



Review

Natural Polymers Used in Edible Food Packaging—History, Function and Application Trends as a Sustainable Alternative to Synthetic Plastic

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Abstract: In this review, a historical perspective, functional and application trends of natural polymers used to the development of edible food packaging were presented and discussed. Polysaccharides and proteins, i.e., alginate; carrageenan; chitosan; starch; pea protein, were considered. These natural polymers are important materials obtained from renewable plant, algae and animal sources, as well as from agroindustrial residues. Historically, some of them have been widely used by ancient populations for food packaging until these were replaced by petroleum-based plastic materials after World War II. Nowadays, biobased materials for food packaging have attracted attention. Their use was boosted especially because of the environmental pollution caused by inappropriate disposal of plastic packaging. Biobased materials are welcome to the design of food packaging because they possess many advantages, such as biodegradability, biocompatibility and low toxicity. Depending on the formulation, certain biopolymer-based packaging may present good barrier properties, antimicrobial and antioxidant activities. Thus, polysaccharides and proteins can be combined to form diverse composite films with improved mechanical and biological behaviors, making them suitable for packaging of different food products.

Keywords: food packaging; biobased materials; biodegradable packaging; edible films; composite films; alginate; carrageenan; chitosan; starch; pea protein



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1. Introduction

Plants, algae and marine seafoods are important resources to obtain natural polymers that can be used for designing sustainable packaging. Historically, different natural materials, e.g., plant and animal parts, shells, ceramics, and others, were the firstly used materials by the ancient populations for food packaging [1]. The Industrial Revolution and the World War II were significant events that boosted the discovery, design of new packaging materials and novel processing strategies. Aluminum cans by canning operation and the coming up of petroleum-based plastics are some typical examples [2,3]. Afterwards, conventional plastics were extensively used for packaging. As their majority has a single use only, usually ends up polluting landsites and aquatic environments. It was estimated that, only in 2015, around five billion tons of plastic waste ended up in natural sites [4]. Because of this problem, reducing petroleum-based pollution should be top priority of all nations around the world. More than 170 United Nations Member States have committed to reduce the use of plastics until 2030 [4]. In this way, replacing conventional plastics for biobased materials is a sustainable approach that contributes to reducing plastic pollution. Consequently, the synthesis and use of biobased materials for packaging design has increased over the past few years.

Besides being environmentally friendly and emitting low greenhouse gases, natural materials can be obtained from different renewable sources, such as plants, animals, microorganisms and their residues, making them extensively available [5]. This is also an advantage when compared to conventional plastic polymers, since the latter are petroleum-based, a nonrenewable resource. Natural polymeric materials, such as plant- or marine-based polysaccharides and proteins, are naturally biodegradable, which means that they can be degraded by local microorganisms and return to nature [6]. Natural polymers have important technological and biological properties, which allow their utilization for food packaging. Generally, these materials display good film-forming and gelling properties, which leads to consider them distinguishable and sustainable choices to prepare edible films and coatings. When applied to this end, biopolymeric films may exhibit some processing challenges. They present brittleness, low water and gases barrier properties, inferior mechanical behavior, characteristic of their nature [7]. The incorporation of hydrophobic substances or blending two or more biopolymers to form composites consist of alternative approaches to overcome these technological issues [8].

In this context, this review aims to summarize a historical perspective, technological trends and physicochemical properties of five natural materials, as alginates, carrageenan, chitosan, starch and pea protein, used to the design of edible films and coatings. Two platforms, Web of Science® and Google Scholar®, were used to search published articles within the last 10 years, although some older references may appear, mainly linked to the historical point of view of food packaging and these natural polymers.

2. Historical Perspective of Food Packaging

It has been exceedingly difficult to study the firstly used materials for packaging food supplies, since so little of them have survived along the centuries. These primitive packaging used by the ancient populations thousands of centuries ago were made from natural materials, such as wooden parts, bamboo, shells, animal skin, leaves, grasses, jute, glass, and ceramic [1,9–11]. Some evidence of primitive pottery and glass used as packaging is dated around 7000 B.C. [9,10].

When visiting museums, is common to find ancient ceramic jars named amphoras, and potteries made from glass, which were frequently used in Mediterranean region as packaging for different goods. Some of these, dating about 1500 B.C. to 500 A.D., were used to transport and commercial trade of wine, oil, grains and other foods by Egyptians, Greek and Roman civilizations [10,11]. On the other side of the Atlantic Ocean, Latin-American indigenous populations also had used ceramic materials to prepare and dish their foods during ritual events, dating around 1000 B.C. [12].

