Cultivos Tropicales, 2020, vol. 41, no. 3, e09
ISSN impreso: 0258-5936
ISSN digital: 1819-4087

julio-septiembre
Ministerio de Educación Superior. Cuba
Instituto Nacional de Ciencias Agrícolas

http://ediciones.inca.edu.cu

Bibliographic review

Polysaccharide-based films as biodegradable coatings in fruits postharvest

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ABSTRACT

The postharvest preservation of fruits and vegetables is a vital issue due to the large losses generated during its handling in the domestic market and exportation. Microbiological and physiological deterioration, inadequate collection process, the use of inappropriate packaging and insufficient transportation routes are key factors for these losses. The use of biodegradable films and coatings is an alternative to decrease these losses. Biodegradable films and coatings are generally either composed of carbohydrates proteins and lipids individually or combined. These coatings constitute a semipermeable barrier to water vapor, oxygen (O_2) and carbon dioxide (CO_2) between fruits and the surrounding environment. In general, they can slow down the senescence process and microbial growth on the surface of the fruits, which allows quality preservation and facilitates their distribution and commercialization. Due to the potential and importance that this type of technology currently presents in our national context, this review was carried out taking into account the main components used in the formation of these materials, the practical application in fruits and vegetables, and the principal characterization procedures of these biodegradable films and coatings.

Key words: chitosan, pectin, starch, proteins, lipids

INTRODUCTION

The conservation of the fruits in the postharvest is a matter of vital importance for the commercialization and distribution of the same. Post-harvest losses of fruit and vegetable products rise from 5 to 25 % in developed countries, and from 20 to 50 % in developing countries [(1)]. Generally, these losses are due to microbiological and physiological deterioration, to an inadequate collection process, to the use of inadequate packaging and insufficient transportation routes. To mitigate these problems, a group of alternatives have been developed such as storage at low temperatures [(2)], the application of gamma and ultraviolet radiation [(3)], the application of biological controls [(4)], the use of controlled atmospheres and plastic packaging [(5)], the use of aminoethoxyvinylglycine [(5)] and 1-methyl chloropropene to delay fruit ripening [(2)]. Also, the use of biodegradable films and coatings [(6)], among others.

In particular, the development of biodegradable films and coatings for fresh and processed food products has gained momentum in recent times. This is because this technology is environmentally friendly and can improve the quality of food, its biosecurity and stability. These characteristics are achieved by providing a semipermeable barrier to water vapor, dioxygen (O_2) and carbon dioxide (CO_2) between the fruits and the surrounding atmosphere. This technology serves as a protective barrier by reducing metabolic processes such as respiration, delaying the senescence process and microbial growth on the surface of the fruit, which allows preserving quality and facilitating the distribution and marketing of these [(7).]

Biodegradable coatings must be toxic free and safe for health, they must require simple technology for their elaboration and protect the fruits from any physical, chemical or mechanical action. At the same time, they must be transparent and cannot be detected during consumption [(8)]. Polysaccharides are the main components present in edible films and coatings. Among them, we can mention high and low methoxyl pectins [(9)], chitosan [(10)], cellulose and its derivatives [(11)], algae extracts (alginate [(12)], carrageenan, agar [(13)] and gum arabic [(14)], among others. Other compounds used are lipids such as beeswax [(15)], waxes extracted from candelilla plants [(16)] and laurel [(17)], as well as proteins, both from animal sources (casein [(18)], gelatin [(19)] as vegetables (wheat gluten [(20)], zein [(21)], soy, peanuts and rice [(12)]), among others.

These compounds can be part of the films individually or combined with each other. Furthermore, they can be linked to additives that improve the antioxidant, antimicrobial and mechanical properties of coatings to extend their shelf life and reduce the risk of pathogen growth on the food surface [(22)]. Taking into account that these films have proven to be effective in terms of food safety and environmental contamination, they could be a viable alternative in the industrial sector. Cuba is a country rich in fruit and vegetable products that are destined for both domestic consumption and export, so having technologies that allow them to extend their useful life is vitally important. For this reason, the objective of this work is to approach the art state on biodegradable films research based on polysaccharides. They are used as a post-harvest coating, their main sources of production, characteristics, analyzes that are carried out and the main fruit and vegetable products in which they are used, in order to develop a line for obtaining biodegradable coatings from Cuban raw materials.

