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Current development of edible food packaging: A review

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Abstract

A desirable solution to prevent landfill wastes generated by non-biodegradable polymers used in food packaging can be obtained through edible food packaging. Edible biopolymers draw attention due to their biocompatible, comestible, and degradable properties. Their composition can be conformed by proteins, polysaccharides, microbial polymers and their composites. Their main advantages are their ingestible non-toxic nature, as they form part of the food product while offering shell life extension, improvements in gas/moisture barrier, mechanical properties, and even antibacterial properties. Additional components like antioxidant agents and plasticizers, enhance the previously mentioned properties and open the way to new applications. Concepts, characteristics, and properties concerning research in food's edible films are defined in the present work in order to give a broad perspective of this emerging and promising field.

Keywords: *Biopolymer, Edible films, Degradation, Food packaging, Intelligent, Sustainable*

1. Introduction

The usage of conventional plastics derived from fossil sources for food packaging is causing serious environmental problems because their final disposal generates accumulation in sanitary landfills due to their characteristic of non-biodegradability and also its questionable recycling process (Soroudi and Jakubowics, 2013; Teixeira, 2012; and Privas, 2013). The problem is being addressed by three of the 17 sustainable goals declared by the United Nations (UN): responsible consumption and production, climate action, and life below water. Due to the relevance and the increasing behavior of this problem, new materials are required by the packaging industry, a market that is tending to grow in the next few years.

Nowadays, the packaging industry requires an increase of new polymeric materials, it is expected for the food packaging industry to grow in the next few years. This can be accomplished by the development of biocomposite materials, edible or degradable. Also, nanotechnology plays an important role, by this, the

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properties of packaging could be enhanced. In 2015, plastics derived from petroleum suppressed 300 million tons (Mehdi *et al*., 2017).

One of the main objectives involved in the development of new packaging is that it should meet environmental standards. For this, natural polymers are used as the primary material to meet certain characteristics since they are biocompatible, edible, biodegradable and can be used for barrier purposes (Padrão, 2016). Biopolymers are mainly divided by two conditions, those that come from renewable resources and others that are degradable (Valero *et al*., 2013), however, both properties can be present. Due to this, classification of bioplastics fits into three main categories, including:

a) Renewable resource-based bioplastics

b) Petroleum-based bioplastics

c) Bioplastics from mixed sources

Biodegradable processes involve three main phases, firstly, biodeterioration correlated to the growth of microorganisms causing variations in polymer properties. Second, biofragmentation involves the breaking down of polymers bonding due to such microorganisms. On the last step of the process, microorganisms take necessary carbon, energy and nutrients transforming polymer´s fragments into CO_2, water, and biomass (Mehdi, 2017). Due to this, biodegradable polymers do not require recycling since they can be left out in the environment to biodegrade (Shahadat *et al*., 2016). A wide interest in biopolymers has emerged due to their capacity for film formation specifically as edible films in the food industry (Pérez *et al*., 2013).

In the following work, we review different kinds of biopolymers (protein-based, polysaccharides, composites, and microbial polymers) that have edible packaging potential. The characteristics of current edible packaging, together with their mechanisms of obtention are presented as well, so that their mechanical, degradation, and permeability properties can be assessed. Finally, current applications of edible biopolymers packaging are described.

2. Biopolymers

Natural polymers are more suitable materials to produce food packaging than synthetic polymers since it is desired to use biodegradable materials (Siracusa *et al*., 2008). The concept of biopolymer-based packaging refers to the components of the package having to come from agricultural and marine sources (Mohan *et al*., 2019). According to the European Bioplastics, biopolymers made with renewable resources must be biodegradable and especially compostable, so they can be used as fertilizers and soil conditioners. It needs to define that plastics based on renewable resources do not necessarily have to be biodegradable or compostable, and bioplastics do not necessarily have to come from renewable materials because the biodegradability is part of the chemical structure properties (Siracusa *et al*., 2008). There are three categories of biopolymers used: a) extracted from biomaterials, such as starch, cellulose, proteins, and marine prokaryotes; b) produced by chemical synthesis of biomaterial-derived monomers; c) produced by microorganisms such as polyhydroxy butyric acid (PHB), hydroxyisobutyrate (HB) and hydroxyvalerate copolymers (HV). However, the practical applications of biopolymers are limited by their production technology as well as high production costs (Kokoszka and Lenart, 2007). Therefore, the use of biopolymers materials such as proteins, polysaccharides, or composite materials has emerged as an option with low production costs that is easily scalable globally.

2.1. Protein-based material

In their natural states, proteins exist as fibrous proteins or globular proteins. The first type is water-insoluble and is used as the main structural materials of animal tissues; globular proteins are soluble in water or aqueous solutions of acid bases or salts and function widely in living systems. Structurally, fibrous proteins extend and associate between parallel structures through hydrogen bonds forming fibers, on the other hand, globular proteins fold into spherical structures by combinations of hydrogen, ionic, hydrophobic, and disulfide bonds (covalent). In both cases, chemical and physical properties depend on the amounts of amino acid residues and their placement along the protein-polymer chain (Hassam, 2018). Generally, proteins must be denatured by heat, acids, bases, and / or solvents to produce its more extended structures that are required for film formation. The formation of the films is through solutions or dispersions of the protein as the solver / carrier evaporates; commonly used solvents are water, ethanol, or ethanol-water mixtures (Wittaya, 2012).

Protein-based edible films can be used for different food products to reduce moisture, absorption of oxygen, migration of lipids, to improve mechanical handling properties, to provide physical protection, or to offer an alternative material to synthetic packaging materials (Mohan *et al*., 2019). The addition of polar groups along the protein chain increases the possibilities of interaction between the chain, which determines the resistance of the edible film; stronger interactions produce stronger films, but less permeable to vapors, liquids, and gases. Proteins can be assembled or selected based on the physical barrier or barrier requirements (Hassam, 2018). Therefore, the protein-based edible film offers excellent mechanical and barrier properties against oils, oxygen, and aroma (Mohan *et al*., 2019). Some proteins that have been used to produce healthy to eat films / coatings because of their film properties are shown in Figure 1.

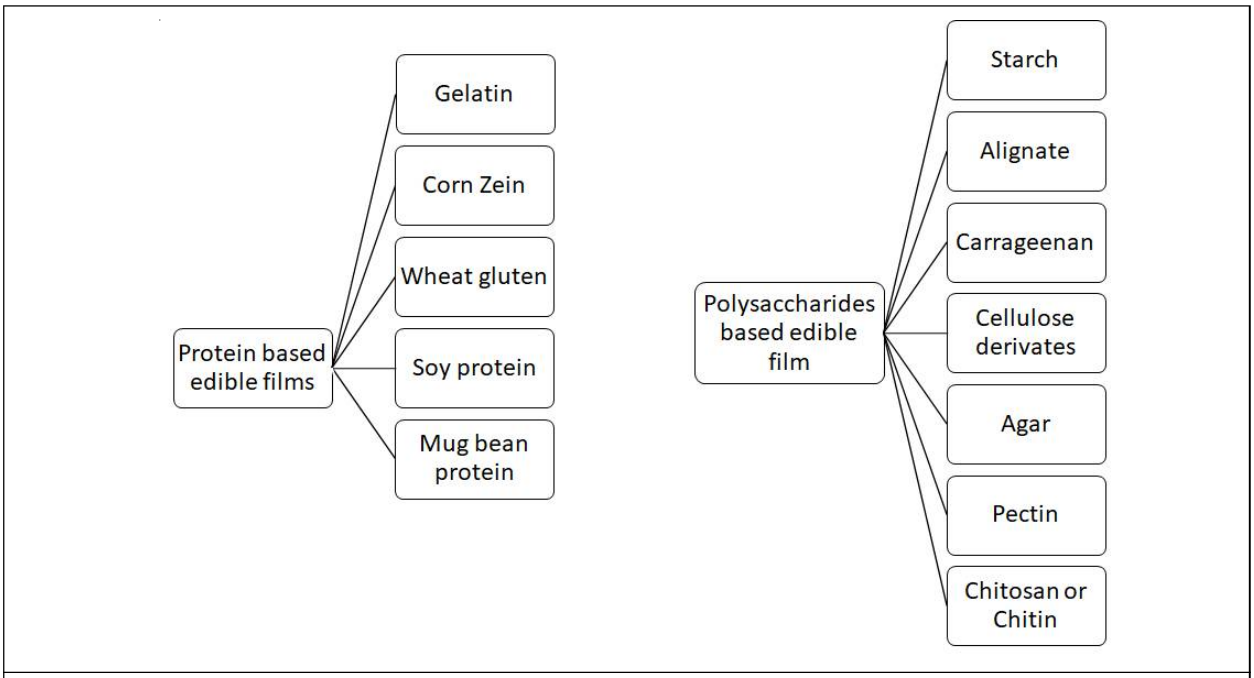


Figure 1: Classification of edible food packaging, Adapted from Mohan *et al*., 2019