Limestone, sand, soda, and silica were the basic materials molded and used to make glass potteries, cups and bowls in the 1200 B.C. [10,13]. These ingredients have not changed in the glass-making process since their discovery. However, molding evolved with the invention of the blowpipe by Phoenicians in 300 B.C., who improved the glass production to make round vessels [13]. It was only in the 17–18th century that the split molding process was developed, allowing the fabrication of irregular shapes and different decorations on glass packaging [13]. From this period until the early 1900s, glass vessels with a wide range of shapes and sizes gained the consumers attention and dominated the packaging market up to the 1960s [13]. Even with the discovery and use of metals for different purposes during the Iron Age, 500–332 B.C., their use as a packaging material was not common until the development of stronger alloys and thinner coatings by the process of tin plating in 1200 A.D. [13].

The major packaging evolution occurred during the Industrial Revolution in the 18–19th centuries. During this period, an event stamps the beginning of the Modern Packaging Era, the discovery of canning operation by Nicholas Appert, who used this method to preserve food supplied to the French Army of Napoleon Bonaparte [2,3,10]. The Appert's method allowed that food products were heated during processing, consequently improving their shelf-life and preserving it against microbial contamination [10]. This

technology was quickly employed in many places, especially in the USA, where it was used in a continuous production in the early 1900s [3].

The development of aluminium cans, which years later was to be used for beer and other drinking food, resulted from the tinsplate shortage during World War II (WWII) [3]. Several of these technologies were especially designed to sustain a safe food supply to the armies during WWII and are used until today. In the 19th century, different materials, such as paper and paperboard, were continuously improved, folded into smaller packages, cartons, and corrugated boxes and used for food packaging [10]. Additionally, the first plastics derived from cellulose and petroleum were discovered in the 1800s [2,9]. The petroleum-based plastics were firstly employed to protect munition during WWII and then became packages used for cereals and biscuits [2]. The development of various polymers in the early 1900s, such as polypropylene, polyester, and ethylene vinyl alcohol, pushed the plastic packaging industry increasingly forward letting the metal, glass, and paperboard industries [2,10].

The use of edible materials as food coating has also few historical reports. Back in the ancient age, edible packaging was used in the production of sausages, preserving meat by stuffing it in animal intestines, a process developed by Sumerians in Mesopotamia around 3000 B.C., and by Chinese settlers in China around 580 B.C. [14]. A particular type of edible thin film made from the skin of boiled soy milk was used in Japan during the 15th century to cover different foods [15]. In China and Europe, during the 16th century animal grease (lard) and wax were used to coat fruits and other food products for their conservation and later consumption [9,15]. And later, back on the 19th century, a coating made from gelatin was patented by USA and used in meat products [15]. As synthetic plastics dominated the packaging market after WWII, the use of edible resources for packaging were left behind until recently.

After WWII, the synthesis of different plastic materials and their development for food packaging were significantly enhanced. The plastic industry has profoundly expanded in the 21st century. With a quick look at some aspects of our daily life is possible to identify many different types of plastics surrounding us. In the food industry, this expansion was also noticeable, mainly because the use of these materials has improved food preservation and extended the storage period of a lot of products. Many advantages have contributed to the extensive use of plastics in food packaging, such as low cost, good resistance to solvents and processability, flexibility, lightweight, easiness to be molded in different sizes and shapes, and others [1]. In the 1960s, more than 25% of bread sold were packaged in low-density polyethylene bags [10]. It was estimated that in the period of 1950 to 2015 the cumulative production of plastics reached more than 7.5 billion metric tons and, only in 2018, the global production of these materials has risen to 360 million tons [5,16]. Around 40% of the petrochemical-based plastics are used for packaging purposes and closely to 60% of plastic packaging are used to pack food and beverages [17].

Giant food industries and packaging companies have announced commitments to the reduction of plastics waste and to practice a circular economy for them. For example, Unilever[®], the owner of the food brands Ben & Jerry's[®] and Lipton[®], has declared in October 2019 an ambitious commitment with the reduction of the use of plastic packaging in more than 100,000 tons and use at least 25% of recycled plastic packaging until 2025 [18]. In Brazil, the actions of the group are supported by a long-running partnership with the retailer Grupo Pão de Açúcar[®], which helps in the waste collection through drop-off stations [18]. Dasani[®], a brand of bottled water commercialized by The Coca-Cola Company[®] in USA, has announced the first fully recyclable polyethylene terephthalate (PET) bottle 50% plant-based, HydriD Bottle[™] [19]. The Coca-Cola Company[®] has announced to make 100% of its packaging recyclable by 2025 and use at least 50% recyclable materials in their packaging by 2030 [20]. Other food companies, such as Keurig Dr Pepper[®], PepsiCo[®] and Mondelez[®], also have agreed to reduce the use of virgin plastic for its rigid plastic packaging [21]. According to the non-profit organization As You Sow [21], Keurig Dr Pepper[®]

used 208,000 metric tons of plastic packaging in 2018, and the company has planned to cut by 20% the use of virgin plastics by 2025.