Edible films and coatings

A biodegradable coating can be defined as a continuous, thin, transparent matrix that is created around a food, generally by immersing it in a coating-forming solution, in order to preserve its quality and also serve as a packaging (8). A biodegradable film is in the form of a solid sheet and is applied by wrapping the product in it (23). Active biodegradable coatings maintain quality and extend the shelf life of fresh fruits and vegetables, as well as prevent microbial spoilage (10). The deterioration in the fruits is mainly caused because they are biologically active and transpiration (loss of humidity), respiration (gas exchange), maturation and other biochemical processes occur in them (24). Biodegradable coatings reduce moisture and solute migration. Its application offers a plausible solution to obtain fresh, nutritious and edible products. They serve as semi-permeable barriers to gases and water vapor, reducing respiration and water loss. Furthermore, they delay deterioration and senescence in a similar way to modified atmospheres (25).

Biodegradable coatings must meet certain functional requirements that allow them to control or lessen the causes of alteration in the foods to be coated (Figure 1) (26–31). However, when coating a fruit or vegetable to delay moisture loss, it is necessary there be a certain permeability to O_2 and CO_2 to avoid anaerobic respiration that could induce physiological disorders and a rapid loss of quality and shelf life of themselves (23).

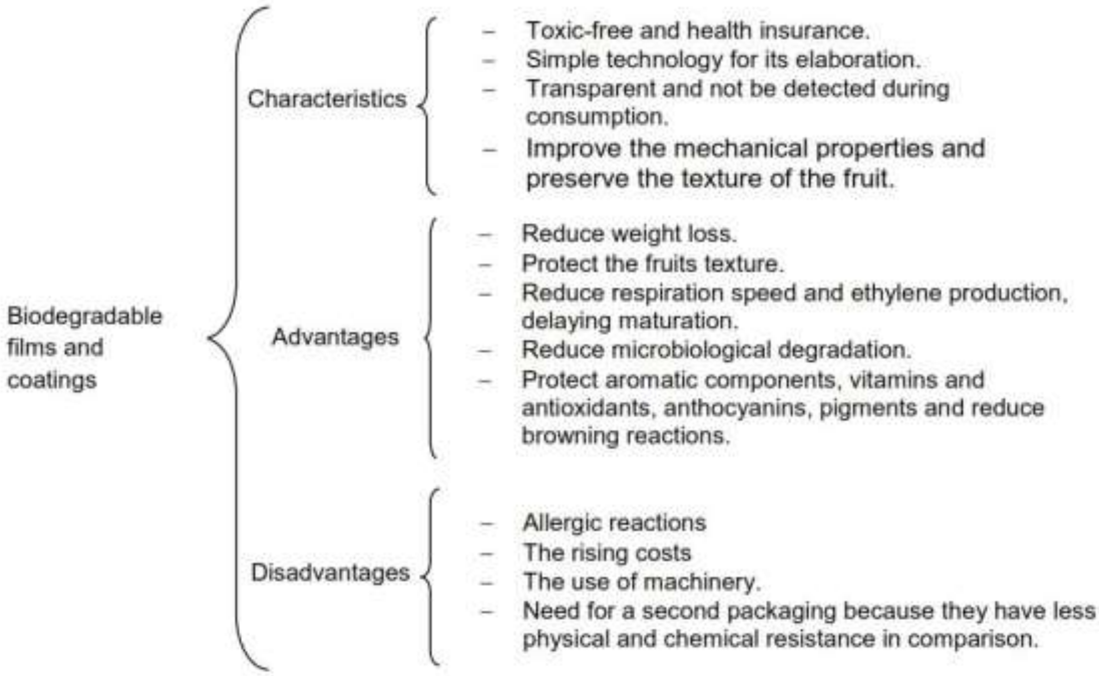

Figure 1. Characteristics, advantages and disadvantages of coatings and biodegradable films

Main components of biodegradable coatings and films

The main components of biodegradable films and coatings are carbohydrates (sugars and polysaccharides), proteins and lipids. Polysaccharides are macromolecules made up of monosaccharides linked by glycosidic bonds and their hydrolysis produces a large number of molecules of one or more simple sugars (monosaccharides) or their derivatives [(32)].

