

## Review

# Polysaccharides: Sources, Characteristics, Properties, and Their Application in Biodegradable Films

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**Abstract:** Biodegradable films emerge as alternative biomaterials to conventional packaging from fossil sources, which, in addition to offering protection and increasing the shelf life of food products, are ecologically sustainable. The materials mostly used in their formulation are based on natural polysaccharides, plasticizing agents, and bioactive components (e.g., antimicrobial agents or antioxidants). The formulation of biodegradable films from polysaccharides and various plasticizers represents an alternative for primary packaging that can be assigned to specific food products, which opens the possibility of having multiple options of biodegradable films for the same product. This review describes the main characteristics of the most abundant polysaccharides in nature and highlights their role in the formulation of biodegradable films. The compilation and discussion emphasize studies that report on the mechanical and barrier properties of biodegradable films when made from pure polysaccharides and when mixed with other polysaccharides and plasticizing agents.

**Keywords:** polysaccharides; natural polymers; sources; characteristics and properties; biodegradable films



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## 1. Introduction

Polysaccharides are the most abundant natural polymer on Earth, which are composed of 10 or more repeating units of monomeric sugars that are linked by glycosidic bonds [1]. These biological macromolecules have vital functions (e.g., structure and energy production) in the organisms that possess them; however, their importance for human beings is due to their functional properties demonstrated, e.g., as antitumor and antioxidant agents, as well as regulators of intestinal flora [2].

In the industry, polysaccharides are valuable because they are a potential replacement for petroleum-based polymers; however, it is known that their application has not been fully exploited, despite their versatility [3]. The study by Souza et al. [4] revealed that the use and application of these natural materials has grown by 17% in recent years (2017–2021), which is equivalent to a world market of up to 10 billion USD; in addition, it is expected that, at the end of this decade (2030), this market will be more than 22 billion USD. In this sense, there is interest in testing alternative polysaccharide extraction processes that improve yields or provide purer extractions, as well as alternative sources (e.g., residues) to extract new polysaccharides [4].

The conventional extraction of soluble polysaccharides is by water extraction, while, for non-soluble ones, mixtures of different organic solvents are used (e.g., ethanol, methanol, and acetone) that allow modifying the loads and carrying out the extraction. However, new methodologies and processes (such as ultrasonic-assisted extraction, microwave extraction, supercritical fluid extraction, and enzymatic extraction) have been tested, due to the advantages they bring in terms of improvements in extraction yield, time reduction, compound purity, and even environmental friendliness [2,5].

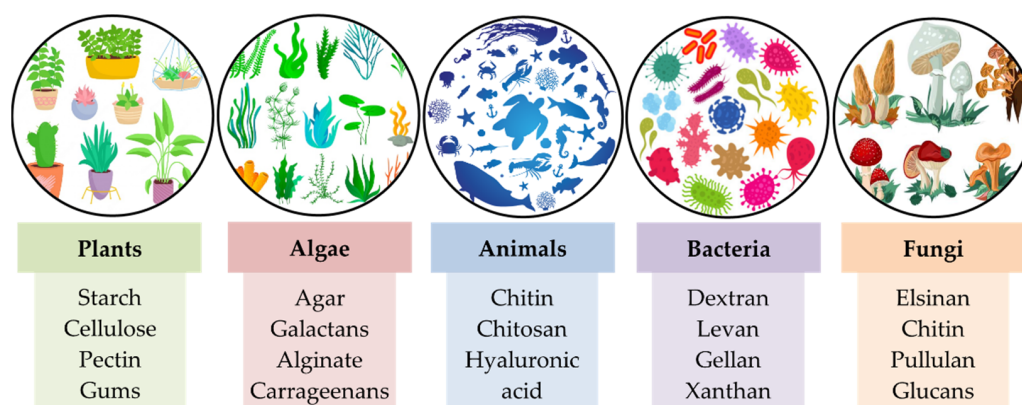
On the other hand, there is evidence that the extraction method significantly affects the properties and functionality of polysaccharides, in such a way that the application options are expanded. For example, polysaccharides with thermal stability can be subjected

to hydrolysis to obtain molecules with a lower molecular weight, polysaccharides with biological activity can be used as food supplements, and polysaccharides with high viscosity are good candidates as food additives or film formers [5].

A biodegradable film is considered a primary packaging made of biodegradable polymers, particularly polysaccharides, which has advantages over synthetic packaging [6]; specifically, it can preserve of quality and extend the shelf life of the minimally processed products [7,8] without damaging or altering the environment [9]. Therefore, this review provides an overview of the current state of the art of biodegradable films, paying special attention to the polysaccharides used for their formulation. In addition, the characteristics that biodegradable films must meet to be functional materials are described. The last section exposes the positive or negative changes that biodegradable films present when they are made with a mixture of polysaccharides and plasticizing agents.

## 2. Sources and Characteristics of Polysaccharides

Natural polysaccharides are synthesized to fulfill various biological functions (e.g., energetic and structural) in the organisms and microorganisms that produce them [1]. Figure 1 shows some examples of natural polysaccharides and their sources. Most natural polysaccharides are found in plants in the form of energy reserves; they can be synthesized by algae, bacteria, fungi, and yeasts or extracted from the exoskeleton of arthropods [10].



**Figure 1.** Natural polysaccharides classified according to their origin. Adapted from [1].

### 2.1. Plant Polysaccharides

Plant polysaccharides are macromolecules composed of identical or different monosaccharides linked by  $\alpha$  or  $\beta$  glycosidic bonds [11]. This class of polysaccharides is divided into two categories according to their functionality, storage, and structure; the first category encompasses polymers that are part of the energy reserves of plants, while the second category encompasses those that are part of the cell walls in such a way that they confer rigidity and flexibility to the plant [12]. Some plant polysaccharides are described below.

- Starch is a homopolysaccharide made up of glucose units, which form linear amylose chains linked by  $\alpha$ -(1–4) bonds and branched amylopectin chains that are connected to amylose by  $\alpha$ -(1–6) bonds [13]. Starch is the main storage carbohydrate of green plants, which has been isolated and used as a raw material for the manufacture of drugs, plastics, paints, and cardboard. Commercially, starch is mainly extracted from corn; however, there are other grains and tubers (e.g., rice, wheat, cassava, and potato) that are alternative sources of extraction. The functionality of starch is based on its properties of viscosity, water retention, and gel formation. However, there is research showing that these properties are influenced by the physical (e.g., particle size) and chemical (i.e., proportion of amylopectin and amylose) characteristics of the starch granules; therefore, it is very common to seek to modify the starch properties natives through physical, chemical, or enzymatic processes [14]. Starch is the most studied

natural polymer because it is a promising candidate due to its availability, low price, and high biodegradability [15].