Innovative packaging designed for food products with differentiated characteristics, such as high moisture content, high pressure and modified atmosphere, natural and fresh foods, among others, and with an environmentally friendly approach are a challenge for the research and development in the packaging industry [22]. Moreover, these biobased materials should be from renewable sources, recyclable and have low greenhouse gas emissions in the ecosystem [5].

Biobased packaging contains wholly or partly materials from biological origin [23,24]. Materials from biological sources (e.g., plants, animal, microorganisms, seafood, woods, and agricultural residues) are naturally biodegradable and compostable. These materials when used for packaging can be degraded by the action of living organisms upon disposal and converted to CO₂, CH₄, H₂O, inorganic compounds, or biomass, returning to nature [25–27]. Once made by biological resources, such as proteins, polysaccharides and microbial polyesters, packaging materials can be named as biobased plastics. Biobased materials are obtained from renewable carbon resources, as plants or biomass, such as chitosan, starch, cellulose, hemicellulose, poly(lactic) acid or vegetable oils [28,29]. Besides renewable resources, biodegradable materials can be produced from petroleum-based products [6,28]. Different biodegradable materials and their sources are displayed in Figure 1.

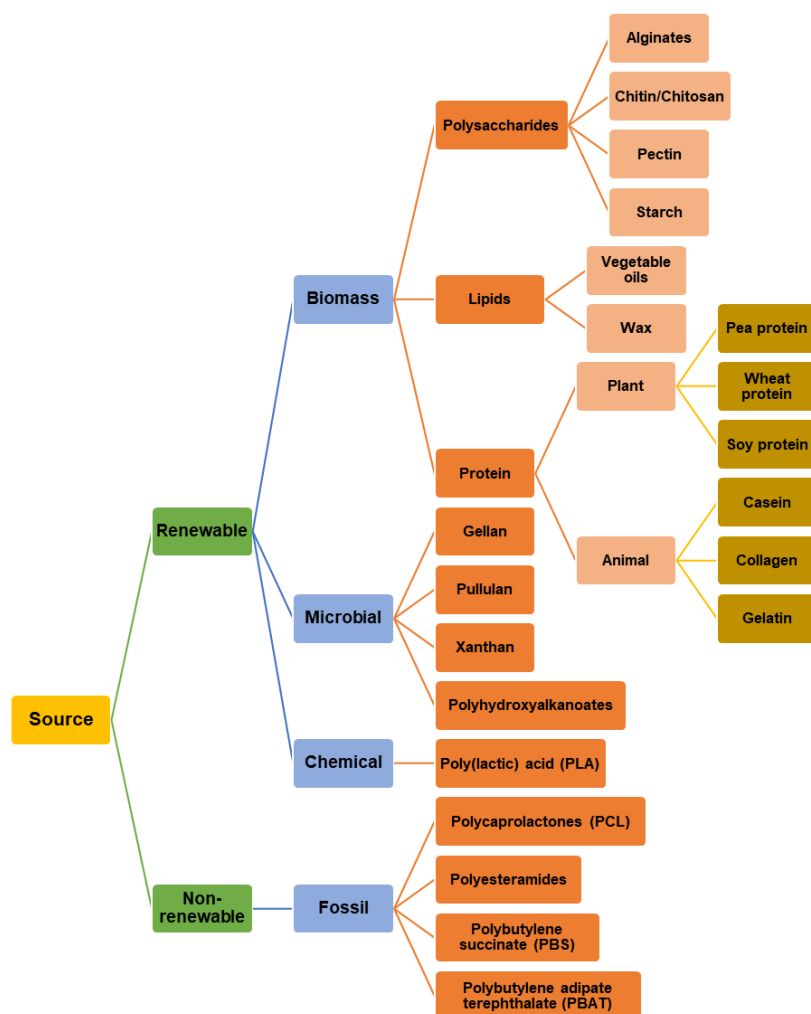


Figure 1. Classification of biodegradable materials used in biobased packaging.