Polysaccharides and proteins are polymers that form cohesive molecular networks due to a high interaction between their molecules, which gives them good mechanical properties and a barrier to O_2 and CO_2 gases, thus slowing down the respiration and aging of many fruits and vegetables [(7)]. These compounds have been widely used in food processing, nanotechnology, medicine, biotechnology, and agriculture [(33)]. Its use in biodegradable coatings has been extensively documented as shown in Table 1. These compounds give the films and coatings hardness, sharpness, compactness, viscosity and adhesiveness [(13)]. The sources of obtaining the polysaccharides are very varied; they can be of plant origin (pectins, cellulose, starch, alginate, carrageenan, among others) and animal (chitin) [(12)].

Table 1. Clasificación de los materiales que forman las películas o recubrimientos biodegradables

Carbohydrates	Protein	Lipids
Animal source: Chitosan [10]. Vegetable source: High and low methoxyl pectins [9]. Cellulose and its derivatives [11]. Starch [18] Gum arabic [14]. Algae extracts (alginate [34], carrageenan [35], and agar [13].	Animal sources: casein [18], gelatin [19]. Plant sources: zein (corn [21]) soy, gluten (wheat) [20], peanuts, rice [13]	Animal Source: Beeswax [15] Vegetable sources: Candelilla wax [17] Laurel wax [17]. Carnauba wax [36] Resins and fatty acids: stearic acid [37], palmitic, lauric, oleic [38] and coconut butter [39]

Chitosan

Chitosan is one of the most widely used polysaccharides in obtaining biodegradable coatings. Its origin is from chitin, the second most abundant polysaccharide in nature after cellulose. It is a semi-synthetic polymer that is obtained by deacetylation of chitin, which is mainly found in the exoskeleton of crustaceans, although it is also found in a smaller proportion in the wings of some insects, and in the cell wall of fungi, algae and others [10]. From a chemical point of view, it is a copolymer composed of units of *N*-glucosamine (D-GlcN), N-Acetyl glucosamine (D-GlcNAc) randomly distributed, and linked by β-(1-4) glycosidic bonds in a structure rigid unbranched. Chitosan is presented as a potential material to obtain biodegradable coatings, mainly due to its capacity to form films, its non-toxic nature, its antimicrobial activity [40], as well as its biodegrabability and biocompatibility. This polymer is recognized to have gas barrier properties, thus ensuring decreased senescence by preventing respiration and perspiration [10]. Chitosan is generally dissolved in acetic acid, although lactic acid has also been used for these purposes [41].

The incorporation of an additive such as calcium gluconate to the chitosan-based coating increases the nutritional value of the fruit, due to an increase in its calcium content [38]. Calcium plays an essential role in the structural maintenance of the membranes and the cell wall. The crosslinking of calcium with free carboxyl groups in adjacent polygalacturonate chains present in the mid lamella of the plant cell wall contributes to adhesion and cohesion between cells [42]. That is why a treatment with calcium can increase the stability of the cell wall and improve resistance to the enzymes secreted by phytopathogenic fungi, while the coating with chitosan reduces respiration, delaying the maturation process and the progressive deterioration of the product fruits of senescence.

Pectin

Another polysaccharide used in edible coatings is pectin. Pectins are located mainly in the middle lamella of the primary cell wall of plants, where they contribute to the adhesion between the cells of the plant parenchyma and the strength of their tissues (43). It is mainly made up of two regions, a uronic region; Extensive and regular, made up of galacturonic acid units linked by α-(1,4)-type bonds, which may be partially methylated, and the second composed mainly of rhamnose that carry neutral sugar side chains (44).