- Cellulose is the most abundant, economic, and available carbohydrate polymer in the world because it can be extracted from plants or their waste [16]. Cellulose is a polysaccharide made up of a linear chain of glucose linked by  $\beta$ -(1–4) bonds. The hydroxyl groups present in its structure are responsible for the intermolecular hydrogen bonds that are formed, resulting in a compact and crystalline structure; however, these links can cause an irregular three-dimensional conformation causing amorphous regions in the molecule [17]. Cellulose is used in health areas for its antibacterial activity, however, its main application is in the generation of materials and matrices [16].
- Pectin is a polysaccharide found in the cell walls of terrestrial plants, mainly in the skin of fruits and vegetables (e.g., citrus). It is made up of a galacturonic acid backbone linked by  $\alpha$ -(1–4) bonds, which may have substitutions of rhamnose units by  $\alpha$ -(1–2) bonds, with side-chains composed of arabinose, xylose, or galactose [18]. Pectin is soluble in water and has a high viscosity, which is appreciated for generating gels; particularly in the food industry, these properties allow it to act as a food additive in the production of jams, jellies, and confectionery [19].
- Gums are highly branched complex carbohydrates composed of various sugars such as arabinose, galactose, rhamnose, and mannose; however, they can also be mixed with proteins or resins, characterized by having colloidal properties [20]. Gums are mainly extracted from plants, seeds, trees, and shrubs (e.g., Arabic, karaya, and cashew gums; however, they are also produced by bacteria. Gums are gelling, thickening, emulsifying, and stabilizing agents, with applications in the food, textile, pharmaceutical, cosmetic product, coating, encapsulant, and film industries [21].

## 2.2. Algal Polysaccharides

Polysaccharides are the most abundant macromolecules in the structure of algae, since they are found as mucopolysaccharides, i.e., structural and energy storage molecules [22]. Although between 4% and 76% of the dry weight of algae corresponds to polysaccharides, the content varies depending on the species of algae; for example, green algae have lignin, cellulose, and hemicellulose, brown algae have only cellulose, and red algae are made of dietary fiber [23]. Some polysaccharides found in algae are described below.

- Agar is a thermoreversible material composed of a linear chain of galactopyranose units linked by (1–4) bonds [24]. This gelatinous substance has gelling, thickening, texturizing, and stabilizing properties, and it is mainly used in the food industry (e.g., beverages, confectionery, dairy products, and dressings) and in bacteriological and biotechnological processes (e.g., culture media) [25].
- Carrageenans and galactans are extracted polysaccharides from marine red algae with very similar characteristics. Galactans are composed of a chain of galactoses linked by (1–6) bonds with (1–3) branches [26], while carrageenans have their main chain of galactoses linked by (1–3) bonds and branches with (1–4) links [27]. Both polysaccharides are used as gelling and thickening agents in the food industry due to their rheological properties. In addition, they are important in the medical, pharmaceutical, and cosmetic areas due to their antiviral, antitumor, and anticoagulant activity [28].
- Alginate is a heteropolysaccharide that is extracted mainly from brown algae, consisting of guluronic and mannuronic acids [29]. It is characterized by its resistance and flexibility, which give it high viscosity and stability, as well as gelling properties [30]. In addition, it is valued for its antibacterial activity, biodegradability, nontoxicity, and biocompatibility. Its industrial application is directed particularly toward the generation of particles, matrix materials, encapsulants, or biocontrol agents [29].

### 2.3. Animal Polysaccharides

Animal polysaccharides are considered natural biopolymers due to their biodegradability, biocompatibility, nontoxicity, and non-antigenicity. Furthermore, these biological macromolecules play a structural and storage role in animals, since they are part of the tissues and cell matrix, and they are a source of energy. These polysaccharides can be divided into chitins and glycosaminoglycans [31,32]; the major animal polysaccharides are described below.

- Chitin is a nitrogenous polysaccharide made up of *N*-acetyl-D-glucosamine that is extracted from the external skeletons of crustaceans (e.g., crabs, lobsters, and krill), invertebrate animals (e.g., octopuses, clams, and snails), insects (e.g., scorpions, ants, and spiders), and some fungi [33]. This polysaccharide has a crystalline structure; however, its association with protein molecules produces amorphous zones, which makes its application extensive. For example, chitin is used as an enzyme immobilizer, as well as for the generation of biosensors, excipients, and drug vehicles, due to its gel-forming properties [34].
- When chitin reaches 50% deacetylation it becomes a semicrystalline material called chitosan. This polymer is the only one with pseudo-natural cationic characteristics [34]. In addition, the structural and chemical change of chitin to chitosan makes it totally soluble, biodegradable, biocompatible, and antimicrobial; thus, its application varies a little with respect to chitin. Studies have demonstrated its use as a flocculant, purifier, gel former, carrier, and microbial biocontrol agent [35].
- Hyaluronic acid is made up of disaccharides of *N*-acetylglucosamine and glucuronic acid linked by (1–3) and (1–4) bonds, and it is found in the body tissues and fluids of vertebrate animals and some bacteria [36]. This polysaccharide is of commercial interest due to its antigenic potential and viscoelastic properties; in particular, the pharmaceutical, dermatological, and cosmetic industries use it as a preservative, healing, and anti-wrinkle agent [37].

### 2.4. Bacterial Polysaccharides

Bacterial polysaccharides are natural biopolymers made up of monosaccharide chains, which, depending on the type of chain, have rheological, biological, and physicochemical properties. These molecules are valued for their viscous, thickening, stabilizing, and gelling properties, in addition to their antitumor, anti-inflammatory, and antimicrobial activities [38]. The production of bacterial polysaccharides can be carried out in two ways, extracellularly and intracellularly, depending on the substrates and requirements of the bacteria [39]; the most commercially important bacterial polysaccharides are described below.