2.2. Polysaccharides material

Polysaccharides are long chains of polymers formed by mono or disaccharides as a repeating unit through glycosidic bonds. Hydrogen bonds are of great importance for the formation of polysaccharide films and their properties since these materials have a high content of hydroxyl groups, as well as other hydrophilic groups present in their structure (Mohan *et al*., 2019). Polysaccharides based coatings are predicted to be an efficient oxygen blocker due to their well-ordered hydrogen-bonded network shape. These coatings are colorless, have an oily-free appearance and a minor caloric content that can be used to prolong shelf life for fruits, vegetables, shellfish, or meat products by reducing its dehydration, darkening of surface, and oxidative rancidity (Hassan, 2018).

Normally, the production of polysaccharide films is from the interruption of interactions between the polymer segments during a coacervation process and by forming new intermolecular hydrophilic bonds and hydrogen bonds after the evaporation of a solvent (ethanol or water). In summary, polysaccharide films have an excellent aroma, oxygen and oil barrier properties, as well as structural and strength integrity, but they have the defect of having little resistance to water migration. As shown in Figure 1, polysaccharides such as starch, alginate, carrageenan, cellulose, agar, pectin, and chitosan are the most frequently used edible materials for the development of edible films (Du *et al*., 2016; and Mohan *et al*. 2019).

2.3. Composite material

A composite is a material where two or come components are combined. The composition consists of a matrix and reinforcement separated by a distinct interface (Mushtaq *et al*., 2016). The reinforcement represents the dispersed phase, which will be the one that increases the properties of the resulting material; it will be protected by the matrix, the continuous phase. Because of their mixed nature, the resulting material will present improvement of properties by the combination of components due to the principle of combined action (Li *et al*., 2011).

Composites can be reinforced with particles, fibers, or through structural incorporations, as presented in Figure 2. Particle reinforced composites present excellent adsorption properties that have gained importance in recent years (Mushtaq *et al*., 2016). Their usage in food packaging is currently under study because, even though particle reinforced composites have high potential applications in the food packaging sector as an innovative material, safety concerns about them, together with the nanocomposites, are relevant due to the fact that only a few studies are available in the literature on the migration of nanoparticles from the package to the food (Li *et al*., 2011; Virkutyte and Varma, 2011). Fiber-reinforced composites are sub-classified by fiber length and orientation. They are very light but present high strength and stiffness as well. The composites reinforced through structural incorporations have properties that depend not only on the properties of the constituents but also on the geometrical design (Malkapuram *et al*., 2009).

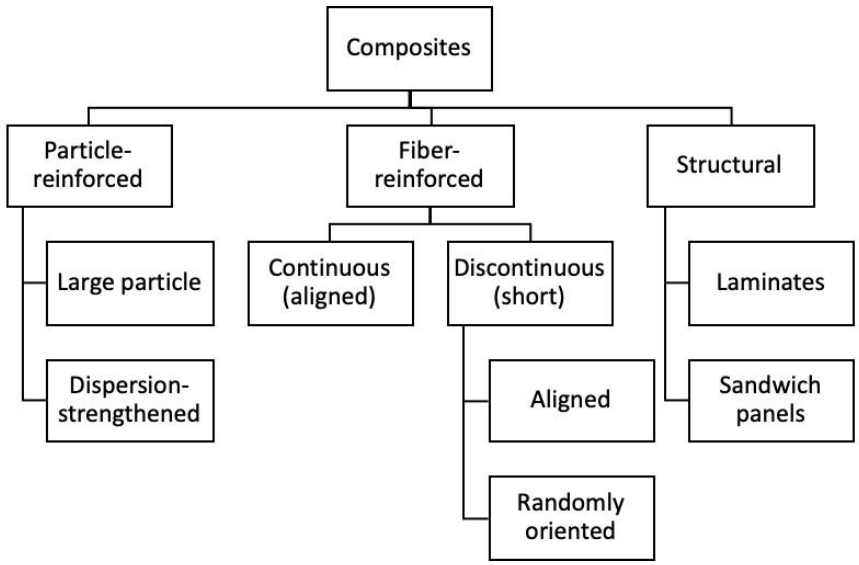


Figure 2: Composites classified by reinforcement, Adapted from Callister and Rethwisch, 2011

Biocomposites are generally fiber-reinforced because traction and a greater elastic modulus compared to the matrix are obtained (Jimenez *et al*., 2018). A strong fiber-matrix adhesion at the interfaces is desired and necessary for effective transfer and load distribution from the matrix to the fibers and vice versa (Thomas *et al*., 2013). Fibers used in biocomposites mainly come from an agricultural origin; leaves, stems, roots, and seeds are the precursors. Vegetable fibers are diverse, such as cotton, ceiba, flax, hemp, jute, ramie, sisal, henequen, and coconut, to mention some (Montalvo *et al*., 2012; Tahir *et al*., 2017; Thomas *et al*., 2013; and Kabir *et al*, 2013). All of the fibers mentioned are composite materials of cellulose fibrils reinforcement linked to a matrix of hemicellulose and lignin (Kabir *et al*., 2013; and Qiuhui *et al*., 2016).

Cellulose is the common factor in any vegetable fiber used for biocomposites (Kabir *et al*., 2012). It is a well-known natural polymer that presents great strength and rigidity per volume unit (Kabir *et al*., 2013). The mechanical properties of the vegetable fibers depend on the cellulose content, the degree of polymerization, and the microfibril angle (Milanese *et al*., 2011). Hence, cellulose-derived fibers vary.

Biocomposites reinforced by natural fibers present advantages and disadvantages. On one hand, the principal disadvantages of natural fibers are their hydrophilic character and their thermal sensitivity (Ausias *et al*., 2013). The consistency of fiber quality, their uncertainty regarding their properties depending on the crop location and processing conditions are also disadvantages in the usage of these fibers (Faruk *et al*., 2012). The advantages, on the other hand, are their low cost, low density, comparable tensile properties, are non-abrasive to equipment (Malkapuram *et al*., 2009). Also, their eco-friendly aspects are relevant, and also related to the sustainable goals established by the united nations, because they involve a reduction of energy consumption, renewable, widely available, the lower health risk to living beings, recyclable and biodegradable characteristics (Kabir *et al*., 2013; and Ausias *et al*., 2013).

The properties of fiber-reinforced composites depend to a large extent on the interfacial adhesion between the matrix and the fibers, as well as on the number of fibers used as reinforcement. In general, an increase in the amount of fiber generates an increase in properties. tensile (Milanese *et al*., 2011). Currently, various types of natural fibers have been investigated to reinforce plastics, such as flax, hemp, jute, wood, rice husk, wheat, barley, oats, bamboo and sugar canes, grass, kenaf, ramie, palm oil, sisal, fique, coconut fiber, pineapple fiber, banana fiber, among others. (Ausias *et al*., 2013; Malkapuram *et al*., 2009; Milanese *et al*., 2011; and Jimenez *et al*., 2018).

3. Microbial polymers

Biodegradable plastics have been proposed as an alternative option due to the high contamination of synthetic plastics. Some bacteria are capable of producing a vast diversity of polymers, some of them have the chemical and material properties that are useful in industrial and medical applications, providing a valuable source of renewable, biodegradable, and biocompatible materials (Kunal and Rajor, 2011).

Bacteria efficiently convert different carbon sources into a diverse range of polymers. The four major classes of polymers produced by bacteria are polysaccharides, polyesters, polyamides, and polyphosphate. These polymerases serve as various proposes due to the biocompatibility of biodegradable properties (Rehm, 2010).

