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Filmes e revestimentos naturais comestíveis aplicados em alimentos: uma revisão bibliográfica

Natural edible films and coatings applied in food: a bibliographic review

Películas y recubrimientos comestibles naturales aplicados en alimentos: una revisión bibliográfica

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Resumo

Hoje em dia, a produção mundial de plástico gera muitos resíduos. Portanto, a necessidade de reduzir os materiais plásticos convencionais não biodegradáveis tem incentivado o desenvolvimento de materiais biodegradáveis inovadores a partir de recursos renováveis. Os filmes de polímeros de base biológica podem ser desenvolvidos usando fontes de ocorrência natural como polissacarídeos, proteínas, lipídios e suas combinações. Polímeros naturais comestíveis são os materiais feitos de constituintes naturais comestíveis que podem ser consumidos por animais ou por seres humanos sem risco para a saúde. Além disso, existem aditivos que também podem ser usados para melhorar as características da embalagem, como óleos essenciais, antioxidantes. Este artigo de revisão tem como objetivo fazer uma extensa revisão de literatura e comparação de dados encontrados por diversos autores em relação aos vários polímeros naturais utilizados na confecção de revestimentos e filmes comestíveis aplicados em produtos alimentícios, e com isso mostrar que seu uso pode melhorar suas características e prolongar a vida útil. Foram utilizados mais de 130 referências bibliográficas para a montagem deste artigo de revisão e foram utilizadas várias plataformas de pesquisa (Scielo, google acadêmico, entre outras). Após a extensa revisão de literatura é possível concluir que o uso de polímeros obtidos de fontes naturais são eficazes para conservar alimentos por mais tempo e com isso ocasionar uma diminuição da perda de vida útil desses produtos.

Palavras-chave: Biodegradável; Alimento; Aditivos naturais; Polímeros.

Abstract

Nowadays, the world's plastic production generates lot of plastic waste. Therefore, the need of reducing conventional nonbiodegradable plastic materials has encouraged the development of innovative biodegradable materials from renewable resources. Biobased polymer film can be developed using naturally occurring sources like polysaccharides, proteins, lipids, and their combinations. Natural edible polymers are the materials made from natural edible constituents that can be consumed by animals or human beings with no health risk. Also, there are additives that could be used as well to improve the package characteristics, such as essential oils, antioxidants. This review article aims to make an extensive literature review and compare data found by several authors in relation to the various natural polymers used in the manufacture of edible coatings and films applied in food products, and thereby show that their use can improve their characteristics and prolong the shelf life. More than 130 bibliographic references were used to produce this review article and several research platforms were used (Scielo, google academic, among others). After the extensive literature review, it is possible to conclude that the use of polymers obtained from natural sources is effective for preserving food for a longer time and thus causing a reduction in the loss of shelf life in these products.

Keywords: Biodegradable; Food; Natural additives; Polymers.

Resumen

Hoy en día, la producción mundial de plástico genera muchos residuos. Por lo tanto, hay necesidades de reducir los materiales plásticos convencionales no biodegradables, fue incentivado el desenvolvimiento de materiales biodegradables innovadores a partir de recursos renovables. As películas de polímeros de base biológica puede ser desenvuelto usando fuentes naturales como polisacáridos, proteínas, lípidos y sus combinaciones. Polímeros naturales comestibles son los materiales hechos de constituyentes naturales comestibles que pueden ser consumidos por animales o por seres humanos sin riesgo para la salud. Además de eso existen aditivos que también pueden ser utilizados para mejorar las características del embalaje, como aceites esenciales, antioxidantes. Este artículo de revisión tiene como objetivo realizar una extensa revisión de la literatura y comparar los datos encontrados por varios autores en relación a los distintos polímeros naturales utilizados en la fabricación de recubrimientos y películas comestibles aplicados a productos alimenticios, y así demostrar que su uso puede mejorar su características y prolongar la vida útil. Se utilizaron más de 130 referencias bibliográficas para producir este artículo de revisión y se utilizaron varias plataformas de investigación (Scielo, google scholar, entre otras). Tras la extensa revisión de la literatura, es posible concluir que el uso de polímeros obtenidos de fuentes naturales es eficaz para conservar los alimentos por más tiempo y así provocar una reducción en la pérdida de vida útil de estos productos.

Palabras clave: Biodegradables; Alimento; Aditivo natural; Polímeros.

1. Introduction

Edible films and coatings can be produced from edible polymers and food additives. They consist of a thin layer of edible material that coats the food directly, or a film which can wrap food without changing its original ingredients or processing method (Azeredo & Walddron, 2016; Wihodo & Moraru, 2013; Zink et al., 2016). Films and coatings have been used to maintain quality and safety, helping to prolong the shelf life of many products (Pavlath & Orts, 2009).

Edible coatings are being used in minimally processed food as a strategy to reduce the effects the minimally process causes in vegetable tissues (Chevalier et al., 2018). Besides, edible coatings can contribute to the increase the life of minimally processed fruits, reducing its moisture and solute migration, respiration and oxidation reactions (Rojas-Graü et al., 2009). Edible coatings can also be applied in the liquid form of the film-forming dispersion, or as thin sheets (films) used to wrap food products. Both are well considered for food preservation purposes due to their ability to improve food quality (Chillo et al., 2008).

In food industry packages are important to products conservation. However packaging attends consumers' expectations protecting food, they still need improvement in their structure, especially when talking about environmental and economic viability, and practicalness (Soares et al., 2009). Plastics are highly versatile materials with excellent physical and chemical properties, aesthetic quality manufactured at low cost, and have numerous applications including food packaging, consumer products, medical devices and construction. Today, the world's plastic production is more than 320 million tons per year, of which more than 40% is used as disposable packaging material to generate plastic waste (Roy & Rhim, 2019). Due to the non-biodegradable and non-renewable nature of petroleum-based plastics, waste plastics cause serious environmental pollution problems and are paying considerable attention to the development of biodegradable, eco-friendly packaging materials that use renewable resources such as biopolymers instead of plastics (Corrales et al., 2014).

The continuous growth in plastic use has brought an increasing environmental awareness from the perspective of waste management, emissions released during manufacture, and resources use (Rasheed et al., 2016). Therefore, the need of reducing conventional nonbiodegradable plastic materials has encouraged the development of innovative biodegradable materials from renewable resources (Garrido et al., 2019).

The biobased polymer film can be developed using naturally occurring sources like polysaccharides, proteins, lipids, and their combinations (Roy & Rhim, 2019). In this connection, various types of polysaccharides have been widely used for their excellent film formability and suitable structure and mechanical properties. Biodegradable films are made of biologic material and they can serve as packaging (Safaei & Taran, 2018).