Pectins are used extensively in processed foods because of their gelling properties (45). Under certain conditions, they are capable of forming gels, a characteristic for which they are considered an important additive in hams, jellies, jams, as well as in the clothing industry. Agricultural sources of pectin are currently underutilized, even though pectin is a potentially important food ingredient available and abundant in agricultural waste. Precisely, its pharmaceutical application and the additional nutritional benefit in a wide variety of food products increase the interest in this polysaccharide (39).

Studies on pectin films date mostly from the 1930 to 1950 (46). These studies focused on derivatized pectins and the use of polyvalent cations such as calcium. The study of films from low methoxyl pectins (<11 %) shows that the tensile strength decreases with the increase of the methoxyl groups (46). The physical characteristics of the gel are the consequence of the formation of a three-dimensional network or of the cross-linking between the polymer molecules (47). Low methoxyl pectins, derived by controlled deesterification, form gels in the presence of calcium ions and can be used to develop biodegradable films. Pectin is highly hydrophilic, since it is composed of at least 17 types of monosaccharides, among which D-galacturonic acid is the most abundant, followed by D-galactose or L-arabinose, and covalently linked to each other (48). Hence, the water vapor permeability (PVA) of pectin films is quite high, in the same order of magnitude as that of cellophane and other carbohydrate-based films (51). Thermomechanical, microstructural and viscoelastic properties of pectin-based films, such as the mixture of pectin and starch, concluding that they are highly resistant (49)

Starch

Starch is another of the carbohydrates used in the development of biodegradable coatings individually or in combination; it is widely used due to its abundance and low cost. Starch is a reserve polysaccharide made from amylose and amylopectin. Starch-based coatings are transparent, colorless, and odorless and have low oxygen permeability (50). Starch has also been combined with carrageenan to form highly resistant biodegradable films (51).

Chitosan and pectin based coatings

The antimicrobial character in biodegradable coatings is a highly desired attribute. In the case of chitosan, antimicrobial activity has been related to the ability of this polymer to cause severe damage at the cellular level to the mycelium of fungi treated with this polymer (52). However, it has been enhanced when combined with the lactoperoxidase enzyme system in the preservation of mango (41). Lactoperoxidase is an excellent system to combat pathogenic microorganisms and has a broad antimicrobial spectrum. The combination of 1 % chitosan with the lactoperoxidase enzyme system is effective against microbial contamination and allows delaying fruit ripening without altering its quality (41). Zinc II and Cerium IV complex with chitosan have also been used to preserve Chinese jujube fruit, extend the shelf life of the fruit and reduce residues of organophosphate pesticides. Zinc is one of the most important essential micrometallic elements in the human body. It is an essential component in a significant number of proteins and is essential for the stability of catalytic functions. Cerium ions have good antibiotic capacity (53). In turn, chitosan has been combined with lemongrass essential oils to preserve pepper against anthracnose (54), and thyme, cinnamon and clove to preserve papaya against *P. digitatum* and *C fungi*. gloesosporioides where it promotes greater mycelial inhibition in *in vitro* studies at a chitosan concentration of 0.5 and 1 % (38).

In the case of pectin-based coatings, essential oils of cinnamon (15), lemon, orange (9) and cinnamaldehyde nanoemulsions (55) have been incorporated, among others. Another example is the modification of the surface of polypropylene (PP) films by multilayers of chitosan/pectin. In these films, chitosan forms a polyelectrolyte complex with pectin, which enables the formation of a stable multilayer structure on the surface of the PP film, with the consequent formation of much better antimicrobial films that can be used to manufacture excellent packaging materials for post-harvest crop protection (56).

In general, polysaccharide-based coatings such as chitosan and pectin constitute a poor moisture barrier due to the hydrophilicity they present. For this reason, many works have been aimed at combining these polymers with lipids, resins and fatty acids (Table 1), with the aim of regulating the hydrophilic-lipophilic balance (39). Lipids are characterized by being hydrophobic, have excellent moisture barrier properties, although their lack of cohesiveness and structural integrity result in poor mechanical properties, leading to the formation of brittle coatings. Lipids reduce perspiration, dehydration, abrasion in subsequent handling, and can improve the gloss and appearance of many foods (8).