The following Table 1 shows the class of polymer, polysaccharides, polyesters, polyamides, and polyphosphate, which is located in the cells, the producer cells, and industrial applications.

Table 1: Classes of bacterial polymers and their characteristics, Adapted from Rehm, 2010

Polymer class	Primary structure	Main component	Precursors	Polymerizing enzyme	Producer Polyamides	Application Polyamides
Polyamides						
Cyanophycin granule peptide	Hetero-polymer	Aspartate and arginine	β-spartatearginine, l-arginine and l-aspartate	Cyanophycin synthetase	Cyanobacteria, *Acinetobacter* spp. and *Desulfitobacterium spp.*	Dispersant and water softener
Polyglutamate	Homo-polymer	d-glutamate and/or l-glutamate	(Glutamate) n-Phosphate, ATP and l-arginine	Polyglutamate synthetase	*Fusobacterium nucleatum* and the archaea *natronococcus occultus*	Humectant, drug delivery and cosmetics
ε-poly-l-lysine	Homo-polymer	l-lysine	l-lysine, ATP and l-lysine-AMP	ε-poly-l-lysine synthetase	*Streptomyces albulus subsp. lysino polymerus*	Food preservative
Polyester						
Polyhydroxyalkanoates	Hetero-polymer	R-3-hydroxy fatty acids	R-3-hydroxyacyl CoA	Polyhydroxy alkanoate synthase	Bacteria and archaea	Bioplastic, and matrices for displaying
Poly Anhydrides						
Polyphosphate	Homo-polymer	Phosphate	ATP	Polyphosphate kinase	Bacteria and archaea	Replacement of ATP in enzymatic synthesis

Aliphatic polyester: poly (3-hydroxybutyrate) [PHB] is recognized as a prototypical biodegradable thermoplastic to solve waste disposal challenges. This polymer is manufactured by a large scale fermentation process, involving the production inside bacterial cells, the bacteria are capable of containing as 90% of their dry weight as polymer. The bacteria capable of performing this feat is *Alcaligenes eutrophus* (Lenz and Marchessault, 2004).

Poly (3-hydroxybutyrate) is accumulated inside the cells when *Alcaligenes eutrophus* is limited to some essential nutrients like nitrogen, oxygen, phosphorus, or magnesium. The production of the copolymer of poly (3-hydroxybutyrate co 4-hydroxybutyrate) [poly(3HB-co-4HB)] can also be produced by *Alcaligenes eutrophus* due to the carbon source used in the synthesis *(* Peoples and Sinskey, A., 1989*)*.

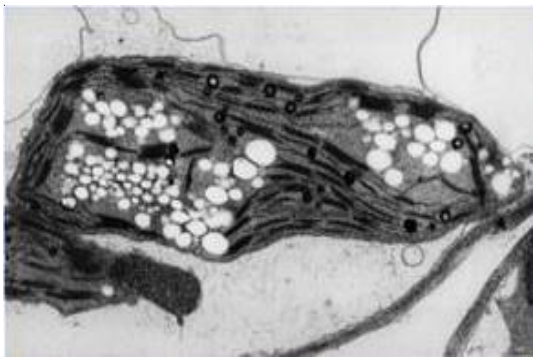


Figure 3: PHB granules in the chloroplast of Arabidopsis thaliana, Retrieved from Lenz and Marchessault, 2004

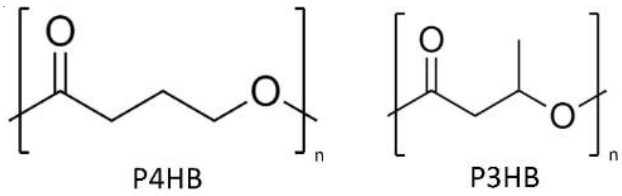


Figure 4: Monomers of poly (3HB-co-4HB). Retrieved from Sigma Aldrich

Table 2 shows the impact of composition between P4HB and P3HB in the synthesis of Poly (3HB-co-4HB) from different carbon sources by *Alcaligenes eutrophus* for 48 h at 30 °C (Saito *et al*., 1995).

Table 2: Production of P(3HB-co-4HB) from carbon sources by *Alcaligenes eutrophus*. Adapted from Saito *et al*., 1995

Carbon source *(a)*	g/litre	Polyester content *(b)*	PHA Composition (mol %) *(c)*		Molecular weight *(d)*	
			3HB	4HB	10^{-3} M_n	M_w/M_n
4-Hydroxybutyric acid	20	16	66	34	392	2 | 5
1,4-Butyrolactone	20	21	83	17	240	1 | 8
1,4-Butanediol	20	22	89	11	209	2 | 7
1,6-Hexanediol	5	36	87	13	120	2 | 3
1,8-Octanediol	5	13	79	21	79	5 | 4
1,10-Decanediol	5	13	91	9	291	3 | 0
1,12-Dodecanediol	4	17	91	9	535	2 | 5

Note: *a*-Carbon source in nitrogen-free medium; *b*-Polyester content in dry cells; *c*-Determined by H NMR.; and *d*-Determined by GPC.

4. Edible packaging

Biopolymers are the matrix of edible packaging. In order to be considered food-grade biopolymers, they must meet certain oxygen / water vapor permeability, tear and tensile strength, and other criteria described in the

following section (Wang *et al*., 2009). The biopolymers, together with the mechanisms their preparation involves, are presented as well.

4.1. Characteristics

Edible films are defined as a thin layer of polymer that can be consumed and used on the surface of a food, which can be applied as a covering during its preparation or as continuous layers between the different components used (Kowalczyc and Baraniak, 2011). Some desirable requirements in the films are control in permeability to water vapor and solutes, selective permeability to gases and volatile compounds, and control in the loss or exchange of flavor in foods, among others (Montalvo *et al*., 2012).

Although edible films made from biopolymers generally have good oxygen barrier properties, protein films are considered to be the most effective in this regard due to the complex structure formed (Okamoto, 1978). This complex structure results from the interaction of monomers that may be present in the matrix, which, depending on their nature (hydrophobic or hydrophilic), will have an effect on the properties of the edible film (Cuq *et al*.,1998). Other factors that may influence the properties of this type of film are the pH value of the protein solution, the use of plasticizers, the formation conditions, and the substances incorporated into the film-forming solution (Montalvo *et al*., 2012; and Okamoto, 1978). Several types of proteins, with particularities depending on their origin, have been used for the formation of edible films, among them are gelatin, casein, whey protein, wheat gluten, and soy proteins (Bhawani *et al*., 2017; Cuq *et al*.,1998; Khwaldia *et al*., 2004; Montalvo *et al*., 2012). In Table 3, Cuq *et al*. (1998) presents a table composed of the main proteins used to form edible films, which includes the previously mentioned proteins, as well as corn zein, keratin, collagen, peanut, cottonseed, egg albumin, and myofibrillar proteins. The tested methods to obtain the edible films are compared in the table, making visible the great usage of film-forming solutions and skin collection to produce edible films.

Table 3: Main proteins used to produce edible films, Adapted from Cuq *et al*., 1998

Proteins	Film-Forming Solution *a*	Collect Skin *b*	Enzymatic treatment *c*	Thermoplastic extrusion
Corn zein	+			
Wheat gluten	+	+		+
Soy proteins	+	+		
Peanut proteins		+		
Cottonseed proteins	+			
Keratin	+			
Collagen				+
Gelatin	+			
Caseins	+	+	+	
Whey proteins	+	+	+	
Egg albumin proteins	+	+		
Myofibrillar proteins	+			+

Note: *a* Thin-layer coating and drying of a film-forming solution; *b* Collect the skin formed after the boiling protein solution; and *c* Enzymatic polymerization.

Film-forming solution methodology is a wet process used to obtain edible films which relay in the dispersion conditions of the proteins in the mentioned solution, and in the spreading condition of the mixture to make films or coatings (Cuq *et al*., 1998). A dry process alternative is one based on the thermoplastic properties of the

proteins used, where they melt and glass transitions are vital to be able to shape the powder or pellets reacting with the proteins in order to obtain films or bio packaging.