The biodegradable characteristic is one of their greatest benefits. There are even applications for non-food products including films for agricultural uses, grocery bags, paper coatings, or cushioning foams. Many functions of biodegradable packaging are similar to synthetic packaging materials and for that, it’s possible to say they could, at least partially, replace them, which would help to reduce the environmental impact of the massive use of synthetic plastics (Chiralt et al., 2018).

Nowadays there is more interest in improving quality in packaged products, and at the same time, reducing the waste of packaging has encouraged studies of new materials for biodegradable films, made of natural resources (Souza, Silva & Druzian, 2012). Among biodegradable materials for films and coatings production the most used are polysaccharides, such as starch, and proteins. There are additives that could be used as well to improve the package characteristics, such as essential oils, antioxidants.

This review article aims to make an extensive literature review and compare data found by several authors in relation to the various natural polymers used in the manufacture of edible coatings and films applied in food products.

2. Metodology

The present study is a literature review on research carried out in the area of films and natural edible coatings applied to food and the methodology used was a bibliographic research. The review covered scientific and review articles, both national and international, published and available in the databases of Scielo (Scientific Electronic Library Online), Google Scholar and Science direct. Bibliographies that did not present important data about the use of films and coatings in food were excluded.

3. Results and Discussion

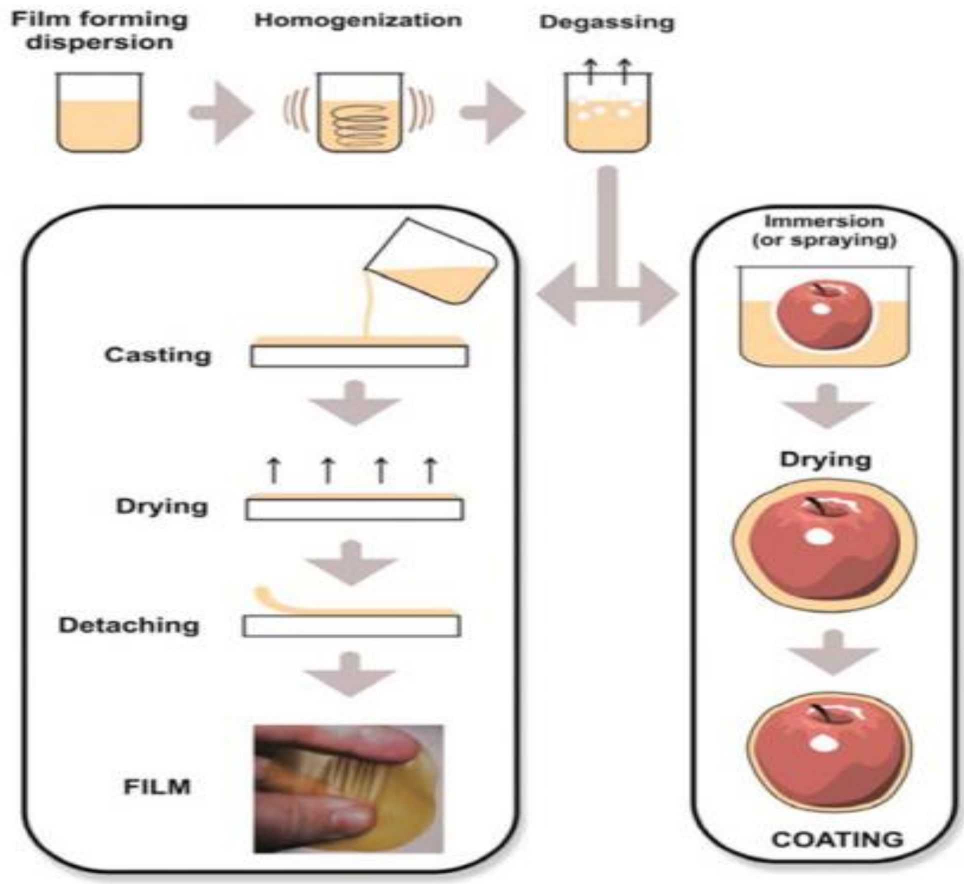
The present study, resulting from a bibliographic review, using more than 130 articles, is divided into topics, which are described below.

3.1 Edible films and coatings obtained from natural polymers

A definition for edible coatings and films is that they are primary packaging made from edible ingredients. Also, it is possible to apply directly a thin layer of edible packaging in the food by coating, immersion, and spraying. Also, previously formed film could be used as a food packaging without changing the process method of the coating and the materials used (Galus & Kadzińska, 2015). These authors mentioned that composite ingredients can be obtained as emulsions or bilayers. In the case of the bilayer system, it is required to create a thin layer firstly of polysaccharide or protein and over this; the second layer is formed of dispersed lipids. The bilayers are less common in the food industry as they need two casting and two drying stages, despite they are providing good barriers against water vapor.

The Figure 1 showed different methods of application to coatings and edible films. The difference between coating and the edible film is that films are obtained as solid laminates then applied to foodstuff while coatings are applied in liquid methods (Falguera et al., 2011; Mohamed et al., 2020).

Figure 1 - Schematic representation for production of films and coatings.



Source: Otoni et al. (2017).

Coatings and edible films are not expected to fully replace conventional wrapping materials, they can be used to extend food stability by reducing the exchange of moisture, lipid, volatiles, and gasses among the food and the surrounding environment. Avoiding surface contamination increased the efficiency of food packaging, and thus reducing necessities for petroleum-derived polymers (Tian et al., 2020).

Coatings and edible films present barriers that depend on the properties of the food that is to be protected. For example, coatings or films for vegetables and fresh fruits should have low water vapor permeability to reduce the rates of desiccation, whereas oxygen permeability should be low enough to retard respiration, but not too low to produce anaerobic conditions favorable to off-flavor formation and ethanol production. New packaging constituents have been developed from natural sources and characterized; but despite that, the presented information for the preparation of food enclosed is not universal for all products, which poses a challenge for the progress of specific films and coatings for each food (Mohamed et al., 2020).

Have various studies about the use of natural sources to obtain edible coatings and films. Table 1 shows recent studies that used different coatings or films obtained from natural sources to increase the shelf life of different foods.

Table 1 - Different natural sources utilized from coatings or films to increase the shelf life of foods.