- Dextran is an exopolysaccharide generally synthesized by lactic acid bacteria. Its structure is made up of glucose linked by  $\alpha$ -(1–6) bonds and branches with  $\alpha$ -(1–2),  $\alpha$ -(1–3), or  $\alpha$ -(1–4) links. Commercial dextran is produced by *Leuconostoc mesenteroides* (generally recognized as safe, GRAS); therefore, its application is mainly directed toward food products (e.g., bakery and confectionery) taking advantage of its gelling, texturizing, and emulsifying properties. However, the properties of dextran are a function of the producing strain (e.g., *Lactococcus*, *Lactobacillus*, and *Streptococcus*) and the structural and physicochemical characteristics they possess [40,41].
- Gellan, commercially known as gellan gum, is a polysaccharide with a linear structure of acetylated tetrasaccharide units synthesized by *Sphingomonas elodea*. Its rheological properties give it a gelling action, which is why it is used in the formation of matrices, tissue engineering, and encapsulation [42,43].
- Xanthan, commercially known as xanthan gum, is a heteropolysaccharide of pentasaccharide units synthesized by *Xanthomonas campestris*. It is important for the textile, medical, and food industries for its stabilizing, thickening, and gelling properties [44].

It is a gum with wide applications, since its stability and characteristics depend on pH and temperature; however, its synthesis is limited by high production costs [45].

- Levan is a polymer made up of fructose linked by  $\beta$ -(2–6) bonds and branches with  $\beta$ -(2–1) links [46]. The structure and molecular weight of the polymer depend on the producing organism (e.g., *Acetobacter*, *Bacillus*, and *Pseudomonas*) and the fermentation conditions (e.g., pH, temperature, and sucrose concentration); hence, each type of levan has its own functional properties, stability, viscosity, and immunogenic activity. Its application is directed toward the food industry as a texture and flavor enhancer, prebiotic, and stabilizer; in addition, it is used as a coating for nanoparticles [47].

### 2.5. Fungal Polysaccharides

Fungal polysaccharides are found in the cell wall or are formed from energetic processes in edible fungi and yeasts [48]. They are polymeric molecules with linear and branched structures that are composed of homo- and heteropolysaccharides that can be joined by  $\beta$ -(1–3),  $\beta$ -(1–6), or  $\alpha$ -(1–3) bonds, resulting in complex structures with different characteristics. For example, the antitumor activity of these polysaccharides is known to be influenced by the spatial conformation of the molecule, the degree of branching, and the molecular mass [49]. Some fungal polysaccharides are described below.

- Elsinan is an extracellular polysaccharide made up of  $\alpha$ -(1–3) and  $\alpha$ -(1–4) linked maltotriose and maltotetraose units produced by *Elsinoe* spp. when exposed to a medium with maltose, glucose, fructose, sucrose, and starch. Elsinan is soluble in water and insoluble in organic solvents, and it exhibits high viscosity; thus, its main function is to form films [50].
- Pullulan is a linear homopolysaccharide synthesized by *Aureobasidium pullulans*; it is made up of maltotriose and maltotetraose units with  $\alpha$ -(1–3),  $\alpha$ -(1–4), and  $\alpha$ -(1–6) bonds, with a three-dimensional structure similar to maltodextrin and amylopectin [50]. In industry, it is used as a substitute for gelatin due to its rheological characteristics, with the capability of forming gels, coatings, films, and encapsulates; in addition, it is a dietary prebiotic and a stabilizer [47].
- $\beta$ -Glucan is a dietary fiber located in the cell wall of algae, bacteria, yeasts, and fungi, particularly *Saccharomyces cerevisiae*. Its structure is made up of glucose monomers linked through  $\beta$ -(1–3) glycosidic bonds in bacteria and algae,  $\beta$ -(1–3) and  $\beta$ -(1–4) bonds in bacteria, or  $\beta$ -(1–3) and  $\beta$ -(1–6) in yeasts and fungi. This polysaccharide may be soluble or insoluble in water depending on its structure and molecular weight; its applications stem from its hypocholesterolemic effect, viscosity, and resistance to acidic pH [18,51].
- Galactan is a polysaccharide made up of galactoses linked by  $\beta$ -(1–4) and sometimes  $\beta$ -(1–6) bonds [52], produced not only by fungi, but also by algae, animals, plants, and other microorganisms. Galactan is important in the pharmaceutical industry due to its antithrombotic, anticoagulant, anti-inflammatory, and antiviral activities, while the food industry uses it as a food supplement [53].

### 3. Biodegradable Films

Biodegradable films and coatings are solid matrices formed by crosslinking between polymers and additives (e.g., plasticizers and crosslinking agents) [54,55]; however, it is important to identify the difference between both. Coatings are generated directly on the product by spraying or immersion techniques, while films are pre-generated before covering the product [56] by casting, extrusion, or electrospinning techniques [57]. The main method for the generation of biodegradable films is the casting technique [58], in which a dispersion between compounds (i.e., polymers and additives) is generated with a volatile solvent, which is poured into a smooth mold and left to stand until the solvent evaporates [59].



### *Characteristics and Properties of Biodegradable Films*

Biodegradable films are an alternative that the packaging industry has targeted, particularly for food packaging. Therefore, the function of biodegradable films is not to completely replace synthetic packaging, but to mitigate the environmental impact generated by solid waste [60] and provide a benefit to food by intervening in its useful life [6,61]. Therefore, biodegradable films must meet the following requirements and characteristics so that they can be functional [58,62]:

- (1) prevent or mitigate mechanical damage,
- (2) prevent or reduce lipid oxidation,
- (3) prevent or reduce microbial spoilage,
- (4) control oxygen absorption,
- (5) generate a selective barrier to carbon dioxide and water vapor,
- (6) regulate the generation of ethylene to delay senescence,
- (7) regulate the release of food additives (e.g., antioxidants, dyes, and flavors).

Biodegradable films require at least one polymeric component, based on lipids (e.g., waxes, fatty acids, and acylglycerol), proteins (e.g., caseins, whey protein, and zeins), polysaccharides (e.g., cellulose, starch, and gums), or their mixtures [9]. In addition, additives (such as plasticizers and surfactants) to modify the intermolecular bonds between polymers can be incorporated, or even bioactive compounds with fungicide, herbicide, and antioxidant capacity (e.g., natural extracts) to inhibit the development and spread of pathogenic microorganisms [63–66]. Accordingly, the thickening, gelling, and emulsifying properties of the dispersion, the texture of the matrix during polymerization, the cohesion and assembly when dehydrated, and the organoleptic and mechanical characteristics in the final film are improved [55,67–71]. However, not all polymers have the intrinsic properties suitable for use in the production of biodegradable films [72].

Polysaccharides are the most used polymers in the formulation of packaging materials for food preservation, due to their low cost and accessibility; however, the choice of polysaccharides in the formulation of biodegradable films is preferred due to the easy modification of nature hydrophilic with additives, which is reflected in its mechanical and barrier properties [9].