4.2. Biopolymers used in edible packaging

As mentioned before, biopolymers enhance desired properties for food packaging. There has been a wide investigation of these, some polymers include edible and biodegradable polymers as prominent polymers for packaging in the food industry (Cerqueira *et al*., 2013) since they improve gas / moisture barrier, mechanical properties, microbial protection, longer shelf life among others. Mostly, edible films in packaging are composite films, therefore, they incorporate different structural materials such as proteins, polysaccharides, and lipids in order to develop an enhanced film due to the properties that each component provides (Galus and Kadzinska, 2015).

One of the optimum characteristics of an edible film involves its biodegradability. In order to accomplish this, edible films are mainly composed of proteins such as collagen, gelatin, whey protein, soy protein, corn zein, egg white protein, and keratin. Main polysaccharides used in film´s development include starch, cellulose, and cellulose derivatives such as pectin, chitosan, and alginate. Lipidic components include the incorporation of animal and plant waxes, vegetable oils, and fatty acids. Specific lipid examples used in edible films are olive oil, sunflower oil, sugar cane waxes, paraffin, beeswax, lanolin, triglycerides to mention a few (Galus and Kadzinska, 2015). Besides the addition of the previously mentioned components, emulsifiers are needed to prevent the separation of lipids from polysaccharides (Chandra *et al*., 2016).

As well, antioxidants and microbial components are also incorporated in order to achieve active edible films (Chandra, 2016; and Piñeros *et al*., 2016). Also, plasticizing agents are generally integrated into the composite film in order to allow modification of polymeric structures by increasing free volume and chain flexibility in order to lower film brittleness (Pérez *et al*., 2013). By the addition of a higher concentration of plasticizers, as reported for xanthan gum, tensile strength and elongation break properties of the edible film were manipulated causing an increase and decrease respectively in the aforementioned properties (Chandra *et al*., 2016). The incorporation of plasticizers is essential because, without them, the formation of pores within the film will be present, enabling water vapor permeability (Rezaei and Motamedzadegan, 2015).

Also, by taking advantage of the use of agricultural residues, the production of such edible films is possible (Tulamandi *et al*., 2016; and Chandra, 2016). Tulamandi, S. *et al*, have designed edible film composites based on papaya, gelatin, and soy protein because papaya harvest losses round up to 75%, and also since it provides high pectin content (Tulamandi *et al*., 2016). As well, Chandra *et al*., 2016 produced spice fused tamarind seed starch edible films. Piñeros *et al*. (2017) incorporated rosemary antioxidant extracts into edible cassava starch film in order to reduce food contamination and spoilage.

4.3. Fabrication methods

Different methods of obtaining edible films for packaging purposes have been investigated. Most methods involve the use of citric acid-starch (CA-starch), chitosan polymer, or acacia lignin, also composites made with gelatin were used to improve the properties of the material.

To prepare the gelatin solution for CA-Starch composite, it was done by mixing gelatin (8 g) and sorbitol (25% w / w) in 100 mL of distilled water for 10 min at room temperature, then the solution was heated in a water bath (60 °C) for 20 min, with constant stirring, the solution then was cooled until it reached 40 °C (Kumar *et al*., 2019).

CA-Starch solution (Kim *et al*., 2017) was prepared by blending 3 g of starch and 25% of sorbitol (% w / w of starch) as a plasticizer in 100 mL of distilled water. The solution was heated in a water bath (90 °C) for 10 min with stirring while stirring citric acid was added into the starch solution (different concentrations of CA were prepared 0.5, 1, 3, 5 and 7% w / w of starch) the solution is kept at room temperature until reach °C then the solution was poured into a mold then it was dried at 25 °C for 24 h.

A Gelatin / CA-Starch composite (Al-Hassan and Norziah, 2012) was prepared by mixing the previously mentioned solutions before dry (different ratios where applied, 1:0, 1:1, 1:4, 4:1, 0:1 CA-Starch: Gelatine) the solutions were homogenized. The prepared solutions were degassed by a vacuum oven. The solution was poured into a mold then dried at 25 °C for 24 h.

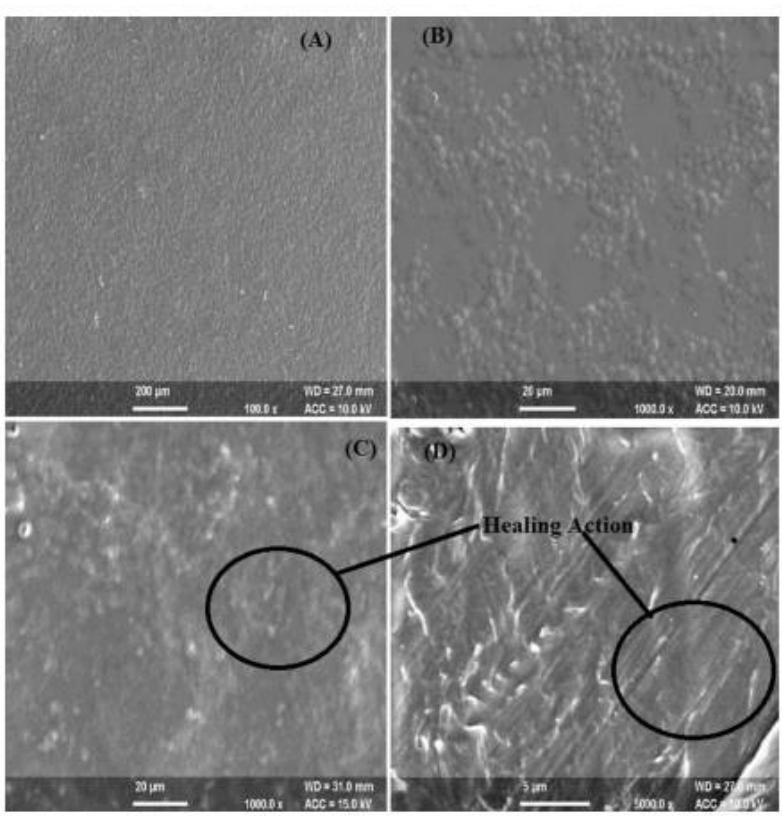
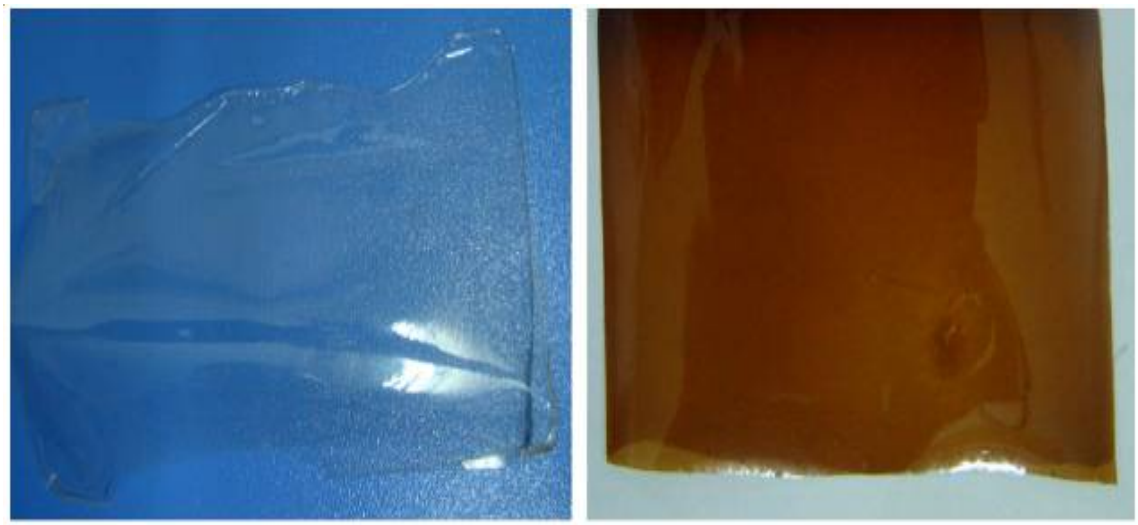


Figure 5: SEM images of citric acid modified (a), potato starch films (b) and, CA-starch/gelatin composite films (c), (d). Retrieved from Kumar *et al.* 2019

The preparation of lignin-gelatin film (Keshaw *et al*. 2016) consists first of the Gelatin powder (4% w / v) dissolved in Millipore deionized water at 60 °C for 30 min. During the time lignin (previously obtained from acacia wood powder) (1% w / v in 0.1 NaOH) was added (different ratios were prepared 100:, 90:10 80:20, 70:30 and 60:40 gelatin:lignin). To plasticize the gelatin-lignin solution glycerol (0.6% w / v) was added. The



Control Film (LG-A) **Lignin-gelatin cross-linked film (LG-D)**

Figure 6: Prepared gelatin (LG-A) and lignin-gelatin cross-linked film (LG-D). Retrieved from Keshaw *et al.*, 2016

mixture was stirred at 60 °C for an hour. After stirring the mixture was poured into a precoated at 60 °C Petri plate, the films were dried in desiccators containing P_2O_2 at 25 °C in a 0% humidity environment.