Source to form coatings/films	Concentration	Application of edible coatings/films	Main results	References
Edible coatings with starch extracted from the pulp of the fruit of banana "Pear," soursop, and stenospermocarpic mango "Ataulfo".	2%	Stenospermocarpic mango (*Ataulfo*) fruit	The starch-based coating extended the shelf life of mango fruit up to 15 days. The fruit stored at 10 °C for 10 days no showed no unfavorable alterations in firmness, color, and total soluble solids.	(Hernández-Guerrero et al., 2020)
Edible coatings of starch extracted from rice grains.	3%	Apple(*Cripps pink*) fruit	Apple fruit coated with starch extracted from rice showed a reduction in weight loss, respiration rate, firmness, skin colour change.	(Thakur et al., 2019)
Edible coatings obtained of the mix of cassava starch, whey protein, beeswax, chitosan, glycerol, stearic acid and glacial acetic acid.	3.5%	Andean blackberries (*Rubus glaucusBenth*) fruits	The application of this mix of edible coatings to Andean blackberries had a positive effect on the physicochemical properties such as pH, acidity, soluble solids, total solids, antioxidant capacity, phenolic compounds, and	(Rodríguez et al., 2020)

			anthocyanins.	
Edible coatings obtained of the cassava starch and starch nanocrystals.	6%	Huangguan pears (*Pyrus pyrifolia*, Nakai)	Coating of cassava starch together with starch nanocrystals can maintain the color, texture, cell membrane permeability, total phenolic, soluble solids and titratable acid contents of the pears and inhibit the peroxidase enzyme and polyphenol oxidase enzyme activities.	(Dai, Zhang and Cheng, 2020)
Edible coatings obtained blend based on cassava starch, gelatin, and casein.	Between 2 to 4%	Guava	The polymeric blend-based was efficient in retarding the fruit ripening process, increasing the shelf life of guavas by 2 days.	(Pellá et al., 2020)
Edible coatings of the chayotextle starch supplemented with different concentrations of resistant starch microcapsules.	4%	Guava	Chayotextle starch coatings were the most effective in delaying maturation changes in guavas, such as titratable acidity, pH, and total soluble solids.	(Martínez-Ortiz et al., 2019)
Film with whey Protein Nanofibrils (WPN) and titanium dioxide nanoparticles (TNP).	WPN 5% TNP 1%	Chilled beef	This combination can prevent lipid oxidation and food deterioration. Can help limit lipid peroxidation and promote antioxidant activity in beef. Addition of titanium	(Feng et al., 2019)

			dioxide can reduce microbial growth and extend the shelf life of chilled beef.	
Film of poly lactic acid with zinc oxide and different essential oils.	1.5%	Tiger Tooth Croaker fish flesh	The application of film of poly lactic acid with zinc oxide and different essential oils could extend the shelf-life of fish fillets in refrigerator up to 16 days as compared to the unwrapped samples with 7 days shelf life, which was supported by the results of the microbial counts and chemical evaluation.	(Heydari-Majd et al., 2019)
Edible coatings soybean protein isolate and chitosan	soybean protein isolate (5%); Chitosan (0.4%)	Apricot fruit (*Prunus armeniaca L.* ‘*Kaite* ’)	The soybean protein isolate and chitosan coating had a beneficial effect on weight loss, retention of firmness, titratable acidity and soluble solids content, as well as retention of the water soluble pectin contents of fruit.	(Zhang et al., 2018)
Edible coating with sodium alginate, pectin and whey protein concentrate.	1.5%	Mini-buns	The results of the presented study show that the application of coating has a retaining effect on the product to some extent. The use of these edible coatings was important to	(Chakravartula et al., 2019)

reduce de water loss and moisture content in the bread.

Source: The authors.

4. General Characteristics of Edible Films

As films and coatings cover materials, they should have packaging properties that protect the inner part from the outside, limiting gas and water vapor transportation between the food product and the environment (. Erkmen & Barazi, 2018). And because of the ‘edible’ term, films can be consumed together with foods they are in contact with, so they need to be considered Generally Recognized As Safe (GRAS) (Erkmen & Bozoglu 2016).

Besides that, there are other important characteristics edible films and coatings should have, which are (Pavlath & Orts, 2009):

- Not contain any toxic, allergic or non-digestible components;
- Provide structural stability to prevent mechanical damage during transportation, handling, and display;
- Have good adhesion to surface of food to protect and to provide uniform coverage;
- Have a control water migration both in and out to protect food and maintain the desired moisture content;
- Provide semi-permeability to maintain internal equilibrium of gases involved in aerobic and anaerobic respiration, thus retarding senescence;
- Prevent loss or degradation of components that stabilize aroma, flavor, nutritional and organoleptic characteristics, which are necessary for consumer acceptance;
- Provide biochemical and microbial surface stability while protecting against contamination;
- Maintain or enhance aesthetics and sensory attributes (appearance, taste etc.) of product;
- Be able to serve as carrier for desirable additives such as flavor, fragrance, coloring, nutrients, vitamins, antioxidants and antimicrobial agents, and
- Be easily manufactured and economically viable.

5. Polysaccharides

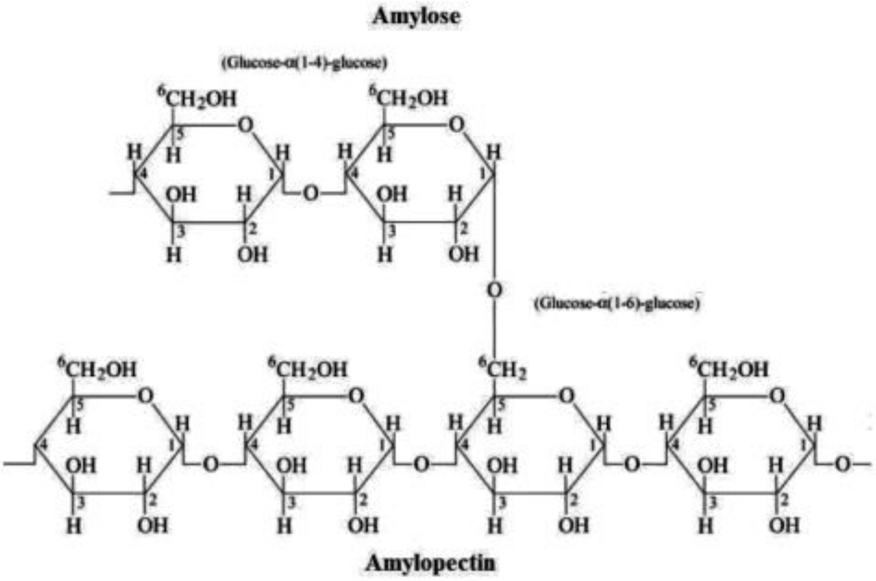
Polysaccharides are widely studied as films and coatings in food. Several polysaccharides are studied, such as starches, gums, fibers. In this work, we will discuss the use of starch, chitosan, xanthan and fibers.