Other additives: proteins and plasticizers

Proteins are another component used in biodegradable coatings. Among the proteins used for the production of these, casein, zein and gelatin stand out among others. Casein has gained prominence in the development of coatings because it is commercially available, has the ability to act as an emulsifier, is water soluble [(57)] and produces transparent and thermally stable coatings [(18)]. The combination of casein and starch together with glycerol and barbatimón extract has been used in the preservation of guava. This coating lengthens the shelf life of guavas, reduces weight loss and firmness [(18)].

Another example is the formation of biodegradable films from chitosan-bound quinoa protein, forming resistant films without the use of plasticizers, the addition of sunflower essential oil, improves water vapor permeability as a result of interactions hydrophobic [(58)]. Zein is a storage protein found in corn kernels. This protein has the characteristic of preventing oxidation and the development of bad odors due to its excellent gas barrier properties against oxygen and carbon dioxide [(59)]. Gelatin films have effective properties as a gas barrier against oxygen and carbon dioxide [(60)].

Plasticizers are non-volatile, high-boiling point substances that, when added to another material, change the physical or mechanical properties of this material [(39)]. Polyols like sorbitol and glycerol effectively plasticize for their ability to reduce internal hydrogen bonding while increasing intermolecular spacing. These plasticizers decrease intermolecular forces along the polymer chain, increasing the flexibility of the film while decreasing its barrier properties [(61)]. Plasticizers have also been used in obtaining biodegradable coatings, such as sorbitol [(61)], glycerol [(62)], polyethylene glycol [(37)] and papaya puree [(55)].

The proportion in which the components are found in the coatings is important. A study on the influence of the amount of pectin and plasticizer on the mechanical properties of the fruit revealed that an increase pectin for and a decrease in the amount of plasticizer increases the tensile strength and modulus of elasticity, while the elongation at break increases with increasing concentration of both [(61)]. Similarly, an increase in the concentration of pectin and plasticizer affects PVA, due to the hydrophilic nature of both compounds. However, the combination with a lipid like beeswax lowers PVA. At low concentrations, the added lipid increases hydrophobicity, thus decreasing the permeability of the film. At higher concentrations, lipids could result in larger globules during the drying stage in obtaining the film, contribute to the interruption of the continuous structure of the films, and consequently to a higher PVA [(63)]. The studies of coatings formed from methylcellulose, stearic acid and polyethylene glycol carried out by an emulsion method, reveal that the increase in the volume of stearic acid up to 22 % causes a decrease in PVA [(37)]. However, the increase in the volume of stearic acid above

22 % leads to an increase in PVA, attributed to inadequate filling of the empty volume inside the stearic acid crystals by the methylcellulose matrix, which favors migration of moisture. On the other hand, the increase in PVA in pectin and sorbitol films at high concentrations of both may perhaps be due to a greater heterogeneity because of the distribution of lipid globules (number of populations) within the pectin matrix [61].

Practical applications of biodegradable coatings and films

Fruits and vegetables are an invaluable source of vitamins and minerals in the daily diet of humans. An adequate conservation of these, allows extending the shelf life in the post-harvest. Biodegradable coatings and films have been used with great success in the preservation of guava (*Psidium guajava* L.) [64], papaya (*Carica papaya* L.) [65], mango (*Mangifera indica* L.) [66], mandarin orange (*Citrus reticula* L.) [67], strawberry (*Fragaria ananassa* Duch) [58], tomato (*Lycopersicon esculentum* Mill.) [68], bell pepper (*Capsicum annuum* L.) [54] and the cucumber (*Cucumis sativus* L.) [27], among others.