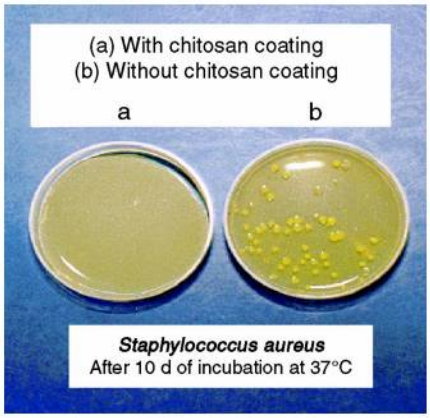


Figure 7: Development of Staphylococcus aureus inoculated on agar plate after 10 days of incubation at 37 °C (a) with chitosan coating and (b) without chitosan coating. Retrieved from Coma et al., 2003

The preparation for chitosan film (Coma *et al*., 2003) was obtained by dispersing chitosan in a 1% (v / v) aqueous acid solution. Then pH was adjusted to 5 using NaOH 1M. The preparation was filtered through membranes of 5.3 μm and then 0.65 μm and finally degassed under reduced pressure.

5. Properties

5.1. Mechanical

Food packaging requires some physical protection to keep it in good condition against shocks and vibrations they are exposed to during transportation (Mohan *et al*. 2019). These protections are mainly composed of cardboard and corrugated materials that resist impact, crushing, and abrasion damage, so they are used in fragile food containers such as fruits and eggs. Thus, some mechanical properties should be optimized such as elastic modulus, tensile strength, elongation at break, compression strength, stiffness, puncture strength, tearing strength, abrasion resistance, adhesion force, etc (Mohan *et al*., 2019).

In a study of Ramos *et al*. (2013) a characterization study of an edible package consisting of two types of proteins was carried out: whey protein isolate (WPI) and whey protein concentrate (WPC) added to three amounts of glycerol (40, 50 and 60% w / w). In this study, the thermal, barrier, tensile, and optical properties were determined to observe the effect of the nature of the protein and the content of the plasticizer.

Figure 8 shows the data of tensile properties for the WPI and WPC films reported by Ramos *et al*. (2013) According to the authors, WPI films showed significantly higher values ($p < 0.05$) of tensile strength (TS),

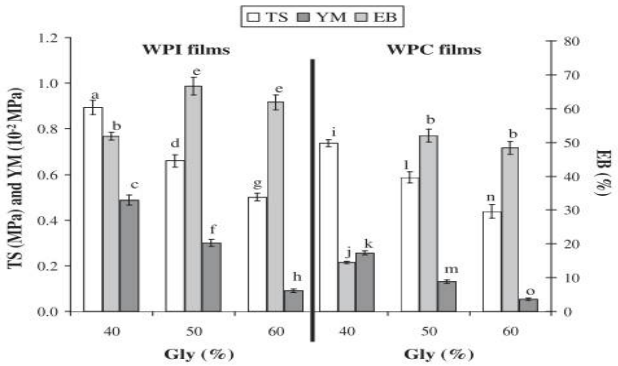


Figure 8: Values of TS, EB, and YM of 10% (w/w) WPI and WPC edible films with various contents of glycerol

elongation at break (EB), and Young's modulus than WPC composite films with the same content of glycerol; which is confirmed by the thermal characterizations and the FTIR. Additionally, they observed an increase in the elasticity and elongation properties proportional to the glycerol content; observing a content percolation of 50% (w / w) in the WPI films. This effect is due to the intermolecular space, and therefore, to the movement of the polymer chains. According to a study reported by Barreto *et al*. (2003) where the characterization of the thermal degradation of films prepared with sodium caseinate, serum and gelatin in the presence of a sorbitol plasticizer was carried out. In this study it is mentioned that the thermal stability is affected by the interaction of sorbitol with the inter and intramolecular hydrogen bonds of the whey protein. Therefore, the effect of increasing glycine produces the aforementioned effect where the interactions between glycine and hydrogen bonds of the protein produces a spacing that increases the elongation and elasticity properties (Barreto *et al*., 2003).

In another study by Chiumarelli and Hubinger (2012) the characteristics of an edible packaging formulation prepared from glycerol, cassava starch and carnauba were studied. In this study the stability of the emulsion and the barrier properties applied to fresh apple slices and the solubility and mechanical properties were evaluated through a 2^3 central compound rotary experiment design.

Table 4 reports the results obtained by Chiumarelli and Hubinger (2012) for the properties of tensile force, elongation at break and elastic modulus. Initially, it is mentioned that the cassava and carnauba starch-based films had a tensile strength that varied between 0.252 and 2,138, having lower values in the experiments with

Table 4: Central composite design matrix with real and coded values (in parenthesis) of the variables and responses of mechanical properties (tensile strength - TS, elongation at break - ELO and elastic modulus - EM), edited from Chiumarelli and Hubinger, 2012

Corrida	almidón de yuca (%)	Glicerol (%)	Cera de carnauba: ácido esteárico (%)	RT(Mpa)	AR(%)	ME (Mpa)
1	2.4 (–1)	1.4 (–1)	0.08:0.92 (–1)	0.830	25.383	0.169
2	3.6 (+1)	1.4 (–1)	0.08:0.92 (–1)	0.940	31.357	0.252
3	2.4 (–1)	2.6 (+1)	0.08:0.92 (–1)	0.341	10.224	0.145
4	3.6 (+1)	1.4 (–1)	0.08:0.92 (–1)	0.492	51.064	0.013
5	2.4 (–1)	1.4 (–1)	0.32:0.92 (+1)	0.566	22.866	0.126
6	3.6 (+1)	1.4 (–1)	0.32:0.92 (+1)	1.193	24.522	0.458
7	2.4 (–1)	2.6 (+1)	0.32:0.92 (+1)	0.360	19.298	0.078
8	3.6 (+1)	2.0	0.32:0.92 (+1)	0.274	28.275	0.133
9	2.0 (–1.68)	2.0	0.2:0.8 (0)	0.252	32.055	0.116
10	4.0 (+1.68)	1.0 (–1.68)	0.2:0.8 (0)	0.536	35.500	0.072
11	3 (0)	3 (+1.68)	0.2:0.8 (0)	1.070	31.089	0.444
12	3 (0)	3 (+1.68)	0.2:0.8 (0)	0.259	23.710	0.143
13	3 (0)	2.0 (0)	0:0 (–1.68)	2.138	34.256	0.166
14	3 (0)	2 (0)	0.4:0.6 (+1.68)	0.352	36.230	0.131
15	3 (0)	2 (0)	0.2:0.8 (0)	0.548	3.766	0.087
16	3 (0)	2 (0)	0.2:0.8 (0)	0.496	34.083	0.119
17	3 (0)	2 (0)	0.2:0.8 (0)	0.388	45.627	0.080

lower starch content (2% w / w). Additionally, the experiments without added lipids obtained the best results because they contribute to forming inflexible and breakable films.

In the case of elongation, it is expressed between the ratio of the length of the sample until the rupture and its original length. The authors reported elongation percentages of between 10 and 51%, which were affected by the concentration of the plasticizer. The results demonstrated a high dependence on the content of starch and glycerol, since the experiments with the lowest content of these obtained the lowest elongation values, as in the study by Ramos *et al*. (2013).

Similarly, glycerol had an important effect on Young's modulus, since in the table results which vary from 0.013-0.458 MPa, the increase in glycerol content resulted in a more elastic material and therefore lower values in Young's modulus. The relationship between starch and carnauba was determined to produce a stiffer material with higher Young's modulus values.