5.1 Starch

Starch is the main carbohydrate which is stored as a food material in plants. It contains two types of glycose units, amylose and amylopectin. Both have α (1→4) linkage between the polymers in short or long chains. Amylose has a linear chain, while amylopectin can have branches due to α(1→6) linkage (Preiss, 2018). Starch is found in the form of semi-crystalline hydrophilic granules. Starch granules absorbed water molecules surrounding the free hydroxyl groups, that make starch granules swell, then swelling continue till achieved a critical concentration. The critical concentration is the required concentration of starch to make the swollen granules occupy the entire volume at 95 °C; on cooling gel is formed (Pelissari et al., 2019).

In the Figure 2 it is possible to observe the chemical structure of amylose and amylopectin.

Figure 2 - Chemical structure of amylose and amylopectin.



Source: Buléon et al. (1998).

The formation of starch films starts with the heating of starch granules in excess water to prepare a viscous solution. Aqueous solutions are normally unstable and tend to gel immediately upon cooling due to the association of polymer chains (glycose units) (Wang et al., 2015). At high temperature and in excess water, starch granules gelatinize, transforming from a semicrystalline phase into an amorphous state (Ratnayake & Jackson, 2008). The loss of crystallinity happens in two steps, the first when the starch molecule swells, normally at 60-70 ˚C (Shanks & Kong, 2012). The second step is observed at temperatures above 90 ˚C and results in excessive granule swelling and solubilization, which leads to a complete loss of structural integrity (Ivanič, 2017).

Starch is known for being biocompatible, biodegradable, nontoxic and for having low cost (Mahmoudian et al., 2012; Qiao et al., 2016). This material is relatively easy to handle. Besides being totally biodegradable, is also widely available in nature from sources such as cereals, roots, tubers and palms (Araujo-Farro et al., 2010; Colla, Sobral & Menegalli, 2006).

Several authors studied the development of starch films from different sources, which ones found good barrier results in their work on the development of green peas starch films; and quinoa starch films (Araujo-Farro et al., 2010). Other authors studied films physical properties of different native starches, such as extraction of potato, cassava and rice starch, added with plasticizers (glycerol and sorbitol) and montmorillonite clay (Borges et al., 2015); and found out that rice starch films had higher elongation values and low water solubility, when compared to potato and cassava starch films, which shows potential for coating food products.

There are studies about extrusion technology as a previous treatment of the casting technique to modify starch structure in order to obtain edible films with improved physicochemical properties, using corn starch (Fitch-Vargas et al., 2016). This study indicates that when combining extrusion and casting technique corn starch edible films showed greater breaking strength and deformation, as well as lower water vapor permeability, which says that these films could be applied on food products to improve their preservation, distribution and marketing.

Starch is recently receiving more attention as a material to be used as packaging. Some researchers studied the comparison of laboratory-induced wheat grain germination and pre-harvest sprouting in the field and their effects on starch characteristics and application in films (Baranzelli et al., 2019). They found that elongation of starch films increased significantly with wheat germination time, but the germination process did not significantly affect the opacity, water solubility, and water vapor permeability of films. With that, it is possible to consider the use of starch from germinated wheat grains in films, adding value to a commonly discarded product due to its inability to be used in baking.

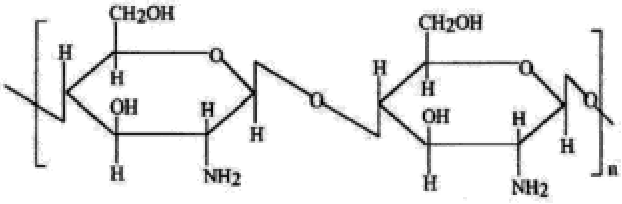
Although starch itself is a good material to work with, there are studies about starch combined to other materials. A study of the dissolution of films based on starch blended with agar and maltodextrin showed great results. The treatments using only starch showed the higher values of solubility in water and acid when compared to the blended treatments. Authors found that the addition of agar improved the barrier properties of films (Wongphan & Harnkarnsujarit, 2020).

In a research about a polymer blend coating based on cassava starch, casein, and gelatin, and using sorbitol as the plasticizer, applied to guavas, the best results obtained were the treatments with higher quantities of starch and casein and low values of gelatin, which were able to retard the fruit ripening process, increasing and increase the shelf-life of the fruits. It confirms the efficient use of starch itself, but also opens the idea of blending for use in coatings and edible films (Pellá et al., 2020). Emamifar, Ghaderi & Ghaderi (2019) related that the application of salep coating as a polysaccharide-based coating can improve physicochemical, sensorial, and microbial quality of fresh strawberries during cold storage.

5.2 Chitosan

Chitosan is a natural non-toxic polymer, biodegradable and economic available, which are reasons for its use in a variety of applications in food industry, counting on the quality control as a substitute for non-biodegradable polymers. Chitosan can form transparent films; it becomes versatile and can fit in any packaging application (Elsabee & Abdou, 2013; Mohamed et al., 2020). Coating food with chitosan films lowers the oxygen partial pressure in the package, keeps the temperature with moisture transfer between food and its environment, delays enzymatic browning in fruits, controls respiration and declines dehydration. Besides, chitosan is used on enhancing of the emulsifying effect, increasing the natural flavor, setting texture, deacidification and stabilization of color (Duran & Kahve, 2016). Chitosan based films are clear, flexible and tough. They show good resistance to fat and oil, oxygen, but they are highly sensitive to moisture (Nayik, Majid & Kumar, 2015). In the Figure 3 is possible verify the chemical structure of the chitosan.

Figure 3 - Chemical structure of chitosan.



Source: Naira & Laurencin (2007).

A study comparing edible coatings based on chitosan and xanthan gum showed the efficiency of chitosan in relation to attributes, such as color, texture, loss of vitamin C and weight, and sensory attributes. It showed that the use of chitosan as coating was more satisfactory in extending the shelf life of minimally processed broccoli for up to 12 days in refrigerated storage when compared to control and the xanthan gum treatments (Pizato et al., 2020).

In a study of the effects of the physicochemical and microbiological qualities of sweet cherry (*Prunus avium L.*) coated with chitosan, authors found that coated cherries had better inhibition to microorganism than control. Besides that, it was also observed the coating provided lower water activity values to the cherries, when compared to control treatments. Although chitosan coatings have not been effective at reducing water loss in this research, it shows potential in its antimicrobial effect (Tokatlı & Demirdöven, 2020).