The efficiency of barrier properties such as water vapor permeability (WVP), ethylene content, and oxygen level maintain control of the transfer of ambient moisture and volatile components (e.g., aromas and flavors) [73], which influence the food preservation [74], dehydration of fresh products, hydration of dry products, or oxidation of polyunsaturated fats in food [9]. On the other hand, mechanical properties such as tensile strength (TS), elongation at break (EB), and young modulus (E) are related to the integrity and brittleness of films during handling and storage [75]. Some investigations reported that mechanical properties improved with the incorporation of hydrophobic additives (e.g., glycerol or sorbitol) [76]. Physicochemical characteristics such as color, opacity, luminosity, morphology, and roughness are directly related to the type and concentration of polysaccharides, as well as the film-making method [77,78]. However, the need to understand the behavior of polysaccharides and additives to improve the properties and functionality of biodegradable films is reflected in the increasing number of reported studies, as shown in the next section.

### **4. Biodegradable Films Based on Polysaccharides**

According to the Scopus database [79], studies on biodegradable films have had an average annual increase of 13% in the last decade, highlighting a 24% increase in publications last year (2021). According to published studies [79], polysaccharides are the most widely used materials for the generation of biodegradable films, due to their affordable cost, variety of extraction sources, and functional properties. Current studies that have characterized biodegradable films from different sources (i.e., plants, animals, bacteria, algae, and fungi) are grouped in Table 1. When analyzing the mechanical and barrier properties, different results can be observed, apparently without any relationship;

however, each study followed its own methodology where the type and concentration of polysaccharides or additives may have varied.

**Table 1.** Properties of biodegradable films based on polysaccharides.

Polysaccharide(s) or Derivate(s)	Plasticizer(s) or Additive(s)	Mechanical Properties			Barrier Properties	Reference
		TS (MPa)	E (MPa)	EB (%)	WVP (10 <sup>−10</sup> g·m <sup>−1</sup> ·s <sup>−1</sup> ·Pa <sup>−1</sup> )	
Plant polysaccharide(s)						
Acid alcohol sorghum starch	Glycerol	~1.0–1.4	nr	~5.0–6.0	nr	[80]
Acetylated sorghum starch	Glycerol	~0.2–2.8	nr	~4.0–19.0	nr	[80]
Basil seed gum	Glycerol	21.3	nr	25.9	237.0	[81]
Basil seed gum modified	Glycerol	26.5–46.4	nr	23.8–36.9	120.0–215.0	[81]
Butylated hemicelluloses	nr	~40.0–50.0	~40.0–55.0	~9.0–10.0	nr	[82]
Cassava starch	Glycerol	2.4	51.8	43.8	2.8	[83]
Carboxymethyl cellulose	Glycerol	32.3	nr	35.6	861.7	[84]
Carboxymethyl cellulose	Glycerol	20.0	nr	30.4	~2830.0	[85]
Carboxymethyl cellulose	nr	~37.5	nr	~26.0	~3.4	[86]
Carboxymethyl cellulose/cellulose nanofiber	Glycerol	20.2–23.1	nr	22.8–46.6	1083.3–1421.7	[84]
Carboxymethyl cellulose/inulin	Glycerol	7.3–21.9	nr	20.2–37.8	935.0–1125.0	[84]
Carboxymethyl cellulose/cellulose nanofiber/inulin	Glycerol	16.4–23.8	nr	34.6–41.3	978.3–1540.0	[84]
Carrot flour	nr	~3.0	<0.1 *	<0.1	nr	[87]
Carrot flour/hydroxypropyl methylcellulose	nr	~3.0–7.0	~0.4–0.6 *	~1.0–2.0	~0.4–0.7	[87]
Cellulose regenerated	nr	~70.0	~98.0	~6.0	nr	[82]
Cellulose acetate	nr	~80.0	~80.0	~22.5	nr	[82]
Cellulose carbamate	Glycerol	~45.0	~80.0	~10.0	nr	[82]
Cellulose palmitate	nr	~10.0	~10.0	~27.5	nr	[82]
Cellulose octanoate	nr	~10.0	~10.0	~117.5	nr	[82]
Corn/octenylsuccinated starch	Glycerol	4.4	nr	45.7	2.9	[88]
Guar gum	Glycerol	5.3	8.9	64.8	38.6	[89]
Guar gum	Tween-20	41.9 *	nr	1.8	nr	[90]
Guar gum	Glycerol	18.0	46.7 *	31.6	nr	[91]
Guar gum/potato starch	Glycerol	8.3	nr	8.6	<0.1	[92]
Hydrolyzed achira starch	Glycerol	9.5	388.1	24.1	2.1	[93]
Hydrolyzed-succinated achira starch	Glycerol	10.5	513.4	42.8	3.2	[93]
Hydrothermal sorghum starch	Glycerol	~0.7–6.5	nr	~7.0–14.0	nr	[80]
Hydroxypropyl methylcellulose	nr	~67.0	~1.8 *	~14.0	~0.2	[87]

Table 1. Cont.