In a study by Cao *et al*. (2007) the mechanical, swelling and optical properties of films composed of soy protein and gelatin were evaluated. The authors report that in preparing a film with higher gelatin content, the tensile properties, the elongation at break, the elastic modulus and the swelling property of composite films. This effect can be observed for the compounds with a ratio of 4:6 and 2:8 in Figure 9. In addition, it is mentioned that it must produce a more combustible composite film than it produces a gelatin-only film, so its use as an edible film is convenient.

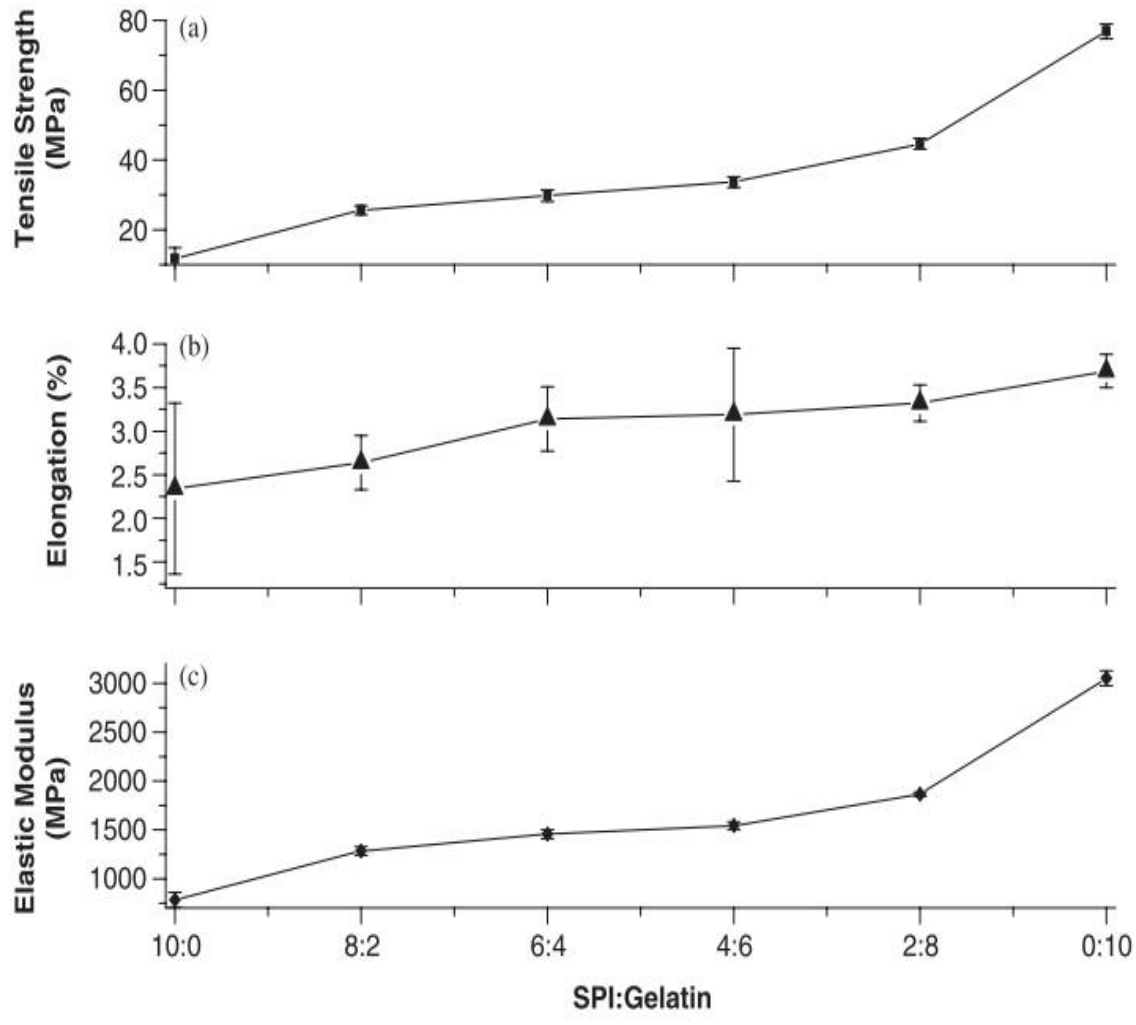


Figure 9: Tensile strength (a), elongation to break (b), and elastic modulus (c) of composite films prepared with different radius of SPI/gelatin ratios (containing 0.1 g glycerin/g protein)

In another study by Kennedy *et al.* (2006) gelatin compounds mixed with konjac glucomannan were prepared, where the compounds were reported to have increased tensile strength and elongation at break. This effect was attributed to the hydrogen bonds between the two polymers that produced a plasticizing effect in films that absorbed water.

5.2. Degradation study

One of the main characteristics of an edible film involves its biodegradation, meaning that it can be degraded by microorganisms through an enzymatic process. By this, the biodegradation process depends on the polymer´s structure and degradation environment. Generally, polymers used in packaging come from non-renewable resources resisting biodegradation. However, some of them can possess biodegradability properties, such polymers include hydrolyzable backbones in their structure, mainly ester, amide, and urethane backbones. By the addition of antioxidant components, polymers will correspond to those from the oxo-degradable group, since they will act against UV light, and by auto-oxidation biodegradation can occur (Vroman and Tighzert, 2009).

Biodegradation studies of starch film with yerba mate extract as antioxidant were developed by the usage of vegetable compost in order to simulate real life conditions. Figure 10 shows the result of the before-mentioned studies, it can be observed that in just less than a week, edible films suffered changes in appearance as well as breakdowns, meaning that biodegradation process started. It has been reported that the starch film degradation process occurs faster than other synthetic biodegradable polymers, for example poly (lactic acid). Because of this, almost total biodegradation resulted at day 12, and the addition of natural antioxidant extract, in example yerba mate extract, resulted in a faster process. As it is shown, edible starch films containing 5 and 20% of extract showed further deterioration at day 12 (Medina *et al*, 2016).

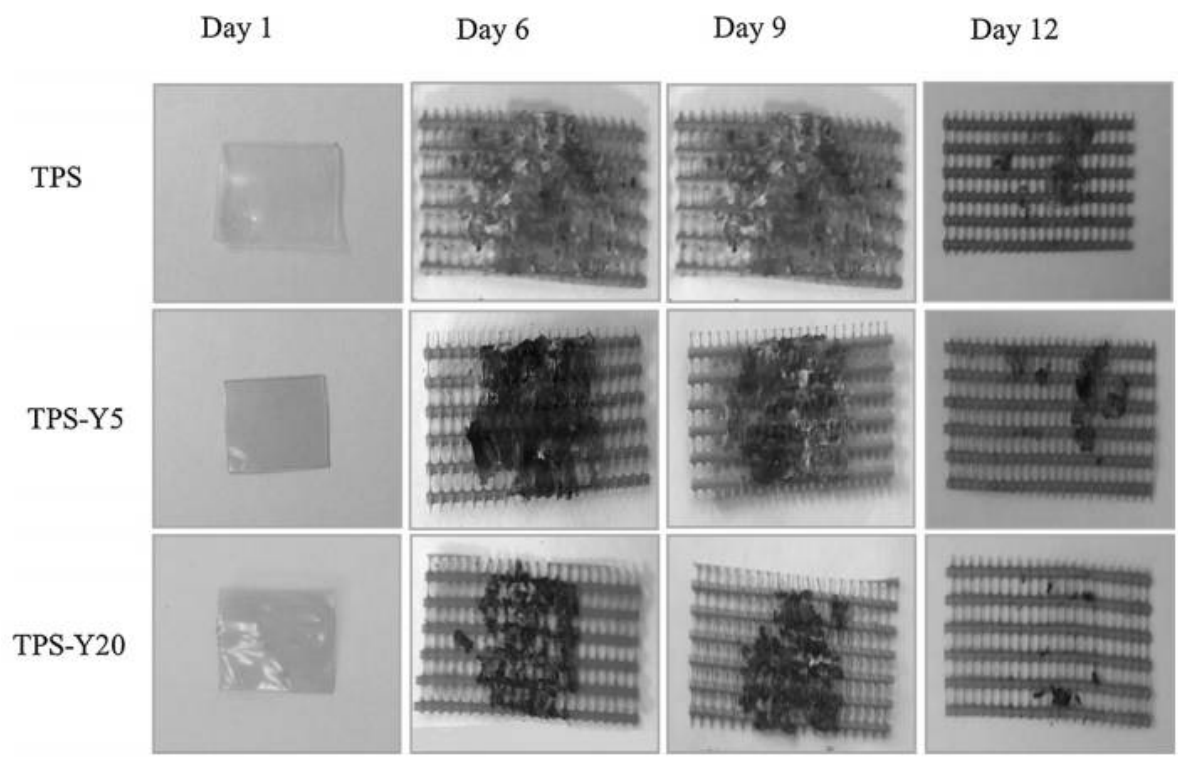


Figure 10: Biodegradation study of cassava-starch containing *yerba mate* extract in vegetal compost. Retrieved from (Medina *et al*, 2016)

By the incorporation of rosemary antioxidant extract into cassava starch edible films, the biodegradation process can be also assured after 7 days of studies (Piñeros *et al.*, 2017). However, the addition of this extract in 5, 10 and 20% lead to a retardation in biodegradation since, at day 14 (Figure 11), major film integrity can be observed when compared to the addition of yerba mate extract in Figure 10.