Combining antimicrobial agents such as plant essential oils directly into food packaging is a form of active packaging. Because of that, researchers studied chitosan-based films containing cinnamon essential oil (CEO) (Ojagh et al., 2010). With the incorporating of the CEO into the films it was possible to observe changes in the properties of the films, such as an increased antimicrobial activity, and also a decreasing in moisture content, solubility in water, water vapor permeability and elongation at break of chitosan films.

Another study confirmed the great interaction between chitosan and an antimicrobial extract (Nguyen et al., 2020). Author developed chitosan films in addition to the leaf extract of *Sonneratia caseolaris L. Engl.* (SCELE) and applied in bananas. The results obtained indicated that films with SCELE had their antimicrobial activities and water vapor barriers improved when compared to control films containing only chitosan.

According to some authors (Fitzpatrick et al., 2013; Veiga-Santos et al., 2005); although chitosan's potential in biodegradable packaging, it shows low values of elongation and high values of solubility and water vapor permeability in films. An alternative to improve its characteristics would be combining chitosan with other polymers, such as xanthan gum. A study of properties of chitosan films added to different concentrations of xanthan gum showed that the combining improved mechanical properties of the films, as high tensile strength and low elongation (De Morais Lima et al., 20017).

Several authors studied the use of chitosan as coatings or films added to complements (De Morais Lima et al., 2017; Veiga-Santos et al., 2005). There is a study on the evaluation of the effects of chitosan coating associated with montmorillonite clay and essential oil of cloves in minimally processed melon (*Cucumis melo L.*) in which the authors were able to notice that the association of chitosan with the other components showed better results than just chitosan, in order to prolong shelf life and retard microorganism growth (Veiga-Santos et al., 2005). In other study authors developed films based on chitosan added with xanthan gum, and protein hydrolysate of Whitemouth croaker (*Micropogonias furnieri*) (De Morais Lima et al., 2017). The addition of xanthan gum increased tensile strength and changed the color parameters of films, and the addition of protein hydrolysate increased the antioxidant activity of the films. It was not observed significant differences in water solubility and water vapor permeability by the addition of xanthan gum and protein hydrolysate.

Another research using chitosan shows the success of its use as coating. Authors developed chitosan (Q) and alginate (A) coatings, in different proportions, added to zinc oxide nano particle (nanoZnO). They were applied to guavas (*Psidium guajava L*) in order to extend their shelf-life. Besides the antimicrobial effect of chitosan, ZnO has an antibacterial action. The treatments were compared in aspects of water loss, texture, color, rot index, and physic-chemical analyses. The results showed that treatments with greater quantities of chitosan (100%Q or 90%Q-10%A) were useful to retard guavas senescence (Arroyo et al., 2020).

5.3 Xanthan gum

Xanthan gum is a water-soluble polysaccharide synthesized by Xanthomonas and can be used as emulsifier, suspension stabilizer, flocculant, gelling and viscosity agent. It is widely used in different industrial applications (Oliveira et al., 2013); such as in food, toiletry, oil recovery, pharmaceutical and cosmetics industry.

Xanthan gum is considered safe, biodegradable and effective in forming a cohesive and continuous matrix with uniform physical and chemical properties (De Morais Lima et al., 2017; García-Ochoa et al., 2000). Besides, xanthan gum film forming properties embrace pseudo-plastic rheological behavior in an aqueous environment that is favorable to film production due to its readily dispersion in cold or hot water with very little effect on its viscosity from either temperature or pH (Baldwin, Hagenmaier & Bai, 2011).

Some authors had applied xanthan gum coating in fresh-cut apple added to calcium chloride, ascorbic and citric acid, in order to carry these preservative agents, to extend the shelf life of the apples (Freitas et al., 2013). They noted that xanthan gum was effective in carrying agents in order to avoid browning and lower microorganism growth.

In another study, Cortez-Vega et al. (2013) showed the comparison between polysaccharides on the conservation of minimally processed papaya with xanthan gum-based coatings applied in it, in different treatments, added to guar gum and chitosan. They found that the coating with 100% xanthan gum showed the best results in reduction of mass loss when comparing to the others, which shows the use of this material is promising.

5.4 Fibers

Composites in forms of fibers have been widely used in plastic industries to reach desirable properties or to reduce prices. When compared to inorganic composites, the organics have some advantages, such as being originated from renewable natural sources, have great availability, low energy consume, low cost, low density and reactive surface, which could be used to introduce specific groups (El Halal et al., 2018).

Researchers developed and evaluated films produced from potato and cassava starches, reinforced with cellulose fibers and/or nanoclay (Anglès, Salvadö & Dufresne, 1999). These components increased the tensile strength of the cassava starch films and decreased water vapor permeability, showing that the addition of reinforcement agents in potato starch films produced more resistant films.

6. Proteins

Among the natural biopolymers most used in film making are polysaccharides and proteins, because they are from renewable sources and have the capacity of forming a continuous and cohesive matrix on film development (Rhim & Ng, 2007). The properties of this matrix depend not only on the material type, but also in the composition and processing methods and conditions. For this reason, proteins have been widely explored for the development of antimicrobial films (Da Rocha et al., 2018). Proteins can be found naturally, as fibrous proteins or globular proteins; globulars are rolled over their selves, and the fibrous ones are bonded to each other on parallel (Enujiugha & Oyinloye, 2019).

A recent study shows the potential of egg proteins in the composition of edible films (De Pilli, 2020). The author developed films from a mixture of vegetable oil, egg proteins and hydrocolloid components and applied in sweet baked products, such as cake, with the objective of the substitution of industrial preservatives. The films produced had gas barrier similar to polyethylene terephthalate (PET), which indicates good barrier properties. Beyond that, the films were able to retard changes in the color and texture and inhibited microorganism growth. (Ulutasdemir & Cagri-Mehmetoglu, 2019) showed improved whey protein concentrate (WPC)-based coating containing *Williopsis saturnus* can be applied on surface of roasted peanuts to prevent growth of *Aspergillus flavus* and aflatoxin production.

6.1 Fish protein

The waste of fish industrialization normally is used to process flour, or it is just discarded. An alternative to use this waste is recovering its main components, among them the proteins (Oetterer et al., 2006). The development of fish protein films can add value to a material that would be discarded, contributing to the reduction of environment impacts and offering a viable and low-cost alternative.