Biobased, biodegradable, and composting plastics are terms frequently misunderstood and cannot be considered synonymous [25,30]. Not all biobased material is biodegradable

and otherwise, not all biodegradable material is derived from biological sources [31]. Different institutions and experts have diverted about a unique definition for the term biodegradable. According to the standard designation, D 996–04, from the American Society for Testing and Materials, biodegradable can be defined as being capable of decompose into carbon dioxide, methane, water, inorganic compounds, or biomass, by predominantly microbial enzymatic mechanism, which should be measured by standardized tests, in a specified period of time, reflecting available disposal conditions, compostable is the capacity of being biologically decomposed in particularly composting sites, breaking it down in carbon dioxide, water, inorganic compounds, and biomass, at a rate consistent with known compostable materials [32].

The biodegradability rate of biobased materials is intrinsically linked to their chemical structure. For example, the biobased polypropylene (PP) and polyethylene (PE) (vinyl polymers) obtained from the sugar cane processing are nonbiodegradable materials and are used similarly to the fossil-based PP and PE [33]. It is critical to underline that biodegradability do not imply a short time process and this should be expected to happen on a specific ecosystem at a particular time [25].

Nowadays, the global production of bioplastics, considered low, has reached a total of 2.11 million tons in 2020 and is expected to grow 15% until 2024 [33]. According to the report published in November 2020 by Intrado Globe Newswire [34], the world bioplastic market has generated more than U\$4.5 billion in 2019 and it is estimated to reach more than U\$13 billion until 2027. To them, the starch-based bioplastics market should totalize U\$561 million until 2023, with an annual growth rate (CAGR) of more than 3.5%. The major contribution to the bioplastic market was from the Asia-Pacific region. Europe should have the highest CAGR, more than 16%, until 2027 [34]. The distribution by sector of the bioplastics global production market in 2020 [35] is presented in Figure 2. Thus, the plastics and packaging markets are witnessing a significant increase on demand for materials from renewable resources.

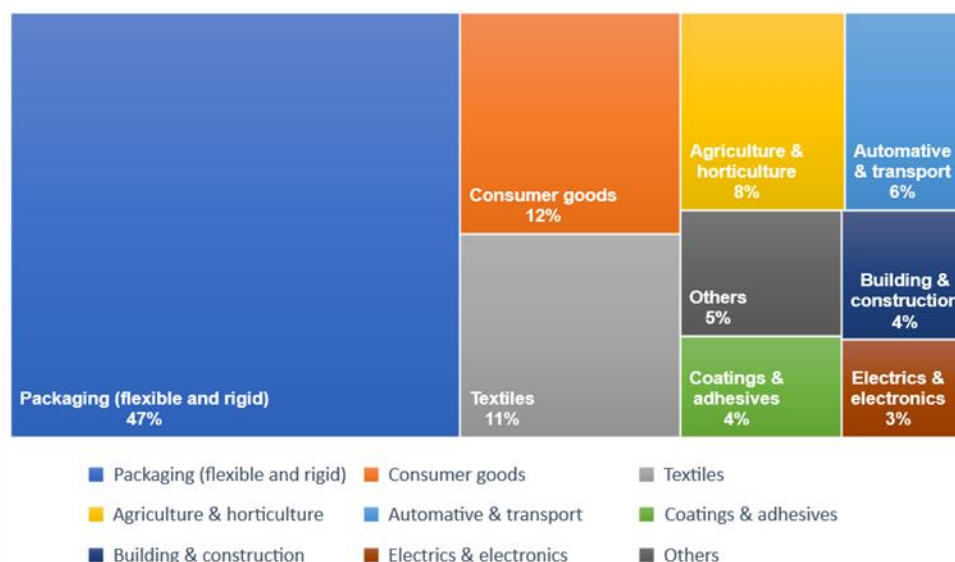


Figure 2. Distribution by sector of the bioplastics global production market in 2020 (total of 2.11 million tons).

Even with this high demand for biobased materials, some challenges must be overcome. Some examples may be cited.

- Improvement of national and international legislation to correctly establish the concepts and required standards related to bioplastics.
- Revision of industrial facilities for the processability of these materials while maintaining optimized and efficient performance.
- Maintaining the adequate quality of products and ensuring their biodegradability and compostability.

- Adjustments of the waste management system of each country/region to the collection of biobased plastics since each city has its own program.

