drying time and energy consumption are expected to decrease with no loss of quality (of mechanical properties, for instance). High drying rates may lead to large gradients of concentration and form a vitreous superficial layer, with a central region in the film with high moisture content. Increase in temperature increases vapor pressure, which may lead to bubble development, a fact that damages film quality. Although most studies have reported suspension drying either at room temperature or in forced air ovens at moderate temperatures, recent studies have shown that drying time can be decreased.[38]

Film development by casting, followed by drying of suspensions made from starch-glycerol and starch-sorbitol by convection – at temperatures ranging from 25.9 to 54.1 °C and relative humidity (RH) ranging from 33.8 to 76.2%, showed that drying velocity influenced mechanical properties and solubility of films.

When films are produced in laboratory scale, they are developed after starch solubilization in a solvent; a filmogenic solution is formed and immediately placed on a surface to dry after solvent total evaporation. This technique is called casting.[34] In this technique, in the step after thermal gelatinization of granules in excess of water, for instance, amylose and amylopectin disperse in the aqueous solution and reorganize during the drying process, thus, forming a continuous matrix that originates films.[34]

Crystalline structures of starch films and, consequently, mechanical and barrier properties, are deeply influenced by their drying conditions. In dry films at temperatures above 60 °C, the drying process is faster than re-crystallization (a process that leads to film aging) and generates material that is more stable to storage than films dried at lower temperatures.[34] RH throughout film drying is also an important factor since films dried at high RH exhibit structures with high crystallinity degree and high content of residual humidity, a parameter that makes films more susceptible to alterations in storage and use.[34]

Casting has been used and discussed in research into biodegradable films based on starch and has shown good results in laboratories lately.[34] However, in the case of production in industrial scale, it has some disadvantages, such as long processes and high cost, mainly because of the cost of electrical energy needed to dry films. A solution would be the use of extrusion to produce films in industrial scale; advantages would be quickness and lower production costs. Another important factor is that this process has already been used for producing conventional synthetic packaging, such as the one made from PE, PP and PET.[34]

### 3.5. Active films incorporating essential oils and plant extracts

Protection properties of films may be reinforced by incorporation of natural products (essential oils and plant extracts) which release compounds that increase shelf life of food. Natural products have been used because they have several biological activities, such as antimicrobial, bacterial, fungicide and antioxidant ones.[39] It should be highlighted that, since consumers have required safe food and have worried about side effects of artificial additives, it is very important to carry out studies of new natural and atoxic additives in order to replace artificial ones and keep food quality.[39]

Active biodegradable films may incorporate essential oils or any bioactive compound which interacts directly with food and bestows them desirable sensory and nutritional attributes.[40] Essential oils extracted from aromatic plants are concentrated hydrophobic liquids that contain several bioactive compounds, such as phenolics and terpenoids. It has been shown that essential oils exhibit strong antimicrobial and antioxidant activities, which may be potential preservative agents of perishable foods.[40] Mahcene *et al.* (2020) revealed the power of different essential oils when they were incorporated into sodium alginate-based films.[40] On the other hand, essential oils from *Citrus limonia* leaves were also promising to develop chitosan-based films.[41] The authors also described essential oils as complex mixtures of volatile organic compounds consisting of oxygenated compounds and hydrocarbons, such as sesquiterpenes and monoterpenes, produced by aromatic plants as secondary metabolites. These compounds are responsible for the antimicrobial activity of essential oils and have mechanisms associated with their lipophilic nature, which interacts with microbial membranes leading to leakage of cell compounds and causing energy loss from microbial cells.[41] Much bioactivity exhibited by essential oils has drawn researchers' interest worldwide. They have developed and tested their bioactive films incorporating essential oils extracted from different plant species, such as clove.[42]

Plant extracts, which are natural products rich in bioactive secondary metabolites, have had their incorporation into films and edible coatings widely explored. The literature has described that plant extracts exhibit important antioxidant and antimicrobial compounds and, just like essential oils, may replace synthetic additives (e.g., sorbate and benzoate) and have increased in foodstuff due to their safety, biocompatibility, biodegradability and low toxicity.[43] For instance, edible chitosan-based films – pure or mixed with gelatin – have already been tested, with and without boldo.[43] Other active extracts from *Garcinia atroviridis*[44], *Eugenia brasiliensis*[45], orange peel[46], mango kernel[47], *Acca sellowiana*[48], *Thymus serpyllum*[49] and *Capsicum chinense*[21] were incorporated into different compositions of edible films and had satisfactory results.

### 3.6. Films, edible coatings and their effects on human health

As discussed before, films and edible coatings are layers – of different thickness – composed of distinct natural and/or synthetic compounds which polymerize and promote protection/isolation of food from external agents, with no risks to consumers' health since they are not metabolized by the body and their passage through the gastrointestinal tract is innocuous.[50]

They are called innovative packaging worldwide because they are considerably different from old and non-biodegradable packaging which not only polluted the environment but also poisoned consumers with metals, such as cadmium and mercury, and other residues.[51] Concerning safety and human health, the food industry and new research patterns have reassured consumers that human health is fundamental and made sure the environment is clean by providing food that is thoroughly entire, protected and healthy.[51]

## 4. Conclusion

This study compiled promising data on preparation and application of different types of films and edible coatings. They are different possibilities that may be used to protect food, from fruit to meat. It deals with the so-called innovative, sustainable and mainly biodegradable packaging that may replace synthetic plastic that is harmful and environmentally unfriendly. This paper aimed at clarifying that films and coatings exhibited distinct forms. Films are thin membranes that are pre-formed separately from products, for instance, by casting, which consists in applying a filmogenic solution to a surface up to drying and film formation, followed by its application to food. Coatings or covers are suspensions or emulsions that are directly applied to food surfaces and become films. In short, bio-packaging has shown to meet requirements of food protection; its use suggests savings related to loss of food due to natural maturation, and extension of shelf life of products.

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