Fish-protein based films form a chain that presents good mechanical properties, like plasticity and elasticity, and good oxygen barrier. Although it absorbs a lot of water due to the hygroscopicity of the protein amino acids. This characteristic can be changed by adding plasticizers and/or additives (Paiva, Morales & Guimarães, 2006; Zavareze et al., 2012).

Researchers have been studying the use of fish protein isolate to develop films and coatings, and their application in different food products. A study of nanocomposite films produced from Whitemouth croaker (*Micropogonias furnieri*) protein isolate with montmorillonite showed promising results from the standpoint of mechanical properties, visual appearance and easy handling, as well as for their low water vapor permeability and low water solubility (Cortez-Vega et al., 2014). Other authors also evaluated films from Whitemouth croaker protein isolate (CPI) modified with montmorillonite (MMT) and had similar results about the decrease in the solubility, transparency and water vapor permeability (WVP) (Pizato et al., 2015).

In other works, authors used croaker coating applied in fresh-cut papaya (*Carica papaya L.*) (Cortez-Vega et al., 2014); and fresh-cut pear (*Pyrus communis L.*) (Pizato et al., 2015). In both works they did three different treatments, T1 for control, T2 with croaker coating and T3 with croaker coating added to montmorillonite. For both works the result was close. Both treatments with the croaker coating showed low mass loss, low microbiological growth, small decrease of firmness, lightness and pH. However, the treatment with montmorillonite added showed the best results for coating.

In a study of physical, mechanical and antimicrobial properties of protein films from Argentine anchovy (*Engraulis anchoita*) incorporated with organic acids (sorbic or benzoic acids), authors found that the increase in concentrations of the acids resulted in greater thickness, color difference, opacity and elongation at break, but decreased tensile strength of the films (Rocha et al., 2014). They also observed that the more percentage of organic acids, higher the values of water vapor permeability. The films were effective against the microorganism tested, except for *Staphylococcus aureus* that showed no inhibition in any of the organic acid concentrations tested.

An investigation of the influence on different times of cold plasma application as a surface modification strategy for films prepared from fish myofibrillar proteins showed changes in the mechanical performance, water vapor permeability, solubility in water and color properties (Romani et al., 2019a). Films treated for 2 minutes showed increased elongation at break and decreased tensile strength, while the opposite behavior was observed after 5 minutes of treatment. Solubility in water increased with 5 minutes and water vapor permeability increased with 2 minutes of plasma treatment. Color and opacity also increased. According to some authors (Chu et al., 2002; De Geyter & Morent, 2012; Mahmoud, 2016), plasma is a partly ionized mixture of ions, radicals, free electrons and neutral species in the gaseous state. When these particles get excited and ionized, they carry enough energy to induce chemical reactions at the interface with solid surfaces. Researchers say that cold plasma could promote adhesion or anti-adhesion properties in polymers, to improve printability and sealability and to increase the resistance of materials to mechanical failure (Pankaj et al., 2014).

Also, the effects of operating parameters (pressure, power and time) of alternating current (AC) glow discharge plasma were investigated by the same authors to decrease the sensitivity of fish protein films to water (Romani et al., 2019b). In their other study they evaluated physicochemical properties of the films and observed a decrease in water vapor permeability and solubility in some treatments, and concluded that, according to the specificity of the application intended, setting could be changed through the adjustment of parameters of exposure.

6.2 Collagen and gelatin

Collagen can be originated from fish skin, bones, fin and scales, and is used as gelling, stabilizing, foaming and emulsifying agent in food. Properties such as insolubility, biodegradability and fibril forming capacity allow its use in active food packaging material (Bhuimbar, Bhagwat & Dandge, 2019).

The use of collagen is increasing in food, cosmetic, pharmaceutical, tissue engineering and biomedical industries day by day because of its excellent biodegradability and biocompatibility Gómez-Guillén et al., 2011). Collagen can also form insoluble fibers with high tensile strength and stability (Gelse, Pöschl & Aigner, 2003). Meat industry uses collagen films before processing of meat products. On heating, collagen film acts as an edible skin and assists the meat product cooking (Jeevahan et al., 2017).

Gelatin, obtained by the controlled hydrolysis of insoluble fibrous collagen components of skin, bones and connective tissues generated as waste during animal slaughtering and processing (Guo et al., 2014); is presently the most preferable protein derivative as the base material for formulating edible films.

Gelatin films have poor water-vapor barrier and water resistance due to its hydrophilic nature. This could be a disadvantage in gelatin-based films used as food packaging, especially for moist foods (Bhat & Karim, 2014; Etxabide et al., 2017).

Researchers studied collagen films combined with other components, like chitosan (Bhuimbar, Bhagwat & Dandge, 2019); essential oil and montmorillonite (Nakashima, Chevalier & Cortez-Vega, 2016) to investigate their influence in the mechanical properties of the films.

In the study of collagen films added to montmorillonite and essential oil of clove authors noticed that the films obtained had good mechanical properties, adequate visual appearance, easy handling, low permeability to water vapor and low water solubility (Nakashima, Chevalier & Cortez-Vega, 2016).

Other author extracted acid soluble collagen from skin of black ruff, which showed emulsifying activity and emulsion stability (Bhuimbar, Bhagwat & Dandge, 2019). The collagen was used to produce collagen-chitosan films, added to different concentrations of pomegranate peel extract (PPE). It was observed that all films prepared had good mechanical strength. Although chitosan has antibacterial properties, collagen-chitosan blend (control) did not reveal antimicrobial activity due to its low concentration, but the addition of PPE showed some inhibition in pathogens growth.

7. Lipids

Lipids can be divided according to their structure: natural waxes and resins, acetoglycerides, fatty acids, and different types of vegetable oils (Galus & Kadzińska, 2015).

Differently from polysaccharides and proteins, lipids are not biopolymers and do not have the ability to form independent films. Therefore, they can be either used as coatings or incorporated into other biopolymers to make composite films. Waxes, for example, are esters of long-chain aliphatic acids with long-chain aliphatic alcohols. Because of their very low content of polar groups and high content of long-chain fatty alcohols and alkanes, they are more resistant to water migration than most other edible film substances (Cordeiro de Azeredo, 2012).

The origin of waxes is vegetal and animal; they have a function of protective covering tissues (Mohamed, El-Sakhawy & El-Sakhawy (2020). Spotti et al. (2016) mixed beeswax, Brea gum (from *Cercidium praecox*), and glycerol, but they decided that beeswax did not help in this film because of decreased water vapor permeability, microstructure, and mechanical properties. (Kowalczyk, 2016). created films with 5% (w/w) aqueous biopolymer solutions containing 0.5% (w/w) Candelilla wax, 3% (w/w) sorbitol, and 0.35% (w/w) Tween 40 for carrier ascorbic acid. But, waxes are not bad materials for films; for example, Chiumarelli & Hubinger (2014), presented a film with amazing properties, having good mechanical, good barrier, physical, thermal, and structure, and it was composed for cassava starch, carnauba wax, glycerol, and stearic acid.