3. Some Natural and Renewable Macromolecules Used to Prepare Edible Films and Coatings

A wide range of natural polymers from renewable sources has been used in the development of biobased food packaging. The main natural sources for this purpose are derived from polysaccharides, proteins, lipids, or blends of these macromolecules [36,37] (Figure 3). The utilization of these natural materials is partly linked to its biodegradability and renewability. However, some other advantages are expected when used for food packaging, e.g., as these materials can act as carriers of functional substances adding well-being benefits, incorporating flavorings and colorings aspects, enhancing organoleptic characteristics, improving mechanical and barrier resistance, and others [8].

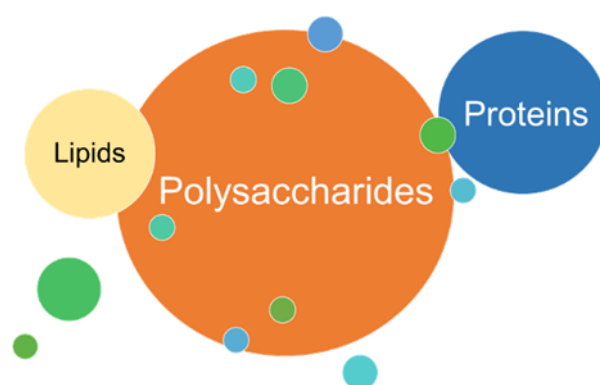


Figure 3. Major natural biological materials used in food biobased packaging.

Polysaccharides are known to possess good oxygen barrier and to have sites for hydrogen bonding formation, which can be used for incorporation of functional substances, e.g., coloring, flavoring, and antioxidants agents. By contrast, these materials do not exhibit good barrier to water vapor, which can be overcome by blending with other hydrophobic macromolecules, such as lipids [7,8]. Polysaccharides have been used to develop natural-based packaging [7,36].

Plant-derived proteins have gained remarkable attention of food manufactures and consumers in search for natural food resources and alternative materials to vegetarian, vegan, and food allergy diet restrictions. Protein-based film packaging exhibit extraordinary mechanical and barrier properties, especially to oxygen and carbon dioxide gases, when compared to polysaccharides [7,38]. Besides being eco-friendly materials, these films can nutritionally improve food quality and preservation. Moreover, the amphiphilic attribute of proteins contributes to their utilization as emulsifiers by stabilizing oil/water interface due to changes in interfacial tension [39]. Nowadays, a great variety of plant proteins have been used in food industry and packaging, such as from soybean, wheat, corn, sunflower, and pea [40].

Edible films can be defined as a thin layer of a material coating or placed between foods, in which act as a barrier and that can be consumed without any health risk [8,38]. Both are a primary food packaging and can be thought similar, although they differ substantially. Mainly, films are solid laminates, separately prepared, dried, processed and then used to cover food surface, placed between food parts, or used as an edible sealed bag [7,41]. Differently, coatings are prepared as a solution, directly sprayed or dipped on the food surface and then dried [42]. Thus, coatings can be considered as a part of the food product, since they are not made to be removed [8]. Moreover, films can be prepared as a mono, bi or multilayers. The latter provide a better water barrier to food but are less commonly used as they need two or more processing steps of casting and drying [7].

Both films and coatings should be composed only by food-grade components, GRAS (Generally Recognized as Safe), including any additives, such as plasticizers [42]. As any other packaging, these materials must protect the food integrity and maintain its quality [8,41,42]. In Figure 4 is presented the main purposes of edible food packaging. Although, it is not expected that the edible films could replace completely all conventional packaging, they can be used to significantly reduce the use of petroleum-based plastics, decrease food losses, and reduce the environment pollution over a long-term.