The practical application of coatings in most cases reduces weight loss and ripening rate, and increases the shelf life of the fruits (Table 2). In the ripening process during storage, a series of physical and biochemical changes occur that vary depending on the type of fruit [9]. Among the changes, we can find the modification of the color through the alteration in the content of chlorophylls, carotenoids and the accumulation of flavonoids. In turn, the modification of the product texture to the alteration of the cell turgor and the structure of the cell wall by metabolism. As well as, the modification of carbohydrates, organic acids and volatile compounds, which affect the nutritional quality, flavor and aroma of the fruit. Last but not least, the increased susceptibility to attack by opportunistic pathogens that are associated with loss of cell wall integrity [24].

It is recognized that during the maturation process the total soluble solids (TSS) are increased due to the respiration process [9], during which the degradation of carbohydrates and organic acids (ascorbic, citric and malic) occurs due to the use of these as a substrate in the breathing process [54]. The increase in the TSS content and the reduction in the measurable acidity are indicators of the maturation process [69].

In the case of tomato, pectin-based coatings increase the retention of ascorbic acid, measurable acidity and total sugars during storage compared to the control [65]. These results are positive since they indicate a delay in maturation due to the action of the coating, since they limit oxygen permeability, reducing the oxidation of ascorbic acid, and limiting the use of citric acid as a substrate in the respiration process.

The chitosan-salicylic acid combination has been studied in the conservation of cucumber at low temperatures of 2 ºC [27]. These study shows that the combination reduces the damage caused by the low temperatures on the surface of the cucumber and that the combined action exceeds that of the components individually. Chitosan-salicylic acid also reduces weight loss and respiration rate, causes increases in malondialdehyde content and electrolyte leakage, and maintains a higher content of TSS, chlorophyll, and ascorbic acid.

Table 2. Biodegradable coatings and films applied to the fruits

Application	Composition of biodegradable films and coatings.	Function	Reference
Guava	Chitosan 1 %, acetic acid	Extension of the useful life of the fruit.	[64]
	Casein, starch, glycerol and extract of barbatimón	Reduces weight loss and firmness. Delays the maturation process by inhibiting respiration rates.	[18]
Papaya	Chitosan 2 %, Acetic Acid 0.5 %, Tween 80.		[70]
Tomato	Chitosan 0,25 %		[68]
	Pectin 3 %, water		[65]
	Chitosan 1 %, zeolite (Ø=35-45 µm), lactic acid, Tween 80	Delays fruit ripening, but is a poor barrier to weight loss	[71]
Mango	Acetato de quitosana 20 %	Reduction of the incidence of anthracnose. Delays the maturation process.	[66]
Cucumber	Chitosan Acetate 20 %		[72]
	Highly methoxylated pectin 2 %, beeswax, sorbitol and water	Extension of the useful life of the fruit, reduction of respiration rates. Reduces weight loss and firmness	[27]
Mandarine	Pectin 8 % and water	Extension of the useful life of the fruit. Reduces weight loss and firmness	[67]
Pepper	Chitosan 1 %, NaOH 1M, Lemongrass Essential Oil 0.5 %, Tween 80	Reducción de la incidencia de antracnosis. Retrasa el proceso de maduración.	[54]
Strawberry	Pectin 1 %, glycerol 0.02 %, lemon essential oil 1 and 0.5 %	Extension of the useful life of the fruit. Reduces weight loss and firmness	[9]
	Quinoa protein 0.1 % -quitosan 2 % in citric acid-sunflower essential oil 2.9 %	Extension of the useful life of the fruit. Reduces weight loss and firmness. Reduces mold and yeast growth	[58]

Both zein and gelatin have been successfully tested in post-harvest preservation of mango [59]. Both proteins combined with glycerol positively affect weight loss, TSS, titratable acidity, pH, total sugar content, and carotenoid content. Additionally, they maintain the content of total phenols and ascorbic acid at high levels compared to the control [59].

When carrying out practical applications of the coatings, it is necessary to take into account the concentration of the carbohydrate or the component in question. A chitosan concentration of 1.5 % inhibits the growth of the fungi during the storage period in the preservation of the pumpkin fruit, while the increase above 2 % leads to inadequate maturation [(70).]