Polysaccharide(s) or Derivate(s)	Plasticizer(s) or Additive(s)	Mechanical Properties			Barrier Properties	Reference
		TS (MPa)	E (MPa)	EB (%)	WVP ( $10^{-10} \text{ g}\cdot\text{m}^{-1}\cdot\text{s}^{-1}\cdot\text{Pa}^{-1}$ )	
Karaya gum	Glycerol	6.5	nr	8.0	58.5	[94]
<i>Lemang bamboo</i> microcrystalline cellulose	Glycerol	17.2–41.9	129.0–253.0	8.7–27.1	2.5–4.9	[95]
Methylated guar gum	Glycerol	3.4	7.2 *	40.1	nr	[96]
Methylcellulose	nr	~70.0	~80.0	~17.5	nr	[82]
Methylated guar gum	Glycerol	5.6–11.3	25.0–47.4 *	36.0–49.4	nr	[91]
Microcrystalline cellulose	Glycerol	13.7–35.7	116.0–206.0	18.0–23.1	3.0–5.4	[95]
Native achira starch	Glycerol	1.1	532.7	5.9	2.7	[93]
Native potato starch	Glycerol	3.9	45.1	81.0	2.4	[97]
Native sorghum starch	Glycerol	~0.5–4.2	nr	~2.0–10.5	nr	[80]
Nanocellulose	nr	~110.0	~115.0	~12.5	nr	[82]
Ozonated potato starch	Glycerol	3.3–4.2	61.1–64.1	19.2–28.4	2.6–3.0	[97]
Ozonated cassava starch	Glycerol	3.8–5.5	71.2–82.0	37.4–39.5	3.1–3.5	[83]
Pectin	nr	6.8–7.3	27.2–33.6	18.9–21.8	nr	[98]
Pectin	Glycerol	9.6	nr	14.5	6180.0	[99]
Pectin	Sorbitol	50.0	nr	18.0	4020.0	[99]
Pectin	Glycerol	14.2–18.4	nr	0.6–1.3	14.5–15.5	[100]
Pectin	Natural deep eutectic solvent	10.7–14.3	nr	0.5–1.2	22.0–24.8	[100]
Pectin	Choline chloride	3.0–10.5	nr	0.5–0.8	18.3–27.1	[100]
Pectin/gelatin	Glycerol	16.9	132.4	73.0	nr	[101]
Pectin/potato starch	nr	22.3	305.4	13.0	2.8	[102]
Persian gum	Glycerol	<0.1	nr	~45.0–50.0	130.0–150.0	[103]
Persian gum	Glycerol	~0.8–16.0	~5.0–375.0	~3.0–60.0	nr	[104]
Potato waste starch	Glycerol	~3.0–17.0	nr	~4.0–12.0	nr	[105]
Potato waste starch	Sorbitol	~4.0–24.0	nr	~2.0–9.0	nr	[105]
Potato starch	nr	5.1	nr	33.7	nr	[106]
Succinated achira starch	Glycerol	7.5	321.9	82.3	2.6	[93]
<i>Salvia macrosiphon</i> seed gum	Glycerol	4.2	nr	39.1	~50.0	[107]
<i>Semantan bamboo</i> microcrystalline cellulose	Glycerol	25.8–43.1	74.0–152.0	6.6–22.1	2.2–4.6	[95]
Tragacanth gum	nr	~11.0	nr	~1.0	nr	[108]
Tragacanth gum/polyvinyl alcohol	nr	~12.0–15.0	nr	~5.0–7.5	nr	[108]



Table 1. Cont.

Polysaccharide(s) or Derivate(s)	Plasticizer(s) or Additive(s)	Mechanical Properties			Barrier Properties	Reference
		TS (MPa)	E (MPa)	EB (%)	WVP (10 <sup>−10</sup> g·m <sup>−1</sup> ·s <sup>−1</sup> ·Pa <sup>−1</sup> )	
Algae polysaccharide(s)						
Agar	nr	~900.0	~30.0 *	~8.5	nr	[109]
Agar	Glycerol	~500.0–650.0	~16.2–22.5 *	~15.0–19.0	nr	[109]
Agar	Glycerol	22.0	1.0 *	8.3	3.17	[110]
Agar	Glycerol	34.9	1.2 *	12.0	10.7	[111]
Agar	Glycerol	34.8	1.2 *	11.8	11.6	[112]
Agar	Glycerol	28.0–55.5	nr	13.0–27.5	0.8–0.9	[113]
Agar	Glycerol	47.3	nr	14.0	nr	[114]
Agar	Polyglycerol	0.4	278.5	18.3	nr	[115]
Agar	Glycerol	18.7	361.7	29.9	93.6	[116]
Agar	Glycerol	40.3	1.4 *	19.4	19.9	[117]
Agar/alginate	Glycerol	45.2	nr	33.0	~0.9	[118]
Agar/carboxymethyl cellulose	Glycerol	60.4	2.0 *	14.7	22.1	[119]
Agar/carboxymethyl cellulose	Glycerol	44.9	1.6 *	16.0	7.1	[120]
Agar/ <i>k</i> -carrageenan	Glycerol	45.4	3.0 *	2.5	6.6	[121]
Agar/carboxymethyl cellulose/cellulose nanocrystals	Glycerol	57.5	2.6 *	10.0	8.1	[120]
Agar/chitosan	Glycerol	~35.0–48.0	nr	~15.0–21.0	nr	[122]
Agar/gellan gum	Glycerol	29.9	nr	29.5	19.0	[123]
Agar/gellan gum/montmorillonite	Glycerol	35.3–44.0	nr	19.9–24.1	16.6–18.1	[123]
Agar/gelatin	Glycerol	3.7–13.6	21.6–186.8	38.9–45.17	145.5–201.0	[116]
Agar/konjac glucomannan	Glycerol	~35.0–47.0	nr	~20.0–38.0	nr	[124]
Agar/lignin	Glycerol	44.1	1.5 *	16.1	18.5	[117]
Agar/pectin	Glycerol	50.3	2.3 *	4.7	4.8	[125]
Agar/pullulan	Glycerol	23.8	nr	37.2	31.7	[126]
Agar/pullulan/montmorillonite	Glycerol	31.4–37.1	nr	28.2–35.2	27.2–30.4	[126]
Agar/pullulan/montmorillonite/quaternary ammonium silane	Glycerol	39.7	nr	26.9	22.0	[126]
Agar/nano cellulose	Glycerol	22.1	nr	10.8	0.9	[113]
<i>k</i> -Carrageenan	Glycerol	44.6	1.5 *	11.0	16.2	[127]
<i>k</i> -Carrageenan	Glycerol	65.9	2.9 *	4.4	18.0	[128]
<i>k</i> -Carrageenan	Glycerol	54.9	2.7 *	8.1	16.7	[129]
<i>k</i> -Carrageenan	Glycerol	17.0–19.1	nr	29.5–63.8	1.2–2.8	[130]

Table 1. Cont.