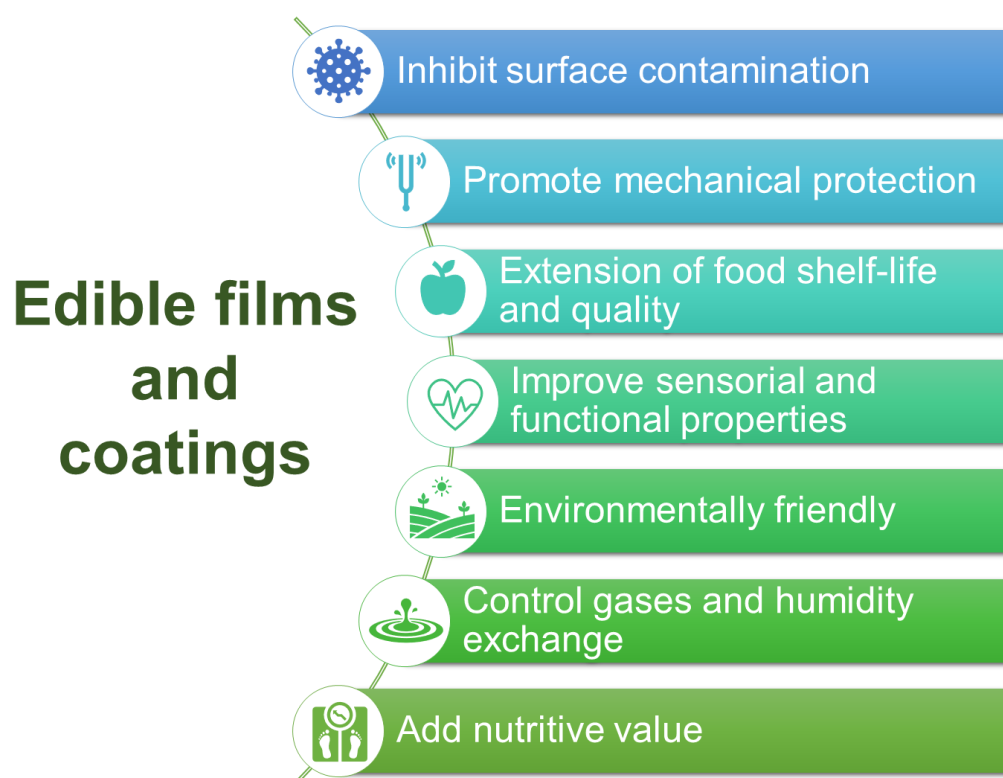


Figure 4. Main purposes of edible food packaging.

Edible films and coatings may have in their composition two main based materials, a biomacromolecule matrix and additives, such as plasticizers, cross-linkers, other reinforcements substances and functional ingredients [14]. These materials can be used alone or blended. Edible packaging composed by natural-based materials can be classified as a subgroup of biobased and biodegradable packaging [8]. Generally, polysaccharides, proteins, lipids, and their mixtures are used for the development of edible films and coatings. The classification and some examples of these natural materials commonly used for this purpose are presented in Table 1.

Table 1. Natural materials used in edible films and coatings.

Biomacromolecules	Raw Materials	References
Polysaccharides	Agar	[43]
	Alginate	[44]
	Cellulose	[45]
	Chitosan	[46]
	Gums	[47]
	K-carrageenan	[48,49]
	Pectin	[50]
	Pullulan	[51]
	Starch	[52,53]
Proteins	Casein	[54]
	Collagen	[55]
	Corn zein	[56]
	Gelatin	[45]
	Pea protein	[57]
	Sodium caseinate	[50]
	Soy protein	[58]
	Wheat gluten	[59]
	Whey protein	[60]
Lipids	Candelilla wax	[61]
	Bee wax	[62]
	Vegetable oils	[63]
Composites	Blends	[5,51,54,55,60]
	Residues and by-products from agriculture	[64–66]
	Fruit pulps	[67]

Edible coatings can be used to decrease weight loss, color changing, inhibit ethylene production and delay softening related to the postharvest ripening of fruits [68]. Different natural polymers, such as chitosan, carrageenan, alginates, gums, starches, their blends, and a wide range of additives can be used to prepare edible coatings and to be applied on fruits, as listed in Table 2.

Table 2. Edible coatings applied on fruits.

Matrix	Concentration	Additives	Food Product	Reference
Carboxymethyl chitosan/pullulan	Different mixing ratio of 5:0, 4:1, 3:2, 2.5:2.5, 2:3 and 1:4 (<i>w/w</i>)	8% galangal essential oil and 20% glycerol (<i>w/w</i>)	Mangoes	[51]
Chitosan	1% (<i>w/v</i>)	40 mmol L ^{−1} of ascorbic acid	Plums (<i>Prunus salicina</i>)	[69]
K-carrageenan	0.2–0.8% (<i>w/v</i>)	0–1% glycerol (<i>w/v</i>)	Papaya (<i>Carica papaya</i>)	[48]
Sodium alginate	0, 1 and 3% (<i>w/v</i>)	20% glycerol (<i>v/v</i>)	Plum (<i>Prunus salicina</i>)	[70]
Sodium alginate	1.5% (<i>w/v</i>)	<i>Ficus hirta</i> extract, 0.7% citric acid and 1.0% sucrose ester (<i>w/v</i>)	Nanfeng mandarin (<i>Citrus reticulata</i>)	[71]
Starch/chitosan	2%/0.5–1.0% (<i>w/v</i>)	2% glycerol (<i>w/v</i>)	Apples Fuji cultivar	[53]
Starch/gelatin	3–5%/10% (1:1)	10% sorbitol	Red Crimson grapes	[72]
Xanthan gum	2.5 g L ^{−1}	1.0 g L ^{−1} cinnamic acid	Pears (<i>Pyrus pyrifolia</i> and <i>P. communis</i>)	[47]