It concludes, the application of biodegradable coatings to fruits and fruit and vegetable products is extremely important because they extend shelf life, allowing for better transportation and marketing of these. This type of technology in our country is highly attractive for the preservation of exportable fruits such as pineapple, mango and avocado, as well as for the internal marketing of the wide variety of fruits and vegetables that are produced in our domestic market and for supplying our tourist facilities.

Main physico-chemical characteristics to be evaluated in edible films and coatings

Among the physical characteristics to be evaluated in the coatings are the thickness of the films from the measurement with a micrometer [(16)]. The determination of the viscosity allows studying the flow properties at different concentrations of the polymers. Generally, for this analysis, a rheometer is used to measure the viscosity and the viscoelastic properties of solutions at different concentrations of the polymers [(61)]. Film surface studies are performed by scanning electron microscopy [(73)] or atomic force [(74)], and Fourier transform infrared spectroscopy (FTIR) allows identifying structural changes made in the polymers from the changes in their characteristic absorption bands [(10)]. These techniques allow for a structural analysis of the film surface that provides information on the composition of the film.

Another analysis to be performed is the permeability to water vapor. This analysis is extremely important in edible coatings because a low permeability slows down the loss of weight in the fruit. Gravimetric methods have been developed for this, among which is the ASTM E96-93 [(75)] standard. This procedure is based on the prior drying of the membrane or film in a waterproof container and the subsequent contact of this container with magnesium nitrate to maintain the relative humidity of the films at 53 %. Later they are stored in desiccators maintaining humidity relative (RH) in 0 % with the help of a drying agent such as calcium chloride at 25 ºC. The water vapor transmission rate divided by the difference in vapor pressure across the film constitutes the permeability of the film [(61)]. The permeability multiplied by the thickness of the film is the transmission to water vapor [(75)].

Another procedure is ASTM E96/E96M-10 [(76)] with some modifications, where the 3 cm diameter membranes, previously stored for at least 72 hours in a desiccator, are placed in containers containing silica gel inside and they are hermetically sealed, taking care that only the membrane is exposed to water vapor. The containers in a desiccator are weighed and placed containing a saturated solution of KCl. It provides a constant water activity equal to 0.8434 at 25 ºC. Subsequently, the desiccator is in a chamber at constant temperature placed. The containers are on an analytical balance every 24 hours removed and weighed. The calculation of PVA is made from the slope of the graph of the variation in mass over time using (Equation 1). The experiment is considered concluded when no increase in the weight of the containers is observed.

$$PVA = \frac{\Delta m}{\Delta t} xA \qquad (1)$$

Where $\frac{\Delta m}{\Delta t}$ is the slope of the weight graph over time and A is the effective area of the membrane in contact with the water vapors (77). Tensile strength (τ) is one of the mechanical properties that are performed to verify the resistance of the films. It is carried out in a texture analyzer (16). In the analysis the samples are stretched to rupture, the resulting force is divided by the cross section of the film, and τ is expressed in MPa. Furthermore, the stretching capacity to rupture (CE) is calculated in percentage, with (Equation 2), where Li and Lf are the initial and final lengths of the films, expressed in millimeters (mm) (16).

$$CE = \left(\frac{L_f - L_i}{L_i}\right) * 100 \qquad (2)$$

CONCLUSIONS

- Biodegradable coatings are of great importance in the post-harvest handling of fruit and vegetable products. Generally, for its formation, the use of polymers from various natural sources such as pectin, chitosan, starch, cellulose derivatives, among others, combined with lipids, emulsifiers and plasticizers is used.
- The great variety of polysaccharides obtained from various plant and animal sources, as well as the combination of these with other additives that allow improving the characteristics of these coatings, extending the possibility of infinite combinations of these compounds, with a view to achieving the best results.
- The use of biodegradable coatings contributes to increasing the shelf life of fruit and vegetable products, which constitutes a useful and promising technology in the conservation of fruits in the post-harvest of our country.

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