Polysaccharide(s) or Derivate(s)	Plasticizer(s) or Additive(s)	Mechanical Properties			Barrier Properties	Reference
		TS (MPa)	E (MPa)	EB (%)	WVP ( $10^{-10} \text{ g} \cdot \text{m}^{-1} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}$ )	
<i>k</i> -Carrageenan	Glycerol	19.2	59.6	4.4	382.0	[131]
<i>k</i> -Carrageenan	Glycerol	43.3	1.5 *	11.2	16.6	[132]
<i>k</i> -Carrageenan	Glycerol	10.0	nr	29.8	0.4	[133]
<i>k</i> -Carrageenan	Glycerol	11.8	40.5	29.2	0.7	[134]
<i>k</i> -Carrageenan	Glycerol	57.0	3.3 *	4.4	17.2	[135]
<i>k</i> -Carrageenan	Glycerol	38.3	nr	21.5	0.9	[136]
<i>k</i> -Carrageenan	Glycerol	22.6	nr	14.5	0.8	[137]
<i>k</i> -Carrageenan	Glycerol	~49.0	~1.5 *	~85.0	nr	[138]
<i>k</i> -Carrageenan	Glycerol	57.0	3.3 *	4.4	17.2	[139]
<i>k</i> -Carrageenan	Glycerol	7.4	nr	32.0	1.5	[140]
<i>k</i> -Carrageenan	Glycerol/ Tween-20	7.4–11.5	nr	19.1–43.6	1.3–1.5	[140]
<i>k</i> -Carrageenan	Glycerol/ Tween-40	4.0–9.4	nr	37.4–49.8	1.6–2.1	[140]
<i>k</i> -Carrageenan	Glycerol/ Tween-80	8.5–12.6	nr	23.5–40.6	1.3–1.5	[140]
<i>k</i> -Carrageenan	Glycerol	37.7–54.4	nr	56.9–80.7	120.1–142.1	[141]
<i>k</i> -Carrageenan/cassava starch	Glycerol	12.2–25.9	4.8–27.0	8.4–26.4	301.0–448.0	[131]
<i>k</i> -Carrageenan/cellulose nanocrystals	Glycerol	38.4–52.7	nr	22.9–28.3	0.5–0.9	[136]
<i>k</i> -Carrageenan/cellulose nanocrystals	Glycerol	~59.0–85.0	~1.7–2.7 *	~67.0–77.0	nr	[138]
<i>k</i> -Carrageenan/pullulan	Glycerol	54.0	3.4 *	2.7	10.0	[142]
<i>k</i> -Carrageenan/nanoclay	Glycerol	~16.0	nr	~20.0	4.0	[143]
Gelatin	Glycerol	6.17	15.0	40.9	121.8	[144]
Gelatin	Glycerol	1.9	nr	91.7	6.4	[99]
Gelatin	Sorbitol	5.8	nr	93.3	2.1	[99]
Gelatin	Glycerol	18.7	103.1	117.1	~5.5	[145]
Gelatin	Glycerol	1.8	9.1	76.7	248.3	[116]
Gelatin/agar	Glycerol	69.1	2.4 *	8.6	5.9	[146]
Gelatin/ <i>k</i> -carrageenan	Glycerol	43.9	3.5 *	3.2	6.1	[147]
Gelatin/cress seed gum/chitosan nanoparticles	Glycerol	7.8–9.4	23.2–45.5	19.7–34.1	109.0–179.3	[144]
Animal, bacterial, and fungal polysaccharide(s)						
Chitosan	Glycerol/Calcium chloride	1.1	2.8	38.5	601.7	[148]
Chitosan/dextran	Glycerol/Calcium chloride	1.5–2–5	1.7–6.5	36.2–87.7	655.0–993.3	[148]

Table 1. Cont.

Polysaccharide(s) or Derivate(s)	Plasticizer(s) or Additive(s)	Mechanical Properties			Barrier Properties	Reference
		TS (MPa)	E (MPa)	EB (%)	WVP ( $10^{-10} \text{ g} \cdot \text{m}^{-1} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}$ )	
Chitosan	Glycerol	~17.5	~775.0 *	~5.0	nr	[149]
Chitosan	Glycerol	23.5	nr	33.4	1.7	[150]
Chitosan	Glycerol/Tween-80	14.2–37.7	nr	15.2–59.2	2.2–3.1	[150]
Dextran	Sorbitol	0.7–7.5	9.1–1755.0	1.0–134.5	<0.2	[151]
Gellan gum	Glycerol	nr	~6.5 *	~1.0	nr	[152]
Gellan gum	Glycerol	2.5 *	nr	55.8	<0.1	[153]
Gellan gum/cassava starch	Glycerol	24.0–40.0	1.3 *	2.1–4.9	17.2–18.6	[154]
Gellan gum/guar gum	Glycerol	2.9–3.0 *	nr	61.2–68.0	<0.1	[153]
Gellan gum/pectin	Glycerol	27.0	0.3 *	41.3	1.6	[155]
Gellan gum/poly( $\gamma$ -glutamic acid)	Glycerol	~5.5–13.0	~10.0–50.0	~1.0–2.5	nr	[152]
Gellan gum/xanthan gum	Glycerol	22.1	nr	30.0	38.3	[156]
Hyaluronic acid/corn starch	Glycerol	6.3	nr	41.7	nr	[157]
Xanthan gum	Glycerol	~17.0	nr	~20.0	nr	[153]
Xanthan gum	Glycerol	7.4–8.7	54.3–68.7	6.3–14.6	2.6–3.7	[158]
Xanthan gum/cassava starch	Glycerol	14.0	0.7 *	3.0	31.7	[159]
Xanthan gum/cassava starch	Glycerol	4.0–10.0	36.0–160.0	12.0–34.0	7.1–14.0	[160]
Xanthan gum/curdlan gum	Glycerol	27.8	nr	12.9	nr	[161]
Xanthan gum/curdlan gum/gelatin	Glycerol	30.3–38.2	nr	14.4–18.9	nr	[161]
Xanthan gum/curdlan gum	Glycerol	~18.0–27.5	nr	~2.5–17.5	nr	[153]
Xanthan gum/maize starch	Glycerol	9.0	0.5 *	11.0	37.2	[159]

nr: not reported. \* Data represented in units of GPa.

#### 4.1. Biodegradable Films Based on Starches

Starch-based biodegradable films are the most studied because they are considered isotropic, colorless, tasteless, odorless, nontoxic, and biologically degradable [162]; however, the hydrophilic nature of starch requires proper formulation to generate functional films (i.e., mechanical strength) with control over water content and WVP [15]. However, the source of production has an influence on the mechanical and barrier properties, due to the fact that, by their nature, starch films have good WVP, but mechanical properties depend on the crystallinity of the starch used [9].

For example, La Fuente et al. [83], Shrestha and Dhungana [80], and Sama and Yuliasih [106] evaluated biodegradable films based on cassava, sorghum, and potato starch, respectively. Although the results did not show a trend in TS, E, EB, and WVP, the cassava starch-based films had the highest E and EB results, while the potato starch films stood out for presenting higher values of TS. This variation can be seen more clearly in the studies by Cabrera Canales et al. [93] and La Fuente et al. [97], who evaluated native starch from two sources (achira and potato, respectively), and the results showed significant differences in the mechanical properties; however, the barrier properties were numerically similar. According to Horstmann et al. [163], the composition of starch was varied in terms of proportion of amylose and amylopectin and their branches, proteins, lipids, ash, and moisture content; this approach generated variations in morphology, granule size, solubility, and

functional properties (e.g., gelatinization and pasting). Therefore, any variation in the composition of the starch positively or negatively affects its properties.