Moisture and oxygen absorption as well as lipids, flavor and odor change due to degradation or migration between packaging, food and the environment are important factors that impact food quality. These aspects are tightly correlated to the mass transfer phenomena during shelf-life of food products and cannot be dissociated from packaging properties [8]. Because of this, films and coatings should be developed accordingly to their

application purpose, e.g., layers for fruits should have lower water vapor permeability to reduce weight loss and reduce respiration.

The investigation of the water vapor permeability (WVP) of edible films are a valuable information to comprehend the mass transfer phenomenon since it implies diffusion and solubility of water moieties through the polymeric matrix. WVP represents the amount of water that infiltrates per unit of area of the film and time ($\text{kg m}^{-1} \text{s Pa}$), considering the film thickness and differential pressure [36]. Usually, polysaccharide-based films and coatings exhibit higher values of WVP than petroleum-based plastics. Natural polysaccharide-based films may present WVP value ranging from 6.6×10^{-13} to 1.7×10^{-8} than synthetic polymers, ranging from 1.1×10^{-14} to 0.5×10^{-11} [36,73]. The addition of lipids, plasticizers, and emulsifiers constitutes a useful method to overcome high values of WVP in edible films and coatings since this may decrease hydrophilicity.

The processing methods used to obtain films or coatings will be influenced by many factors, such as matrix composition, solubility, thermal behavior, flexibility, as well as their compatibility and cohesion forces, e.g., ionic, covalent and H-bonding, between polymer chains, and presence of additives [8,38,41]. Coatings can be applied on foods using dipping, spraying, fluidized bed processing, brushing, and panning methods. For the films production, mainly two types of methods are used, wet and dry processes. In the wet process, also known as casting, polymers are solubilized and dispersed onto a flat surface, then dried under specific conditions until film formation. In the dry processing, different methods, extrusion, injection, blow-molding, and heat-pressing, can be used without the use of liquid solvents. By these methods, high viscous polymers may be processed [8]. However, the dry method could not be used in the case of thermosensitive substances to be present in the matrix composition.

A composite edible film packaging commonly will consist of a blend of two or more hydrocolloids aiming to improve their individual properties and application [74]. Protein and lipid can be supported on a polysaccharide matrix, or the lipid material may be encapsulated on hydrocolloids and dispersed on the polysaccharide film matrix [8]. This configuration may be used to improve mechanical and thermal properties, and to overcome some processing limitations. Furthermore, the incorporation of natural or chemical additives, such as plasticizers, may also contribute to improve mechanical, thermal, functional, sensorial, and nutritional features of edible films [75,76]. The use of plasticizers to improve those properties of edible films have been well reported in the literature [56,77–79]. Nanofibers prepared by electrospinning have also been incorporated in edible films to reinforce their mechanical and thermal properties [14]. This nanofillers change the molecular mobility and relaxation of the matrix, thus leading to significant influence on thermal behavior of nanocomposite films [14]. The incorporation of essential oils in poly(lactic acid) films had a relevant effect in the results of their thermogravimetric curves and thermal stability, due to the formation of hydrogen bonds between poly(lactic acid) and essential oil, which could be linked to its delayed degradation behavior [29].

3.1. Alginates

Alginates are natural anionic polysaccharides from marine origin. These biopolymers are widely used in the food and pharmaceutical/cosmetic industries because of its biodegradability, biocompatibility, bioavailability, nontoxicity, and low price [80,81]. Alginates are largely isolated from different brown seaweeds. *Laminaria digitata*, *Macrocystis pyrifera* and *Ascophyllum nodosum* are the most important sources of alginates [82]. As expected, the seaweed source has influence on their properties. Although they can be obtained from microorganisms (e.g., *Azotobacter* and *Pseudomonas* genera) processing is not economically feasible [80,83]. The main advantage to their use may be attributable to their functional properties. They present crosslinking capacity, good gelling and film-forming properties, fine water absorption, pH-sensitivity, mucoadhesiveness and others [68,81,83]. Moreover, alginic acid and its salts are classified by the U.S. Food and Drug Administration (FDA) and

European Commission (EC) as a food grade additive and approved to be used as gelling, emulsifier, and thickener agents [75].