Investigators used beewax to evaluate the effects of its application as coating on microbiological, physicochemical and sensory properties of Kashar cheese during ripening (Yilmaz & Dagdemir, 2012). The cheese was coated in single and double-layer and was compared to a non-coated sample and a vacuum packaged sample. The results obtained indicated that the coating with beeswax did not affect microbial growth, although thickness of both beeswax has significantly reduced mold growth and extended the shelf-life when compared to control. Also, the coating retarded moisture loss and better maintained the texture of the cheeses.

Studies about extrusion of casein and waxes in order to produce edible films as carrier of potassium sorbate (KS) using different kinds of waxes (beeswax, carnauba and candelilla waxes) and different quantities of KS show antimicrobial activity against *Escherichia coli*, and mechanical and barrier properties (Chevalier et al., 2018). The comparative study showed that several properties are highly depending on wax origin and concentration. Among the waxes studied, only beeswax was efficient to reduce water vapor permeability. The incorporation of waxes improved KS efficiency at low concentration, and this could help the industry on reducing preservative amount in food or food packaging.

The properties of the films from fish gelatin prepared by molecular modification and direct addition of oxidized linoleic acid (OLA), because films from molecularly modified gelatin with hydrophobic substances, such as fatty acids, might not only increase hydrophobicity but also enhance stability. The research showed that molecularly modified gelatin could improve water-vapor barrier of gelatin film more effectively than the one with direct addition of OLA at low levels, and besides, direct addition of OLA had major limitation due to rancid smell in the films, while the molecularly modified gelatin yielded the film with similar appearance to the control gelatin film (Theerawitayaart et al., 2019).

Film from palm fruit oil presented favored water resistance, water vapor barrier, transparency, and elongation. These films can be tried on food (Rodrigues et al., 2016). Vargas, Albors & Chiralt (2011) used sunflower oil in edible coatings and on pork meat hamburgers for enhancing the quality of food, because on meat it was important to oxygen and modulate water vapor to prevent an undesirable reaction.

Resins are substances that plant cells produce in response to injury or infection in trees and shrubs; and some insects can produce them, which is the case of Laccifer lacca that produces shellac resin (Mohamed, El-Sakhawy & El-Sakhawy, 2020). Major of resins are translucent with yellowishbrown tones and physically are solid or semisolid (Baldwin & Hagenmaier, 2011). Chauhan et al. (2015) and Chitravathi, Chauhan & Raju (2014) enhanced edible coatings on a base of shellac; they applied it on green chilies and tomatoes. Those films exhibited quick drying nature, transparency, glossiness, and sound emulsion stability. When applied these coatings on food were optimal, barriers of water vapor and gases could extend shelf life by 12 days and prevented senescence.

8. Other Materials

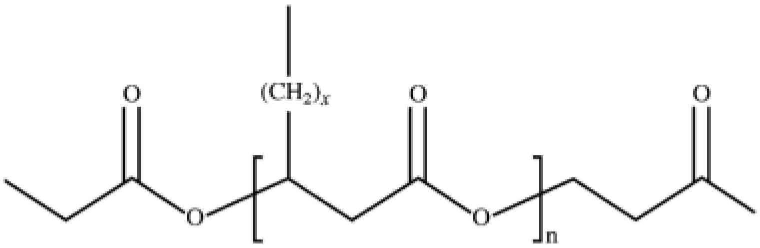
Among the bioplastics produced from renewable sources, are noted for its properties polyhydroxyalkanoates (PHA) and polylactides (PLA). PHA are natural thermoplastics that occur in a wide range of bacteria, while PLA are obtained by the polymerization of lactic acid from fermentation.

8.1 Polyhydroxyalkanoates (PHA)

Polyhydroxyalkanoates (PHA) are the polyesters produced by microorganisms from carbon substrates (Bordes, Pollet & Avérous (2009); Oyama (2009). Its properties and degradability offer potential for non-biodegradable polymers substitution, such as polyethylene and polypropylene (Chen, 2005).

In the Figure 4 is possible verify the chemical structure of the polyhydroxyalkanoates (PHA).

Figure 4 - Chemical structure of polyhydroxyalkanoate.

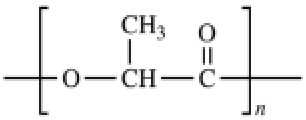


Source: Pachence, Bohrer & Kohn (2007).

8.2 Polylactic acid (PLA)

Polylactic acid (PLA) presents mechanical properties compared to the polymers from fossil sources, specially elasticity, toughness, transparency, thermoplastic behavior, biocompatibility and good moldability (Lim, Auras & Rubino (2008); Liu et al. (2011); Zhang & Sun (2005). PLA is also similar in many ways to polyethylene terephthalate (Oyama, 2009). The Figure 5 present the chemical structure of polylactic acid.

Figure 5 - Chemical structure of polylactic acid.



$$\left[-O-\overset{CH_3}{\overset{|}{CH}}-\overset{O}{\overset{\|}{C}}- \right]_n$$

Souce: Jamshidian et al. (2010).

8.3 Plasticizers

Plasticizers are an important class of compound that have low molecular weight, are non-volatile and are widely used in polymer industries as additives (Sejidov, Mansoori & Goodarzi, 2005).

In most cases, plasticizers are required ingredients for edible films and coatings, especially for polysaccharides and proteins. These film structures can be often fragile and hard due to extensive interactions between the polymer molecules (Krochta, 2002).

The role of plasticizers consists in the elimination of hydrogen bonds and the increase of free volume, leading to higher mobility of the starch chains and decrease of glass transition temperature (Laohakunjit & Noomhorm (2004); Pushpadass, Marx & Hanna (2008). Plasticizers could increase the flexibility of protein films by loosening the protein network. Also, they could affect the water vapor permeability (WVP), because the higher the plasticizer quantity, the higher the WVP (Lieberman & Gilbert (2007); Tanaka et al. (2001).

Polyols, such as glycerol and sorbitol, have been studied for their efficiency in plasticizing hydrophilic polymers (Ghasemlou, Khodaiyan & Oromiehie (2011); Tihminlioglu, Atik & Özen (2010). There are also plasticizers from monosaccharides such as glucose, mannose, fructose, and sucrose (Piermaria et al. (2011); Qiao, Tang & Sun (2011). Several researchers studied how the properties of different films would be affected by plasticizers.