Figure 11: Biodegradation study of cassava starch films incorporating rosemary extracts. Retrieved from (Piñeros *et al.*, 2017)

Biodegradation process is of interest in edible packaging since it will allow polymers degradation in landfills which might result in a decrease in waste packaging leading to environmental protection. As shown previously, antioxidant components might be of help within this process. Yerba mate extract containing low molecular weight compounds in relation to starch led to a faster degradation process, allowing edible film's almost complete degradation in vegetal compost by day 12 (Medina *et al*, 2016). Other methods such as random or block copolymerization will lead to an improvement in biodegradation rate (Vroman and Tighzert, 2009).

5.3. Permeability

Generalization of water vapor properties of films can't be assured, since properties of edible films depend on the structural materials used, their compatibility and film preparation. For example, by using high degree components, more cohesion between them is achieved causing a homogeneous structure in the film. Improving water barrier resistance is one of the main properties to be enhanced since variations among water desorption and absorption affect negatively food's properties (Galus and Kadzinska, 2015). Therefore, carrying out Water Vapour Permeability (WVP) studies will demonstrate the capability of water to pass through the edible films, in other words it will demonstrate its breathability. Generally, in order to achieve water barrier property in the film, incorporation of lipids is required aside from proteins and polysaccharides. Saturated fatty acids are incorporated due to their lower polarity when compared to unsaturated ones (Galus and Kadzinska, 2015).

A decrease in WVP can be achieved by the incorporation of linear and globular proteins (Tulamandi, 2016). Gelatin and defatted soy proteins reduced WVP from 8.45 to 5.55 g mm kPa^{-1} h^{-1} m^{-2} (Tulamandi, 2016). Nonetheless the addition of natural antioxidant and microbial components can significantly WVP (Piñeros, 2017). As reported by Piñeros *et al*. (2017) due to the addition of rosemary extracts into the edible cassava starch film, WVP increased from 5.8 to 11.0 $gs^{-1}m^{-1}pa^{-1} \times 10^{-10}$, however this can be attributed to the surface defects on the film. Nonetheless by the addition of such extract in much lower concentrations such as 5% resulted in similar expected WVP values as control group differing just in 0.2 $gs^{-1}m^{-1}pa^{-1} \times 10^{-10}$. As well, spice powders possess antioxidant properties, high concentration of polyphenols is related to this property, due to this, clove

has been demonstrated as a desired antioxidant when compared to turmeric, black pepper and cinnamon (Upadhyaya *et al.*, 2017).

Plastisizers also influence WVP, it has been reported that by using glycerol, higher WVP values are obtained in comparison with PEG-400 and sorbitol. Sorbitol will provide better moisture barrier properties for the film, since in edible films, lower WVP is expected (Sharma and Singh, 2015; and Rezaei and Motamedzadegan, 2015). Glycerol will be liable to the incorporation of water because of its hydrophilic nature. Also, plastisizers with higher amounts of hydroxyl groups will increase WVP, such as PEG-400 (Chang and Nickerson, 2014).

Another measurement that provides information about water resistance property corresponds to water contact angle (WCA), by this, film's surface hydrophobicity can be evaluated (Tulamandi *et al.*, 2016). Gutiérrez and González developed a plantain flour edible film incorporating *aloe vera* gel, as it can be observed in Figure 11, WCA measurements showed that increasing concentrations of aloe vera gel produced higher contact angles values. The before mentioned, due to the ability of aloe vera gel to cross-link on starch present in plantain flour (Gutiérrez and González, 2016).

WCA of papaya films were reported to be 29.34 ± 0.4º, resulting in wettable films due to their hygroscopic structure. By the addition of 3 g w / w gelatin, cross-linking to papaya occurred resulting in a higher increase of WCA to 78.14 ± 0.57º (Tulamandi *et al.*, 2016). By these results, it can be shown that WCA measurements for edible film's can be similar, or even higher that some synthetic polymers, such as polycarbonate (70º), nylon 6-6 (70º), and PTFE (88º) (Basiak *et al.*, 2017).

6. Applications

The use of edible films and coatings in foods has several objectives, the most important of which are: to reduce moisture loss, to control gas permeability, to control microbial activity, to preserve the structural integrity of products and to allow gradual release of flavor and of antioxidants in foods (Wang *et al.*, 2015).

Many applications of edible packaging can be made in the food processing. Previously, carrageenan-based coatings had been used in the meat industry to prevent superficial dehydration of fresh and frozen meat (Debeaufort *et al.*, 1998). Improvements have been made, and now carrageenan-based coatings are being used as wrapping material on minimally processed chicken breast fillet because of its ability to increase not only its vapor barrier properties, but also the weight loss prevention significantly ($p < 0.05$) compared to the non-wrapping fillets (Yousefi *et al.*, 2018).

Calcium alginate coatings started to be used in beef to maintain the desired color the years ago (Williams *et al.*, 1978). Nowadays, calcium alginate coatings are used as antimicrobials against a wide range of bacteria: *Listeria innocua, psychrophilic aerobic bacteria, yeasts, molds psychrotrophs, coliforms, E. coli and B. cinereal, Brochothrix thermosphacta, Enterobacteriaceae, P. aeruginosa, L. monocytogenes*, and *aerobic mesophilic* bacteria (Sipahi *et al.*, 2013 ; Peretto *et al.*, 2014; Fang and Tsai, 2013 ; Yousefi *et al.*, 2018; and Concha-Meyer *et al*. 2011). Pranoto *et al*. (2005) gave an alginate-based edible film antibacterial properties by incorporating garlic oil as a natural antibacterial agent; the agar diffusion assay, shown in Figure 12, shows their results.

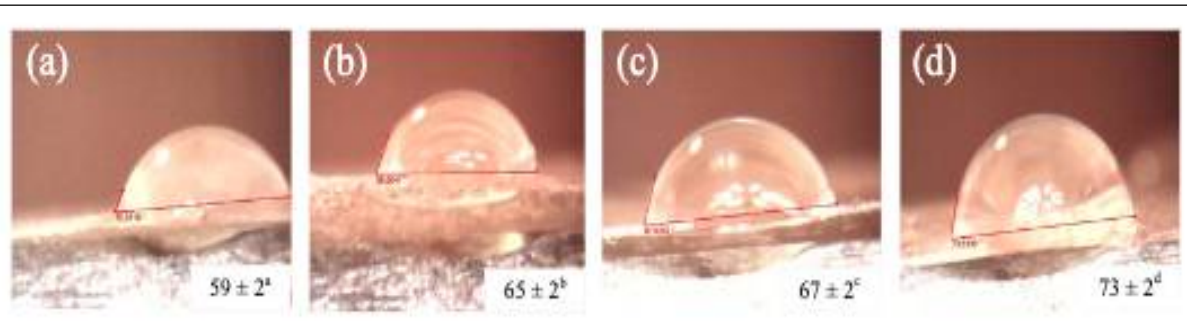


Figure 12: Water Contact Angle measurements of plantain flour film with *aloe vera* gel at (a) 0%, (b) 2%, (c) 4% and (d) 6% (Gutiérrez and González, 2016)

Calcium alginate, as well as other polysaccharides, can also be used with antimicrobial properties in edible coatings. Vu *et al.* (2011) increased the shelf life of strawberries during storage through bioactive coatings based on limonene and the emulsifier Tween 80. They were applied using modified polysaccharides of chitosan, and the results are shown in Figure 13. Is visible how the control fruit was affected by bacteria drastically more when not coated. Vu *et al.* (2010) demonstrated how functionalized chitosan-based edible coating is becoming a promising method to carry specific antifungal agents without detrimental effects on strawberries or other fruits; complete absence of change in flavor or color was reported (Vu *et al.*, 2010).