The source from which the starch is obtained is not the only variable that influences the mechanical and barrier properties. There are studies [164,165] showing that the addition of plasticizers and surfactants such as glycerol, sorbitol, and Tween generates changes in the three-dimensional networks that allow the formation of biodegradable films, affecting their hydrophobicity, permeability, and mechanical resistance. The study by Karki et al. [105] evaluated biodegradable films of potato waste starch with glycerol and sorbitol, and they reported significant variation in the mechanical properties; sorbitol improved the TS of the films, while glycerol benefited EB. Previous studies conducted comprehensive evaluations of plasticizers and their effect on biodegradable films. For example, Sanyang et al. [166] noted that the mechanical properties in sugar palm starch films varied according to the type of plasticizer (i.e., glycerol, sorbitol, or mixture of both). For example, films plasticized with sorbitol presented higher TS and films plasticized with glycerol presented higher EB percentage; however, both plasticizers had an anti-plasticizer effect, which was previously reported when used in high concentrations, particularly in starch films [167]. While the biodegradable films plasticized with glycerol and a glycerol–sorbitol mixture presented similar WVP results, films plasticized with sorbitol presented the lowest WVP of the three, in direct relation to concentration [166]. Specifically, a lower concentration of sorbitol led to a lower WVP due to the interactions generated because of the structure and compactness of the polymer matrix [168].

It has been shown that starches can be physically or chemically modified to improve their film-forming properties [83], in such a way that the acetylation, acid–alcohol hydrolysis, ozone, or hydrothermal methods can be used to reorganize the molecular structure, modify the gelatinization temperature, or improve the solubility of starches [169,170]. Shrestha et al. [80] evaluated biodegradable films of native and modified sorghum starch in three different ways (i.e., hydrothermal treatment, acetylation, and acid–alcohol hydrolysis). In general terms, the hydrothermal and acetylated films improved the mechanical properties of the native starch films; that is, the hydrothermal film had the highest TS values, while the acetylated film had the highest EB percentage of all films. These results were related to the alterations suffered by the starches when they were modified; the improvement of the acetylated films was related to the increase in the degree of solubility and swelling power, whereas the hydrothermal treatment increased the water binding capacity [80].

Cabrera Canales et al. [93] used other types of treatments such as hydrolyzation, succination, or a mixture of both to modify achira starch and generate different biodegradable films. The study showed that the modified starch films improved the TS and the EB compared to the native starch film, which was related to the increase in crystalline areas due to the effect of chemical modifications of starches that caused greater matrix mobility [93]. However, in the modified films, a decrease in E was observed as a consequence of the generation of short chains (by hydrolysis), the remaining granules (by succination), or the little interaction with hydroxyl groups (both processes) between each starch [171–173]. Furthermore, the WVP had significant variations since the hydrolyzed films presented lower values and the hydrolyzed–succinated films had higher values, since the water adsorption, crystallinity, and interstitial spaces depended on the interactions generated between the modified chains of each starch and the plasticizing agent [174,175].

#### 4.2. Biodegradable Films Based on Celluloses and Derivates

Celluloses are used in various food packaging materials because they are versatile, low-cost, and nontoxic, and they have excellent mechanical properties. For example, cellophane is the most widely used cellulose derivative in packaging; however, its formulation with synthetic materials classifies it as a highly polluting material [176]. There are other natural derivatives of cellulose that are used in the formulation of biodegradable films, such as carboxymethyl cellulose, cellulose regenerate, and some cellulose esters (e.g., cellulose acetate, palmitate, and octanoate). Films generated with cellulose or their derivatives

are flexible and moderately resistant, with efficient WVP; however, these properties vary depending on the hydrophobicity and crystallinity modification caused by the incorporated additives [9].

For example, Leppänen et al. [82] evaluated different types of celluloses in the formation of biodegradable films and found that the mechanical properties varied greatly. In general, regenerated cellulose films had the highest E values but the lowest EB values, while cellulose octanoate films had the highest EB values and the lowest E values. These results showed the great influence of the methodologies used to generate biodegradable films, since each one was made in a different way; however, it was also shown that the addition of a plasticizer such as glycerol did not improve the mechanical properties.

Zabihollahi et al. [84], Peighambardoust et al. [85], and Rincón et al. [86] evaluated biodegradable films of carboxymethyl cellulose. Analyzing the WVP results, it is evident that the Peighambardoust et al. [85] presented the highest value, while the biodegradable films of Rincón et al. [86] presented the lowest value; however, it can be noted that the results of the mechanical properties were similar, despite the fact that the methodologies, proportion of polysaccharide, and addition of plasticizer were totally different. These findings suggest that none of these variables is totally influential with respect to the resistance and versatility of the biodegradable cellulose films; even the films with glycerol resulted in lower TS and higher EB than the films that did not contain the plasticizer. This effect was reported by Hidayati et al. [177], who evaluated the effect of glycerol on carboxymethyl cellulose films and noted that, as the concentration increased, the TS decreased while the EB increased. The authors [177] established that the hydrophilic properties of glycerol decreased the intermolecular force of the carboxymethyl cellulose molecules, which allowed the matrix to elongate (higher EB) but, as a result, become more susceptible to rupture by an external force (TS). It has been reported [178,179] that the combination of carboxymethyl cellulose with other polysaccharides (e.g., chitosan and starch) can increase the TS of biodegradable films by increasing their concentration; however, EB and WVP tend to decrease. This is justified by the modification of the crystallinity of the pure matrix (a single polysaccharide), which, when mixed, increases its amorphous zones due to the disorganization of the chains, making them resistant, due to the increase in the interaction between functional groups (e.g., hydroxyl and carboxyl groups), but not very flexible, due to the lack of mobility of the chains [178].

#### 4.3. Biodegradable Films Based on Pectins

Pectin-based biodegradable films are the natural material proposals that have presented the most disadvantages, due to their fragile nature and poor WVP and mechanical properties; however, these films have a positive influence with the addition of plasticizing agents [100]. For example, Shafie et al. [98] and Gouveia et al. [100] generated biodegradable pectin films; in the first study, no plasticizer was used, while, in the second study, glycerol was evaluated. The films with glycerol showed an evident increase in ST, but a decrease in EB. Such an effect is contrary to what happens to cellulose films with the addition of glycerol (Section 4.2); however, it is not a recurrent result in pectin films. In previous studies [180], the effect of increasing glycerol concentration in pectin films was evaluated, and the results were consistent with a decrease in TS and an increase in EB (as in cellulose-based biodegradable films).