The addition of plasticizers to coating or film have a positive effect, as in the case glycerol-sage seed gum film which increase moisture content and thickness (Razavi, Mohammad & Zahedi, 2015). Study realized by Jouki et al. (2013), demonstrated that the morphology of edible films obtained from watercress gum together with glycerol was homogeneous and without cracking.

A work of edible films based on improved cassava (*Manihot esculenta Crantz*) native starches shows the influence of glycerol in the films. Authors analysed the effect of glycerol, peanut oil and soybean lecithin on the water vapor permeability (WVP). They noticed that WVP, moisture content and thickness of the films increased with higher concentrations of glycerol, and the peanut oil and soybean lecithin did not had significant effect on these aspects (Adjouman et al., 2017).

The same behaviour was also observed in other works. In researches of hybrid sorubim (*Pseudoplatystoma reticulatum* × *Pseudoplatystoma corruscans*) protein films (De Souza Silva et al., 2020); and films based on bocaiuva (*Acromonia aculeata*) flour, which is a native fruit from Brazilian savannah (Oliveira da Silva et al., 2019); authors from both works used glycerol as plasticizer and observed that the increase in the concentration of plasticizer increased the WVP values. It leads to think about the use of additives to improve the characteristics affected by hydrophilic plasticizers.

8.4 Additives

A recent study of a smart package based on chitosan and pomegranate peel extract (PPE) and *Melissa officinalis* essence (MOE) was developed to estimate the expiry date of cream cheese. The PPE contains anthocyanin pigments that are sensitive to pH changes, the color change from blue (alkaline environment) to red (acid environment), which made possible follow up the shelf-life of the cheese. The authors observed that the addition of PPE and MOE increased the antioxidant power of the films, and also the MOE showed antimicrobial activity (Pirsa, 2020).

Antioxidants and antimicrobial agents can also be incorporated into film-forming solutions to achieve active packaging or coating functions (Han, 2002; Han, 2003). Zhu et al. (2020) showed that edible film pieces incorporated with carvacrol or cinnamaldehyde in salad bags effectively reduced *Escherichia coli* O157:H7 on leafy greens that were previously contaminated. The use of edible films containing natural antimicrobials could meet the increased consumer demand for both natural and safe salads.

8.5 Essential oils

Essential oils are rich in hydrophobic and volatile materials. They have an important antimicrobial activity owing to its ingredients from terpenoids, terpenes, and other aromatic compounds (Mohamed, El-Sakhawy & El-Sakhawy, 2020). Randazzo et al. (2016) and Liu et al. (2020) tested various essential oils in films to estimate the antimicrobial effect and properties of the matrix. Essential oils (EOs) active ingredients can diffuse from the film into the coated food to control target microorganisms (Valencia-Sullca et al., 2018). Due to their lipidic nature, they can help reduce the water vapor permeability of hydrophilic films (Atarés & Chiralt, 2016).

According Seydim & Sarikus (2006), the directly addition of essential oils in food can reduce microbial population in it, but it could change its sensorial characteristics. Then, the addition of essential oils in films and coatings could be more of interest in the conservation of food. They studied the antimicrobial activity of oregano, rosemary and garlic essential oils in whey protein films.

Other authors studied the effects of other essential oils Chatli & Mehndiratta, 2018; Singh, Chatli & Mehndiratta, 2018). In a study about the antimicrobial effects of cinnamaldehyde, lemongrass oil, clove oil, peppermint oil in starch films, authors found that the optimum level of incorporation of essential oils is 0.5% in starch edible films. The addition increased thickness of films and decreased the moisture content and percent solubility in water. The lemon grass and clove essential oil showed best results in microorganism inhibition (Chatli & Mehndiratta, 2018). In another study, using essential oils from medicinal plants (*R. officinalis L, A. herba alba Asso, O. basilicum L. and M. pulegium L.*) it was also possible to obtain good results in antimicrobial effect. Besides that, due to the dispersion of the EOs in the matrix barrier properties were improved (Mahcene et al., 2020). Randazzo et al. (2016) used citrus essential oils of peels from lemon, mandarin, and orange on methylcellulose or chitosan films, and these essential oils presented better incorporation with chitosan film. This information shows the potential of the use of EOs as coatings not only in the antimicrobial effect but also as a hydrophobic component.

8.6 Antioxidants

Antioxidants in films have the function of protecting food products from the oxidative degradation, avoiding reactions of oxidation by reacting to free radicals and peroxides, extending the products' shelf life. In the context of active and intelligent packaging, the incorporation of antioxidants or antimicrobials to packaging materials shows utility to extend the shelf-life and improve food safety or sensory properties (Valdés et al., 2014).

A study of the addition of antioxidants (α-Tocopherol and ascorbyl palmitate) in whey protein films applied in peanuts (Han et al., 2008); showed results retarding the oxidation of fatty acids presents on peanuts. The coated peanuts oxidized slower than uncoated peanuts, which means that the whey protein coating was a good oxygen barrier and reduced the oxygen penetration rate resulting in the retardation of oxidation of peanuts. However, the author observed no significant difference of oxidation retardation between the coated peanuts with and without antioxidants.

Also, the extraction of antioxidants from sunflower hulls, which are an abundant by-product from food industry, when applied in starch films authors noticed promising results (Menzel et al., 2019). It was observed that 1-2% of extracts were enough to produce starch films with high antioxidant capacity. Higher amounts (4-6%) of extract showed the highest antioxidant activity, and showed the lowest oxygen permeability, high stiffness and poor extensibility.

There are recent studies about the addition of fruit juice in polysaccharides films (Chambi et al., 2020). The authors used juice from jambolan (*Syzygium cumini L.*) and grapes (*Vitis vinifera L.*), both contain phenolic compounds. The authors produced films with an attractive color and observed that the films showed antioxidant capacity, which indicates that fruit juices could be potential ingredients to produce antioxidant edible films.

9. Final Considerations

With that we can see that the use of biodegradable materials, such as polysaccharides and proteins are promising in the development of edible films and coatings, especially when incorporated to fibers, lipids, plasticizers, antimicrobials, antioxidants and other components that could improve their characteristics and properties.

Its application in food products, they can enhance food characteristics and extend their shelf-life, and because they are produced from renewable sources and biodegradable, these materials can be substituted for plastics and thereby greatly reduce the damage caused to the environment.

It is recommended to develop new studies that contribute to the theme emphasized in the various methods that exist for obtaining low-cost edible films and coatings to food.

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