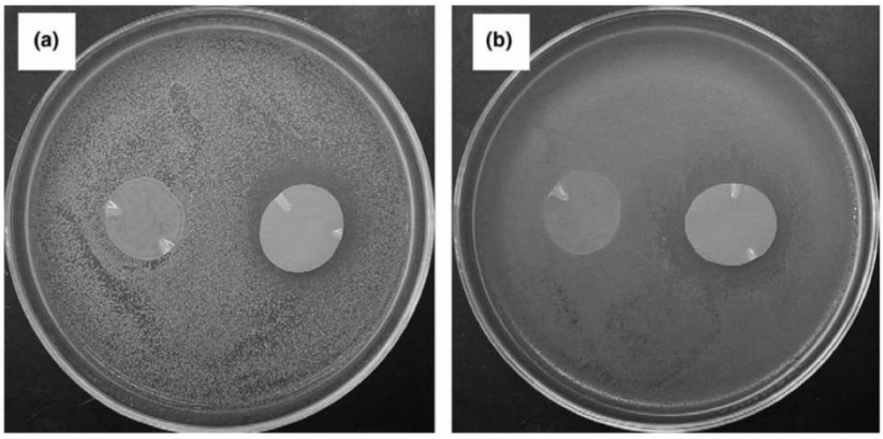


Figure 13: Alginate edible film's inhibitory zone of incorporated with 3% v/v garlic oil (right strip) compared to control (left strips) against (a) *Staphylococcus aureus* and (b) *B. cereus.* Retrieved from Pranoto *et al*., 2005

When edible coatings are applied to fruits and vegetables with minimum processing, the properties offered are a semi-permeable barrier to gases and water vapor and reduce the rate of respiration. In addition, they prevent color changes, mechanical integrity deformation, water loss, texture deterioration, decrease of flavor and microbial growth, giving a longer life to shells (Bhawani *et al*., 2018; Montalvo *et al*., 2012; and Wang *et al*., 2015). A barrier that slows the loss of flavor and water can be created by edible films (Montalvo *et al*., 2012). They can also create a modified atmosphere with relatively high levels of CO_2 and low O_2 to restrict the exchange of CO2 and O2 (Bhawani *et al*., 2018; Montalvo *et al*., 2012; and Wang *et al*., 2015). This atmosphere should not, however, create anaerobic conditions that may cause anaerobic respiration, undesirable taste and growth of anaerobic microorganisms (Balwin and Nisperos-Carriedo, 1995).

Balwin and Nisperos-Carriedo (1995) registered a high respiration rate and ethylene production in minimally processed products, resulting from the process, can theoretically be reduced by applying a semipermeable membrane like edible coatings. Recent studies by Wang *et al*. (2015) with composite coatings based on different polysaccharides, such as cellulose, alginate, pectin, and microcrystalline, showed that "all coatings tested substantially reduced the rate of production of CO2 and ethylene in the product, with a reduction of ethylene of approximately 90%" (Wang *et al.*, 2015). Hence, composite coatings based on polysaccharides are promising in the edible food packaging industry (Balwin and Nisperos-Carriedo 1995 and 1996; and Wang *et al*., 2015).

Besides the promising reported properties of edible coatings and films, the consumers must be taken in account as well. The consumer acceptance of a new product is always challenging, and this difficulty increases if a product that is ingested is the emergent one. Subjective preferences of the consumers depend on properties such as flavor, texture, and appearance, to mention some. All of these properties are organoleptic properties (Han, 2014). In Figure 14, Han (2014) presents the edible film and coating material properties relations with process parameters. How the surface tension (γL) of coating solution affects the adhesion of films during a film-forming process determines the consumer's first interaction with the product; it may make it harder or easier to peel, as shown in Figure 15. This surface tension is caused by the energy in a flat-base solid, being the surface energy γS. When the surface is modified, peeling of the product is made harder. Hence, higher viscosity of the film-forming solution is desirable to reduce this coating phase separation. Precaution of whether edible coatings or films are appealing to the consumers is an important factor to take in account while thinking of the application desired.

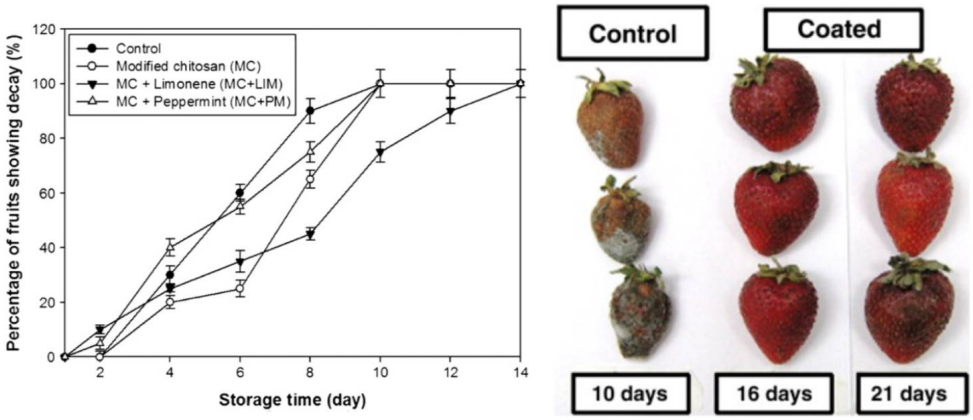


Figure 14: ***Left:*** **Mould contamination percentage in strawberries coated with modified chitosan containing limonene during storage at 4 °C.** ***Right:*** **Appearance of strawberries coated with modified chitosan-based formulation containing limonene, Adapted from Vu *et al*., 2011**

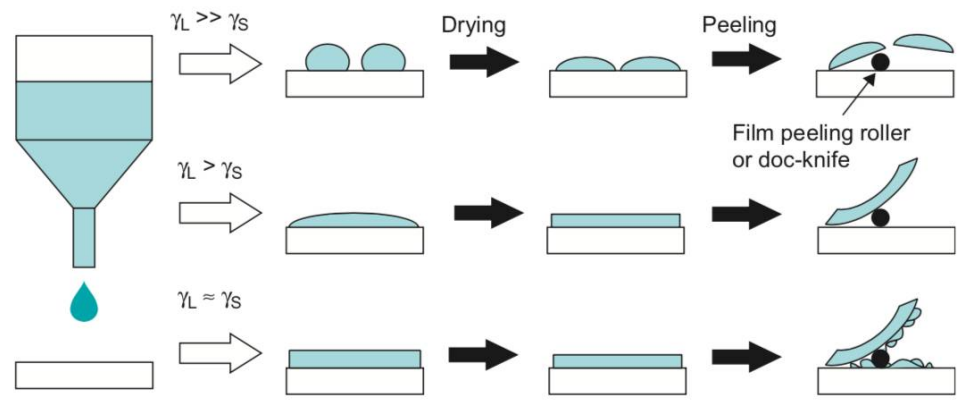


Figure 15: Effect of surface tension (γ L) of coating solution on the adhesion of films during a film-forming process. Retrieved from ch. 9 p. 239, Han, 2014

7. Conclusion

Edible films and coatings are an effective new way of food packaging. Prevention of water content evaporation, loss of desirable odor and flavor volatiles, respiration suppression, gas exchange, and antibacterial behaviors are the desired characteristics obtained through their usage. The extension of shelf-life given by edible food packagings contribute to responsible consumption and production, climate action, and life below water, which are part of the sustainable goals declared by the UN. Because edible films and coatings are meant to be consumed by users, they must address all regulations regarding food products and take in account their sensorial experience using edible films in order to apply them in the market. Their labels should be clearly identified and properly aligned to the regulations established by the country of interest.

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