In addition, the studies by Aitboulahsen et al. [99] and Gouveia et al. [100] tested various plasticizers to evaluate the variation in mechanical and permeability properties. The results showed that sorbitol was able to increase the TS and GE of the biodegradable pectin films, compared to glycerol, natural deep eutectic solvent, and choline chloride; however, the positive effect of sorbitol on the mechanical properties is not specific to biodegradable pectin films, as the study by Ballesteros-Mártinez et al. [181] also reported the highest TS values in sweet potato starch films. The authors [181] suggested that, in the first instance, any plasticizer can modify the density of the film; that is, as the plasticizer is incorporated, the matrix becomes less dense, which causes an increase in the movement of



the chains and, therefore, their elasticity (EB). On the other hand, the results of the ST are related to the interaction of the plasticizer with the polymeric chains; glycerol, having a smaller molecular size, more easily establishes itself in the intermolecular spaces, whereas sorbitol maintains bonds with the polysaccharide surface while avoiding interference with the hydrogen bonds that must be formed to generate resistance [182].

The studies by Jovanović et al. [101] Dash et al. [102] showed that the incorporation of a second polysaccharide in the formulation of pectin-based films also had a positive effect on the mechanical properties and the WVP; in addition, the incorporation of a plasticizer into the mixture improved the TS and the E, which, as in cellulose films, was due to the increase in the amorphous zones of the polymer matrix and the intermolecular interaction between glycerol and this matrix [178].

#### 4.4. Biodegradable Films Based on Gums

Gums are defined as hydrocolloids due to their properties as emulsifiers, stabilizers, texturizers, thickeners, coating agents, and film generators; however, their potential application depends on the type of gum and its source [183]. The gums most used as film-forming agents are guar, gellan, xanthan, Persian, konjac glucomannan, and Arabic gums; however, the study carried out by Pedreiro et al. [184] established that their main application is as a coating (see Section 3) applied directly (by immersion or dipping) on the surface of foods (e.g., tomatoes, guavas, mangoes, and mushrooms) Because they are GRAS, they can perfectly incorporate bioactive compounds (e.g., extracts or antimicrobial agents) and extend the shelf life of products [184].

When comparing the studies that reported the use of gums in the formulation of biodegradable films shown in Table 1, the values of some mechanical properties were lower compared to those reported for biodegradable films of starches and cellulose. Reports have suggested that, for the WVP and mechanical properties to be improved in gum-based films, additives must be incorporated [9].

Kirtil et al. [89] reported lower values of TS, E, and EB in their biodegradable guar gum films compared to the values reported by Leppänen et al. [82] for their cellulose octanoate-based films. However, the variation in the plasticizing agents or the incorporation of a second polysaccharide in the formulation considerably improved the mechanical properties; for example, the biodegradable films of tragacanth gum reported by Janani et al. [108] had increased TS values up to 36% and increased EB values up to 650% upon adding polyvinyl alcohol as a second film former. Mohsin et al. [153] elaborated composite films of guar and gellan gums, while Gao et al. [90] varied the plasticizing agent (Tween-20) in their guar gum films; in both studies, the TS results were reported higher than any starch film in Table 1. In addition, the study by Zhu et al. [161] showed that the incorporation of gelatin as a third polysaccharide in films of xanthan gum–curdlan gum increased TS and EB by up to 43%, however, to obtain a significant improvement, the proportion of gelatin had to be up to 90%.

On the other hand, the studies in Table 1 also showed that gum-based biodegradable films generally maintain the lowest WVP values, which is important for the application of the films in food models. During the respiration processes of natural foods such as fruits and vegetables, sugars undergo an oxidation process in the presence of oxygen, resulting in the release of carbon dioxide, water, and, in some cases, ethylene [185]. In the application of biodegradable films, a WVP is sought (depending on the respiration rate of the product) that ensures that water vapor will be released to delay the maturation of the product [186].

#### 4.5. Biodegradable Films Based on Agars

Agar is easy to extract, safe, and cheap, making it a profitable polysaccharide for biodegradable film formation [187]. The formation of the agar matrix results from the intermolecular interaction of hydrogens of agarose with water molecules; in addition, its properties allow it to interact with bioactive substances such as antimicrobials [188]. According to Mostafavi and Zaeim [189], the greatest limitations of biodegradable agar films

are their fragility, low elasticity, high solubility, low thermal stability, and high permeability; therefore, one solution is to incorporate other polysaccharides and plasticizing agents. However, the studies in Table 1 showed that, in general, the mechanical (i.e., TS and EB) and barrier properties are relatively low (compared to starch films). Furthermore, the incorporation of plasticizers does not significantly increase the values. For example, Campa-Siqueiros et al. [109] developed agar films with high TS and E values but low E values; however, upon adding glycerol in the formulation, the values of TS and E decreased.

It is important to highlight that the studies reported the highest values of E; however, the addition of glycerol, polyglycol, or different polysaccharides (e.g., alginate, carboxymethyl cellulose, and gellan gum) did not generate a linear variation in the values. Therefore, the results depended on the interactions generated during the formation of the matrix.

## 5. Concluding Remarks

Packaging made from biodegradable materials or obtained from renewable sources, such as biodegradable films, have become a viable alternative in product packaging. Biodegradable films based on polysaccharides and plasticizers can assume the role of a primary container and even act as a bioactive container if some antioxidant and antimicrobial components or extracts with some functional activities are incorporated. The initial idea of the knowledge of the mechanical and barrier properties results in the application to biodegradable films. Ideally, the film should be resistant enough for handling during the packaging processes; in addition, the WVP defines the product to which it can be applied. For example, in products with a high respiration rate, it is required that the WVP be high so that the steam generated by the post-harvest metabolism of the food is released into the environment to delay maturation. On the other hand, the characteristics of polysaccharides influence their availability to interact with bioactive components, in such a way that the generated biodegradable films can serve as antimicrobial barriers while delaying the degradation of highly perishable products.

Lastly, it can be said that biodegradable polysaccharide-based films have unique characteristics depending on the polymeric materials, plasticizers, bioactive agents, concentrations, and formation techniques; therefore, the incorporation or mixture of these variables results in multiple options that can serve as packaging even for the same food. However, although some polysaccharide-based films are marketed, packaging based on synthetic or hybrid materials (mixtures of synthetic and biodegradable materials) continue to lead the market; therefore, biodegradable films continue to be an attractive area for further exploration and contribution.

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