Alginates can be extracted from seaweeds with a hot alkali solution, usually sodium carbamate. Afterwards, alginates dissolve as sodium alginate forming a very dense slurry, which may contain undissolved algae cellulose parts, and need to be diluted, pressed and filtered. Finally, the alginate sample is precipitated from the filtered solution, either as alginic acid or calcium alginate. To improve this process, to obtain a less colored product and reduce the loss of viscosity, an acid pretreatment can be performed [68,82]. The monovalent salts from alginic acid are water-soluble [75]. It can be highlighted that sodium alginate is the most used salt from alginic acid.

Structurally, alginates are linear copolymers of α -1,4-L-guluronic acid (G) and β -1,4-D-mannuronic acid (M) repeating units. These units may be arranged as G and M blocks, and as regions of alternating G and M units (Figure 5) [80]. The molecular weight of alginates varies from 32 to 400 kDa, according to their source, and processing methods [84]. In the 1960s, different researchers using partial hydrolysis were able to separate the alginic acid in three fractions, where two of them contained units of G or M, whereas the third fraction contained balanced proportions of both blocks, MG dimers residues. Later on, it was elucidated that alginates have no regular repeating unit and that the monomers distribution along the backbone chain cannot be predicted by statistical analysis, Markov or Bernoullian's models [83]. These models were used to assign the structure of alginates from bacteria, though they do not fit for ascribing the algal monomer sequences [82].

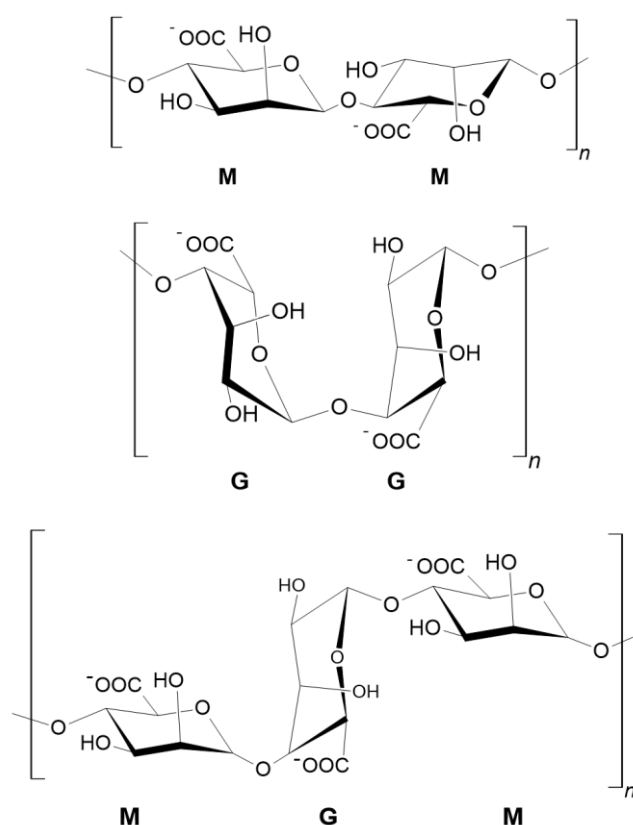


Figure 5. Chemical structures of mannuronic acids diads (MM), guluronic acid diads (GG), and alternated mannuronic acid/guluronic acid/mannuronic acid triad (MGM) in alginate.

The industry of seaweed hydrocolloids, which includes alginates, agars, and carrageenans, has been growing in the order of 2–3% per year, mainly in Asia [85]. China is the major manufacturing market of seaweeds, reaching 10 million tons in 2014, especially of *Laminaria* (*Saccharina*) *japonica*, followed by Indonesia and Philippines in Asia [85,86]. In the Americas and Europe, the seaweed market is dominated by Chile (*Lessonia* spp. and

Macrocystis), with more than 20,000 dry tons in 2015 and Norway and France (*Laminaria* ssp. and *Ascophyllum* ssp.), respectively, with a production of more than 32,000 dry tons in 2015 [85]. Besides the fact that this market is expanding, only a small fraction of the global biomass supply of seaweeds is produced. Some researchers suggest that the cultivation of seaweeds offshore could provide an alternative source of biomass, leading to a sustainable production of food, chemicals, and biofuels [86].

The utilization of seaweeds for food and feed by mankind dates back to antiquity. The earliest written report was tracked in China and dated from about 1700 years ago [86]. For many centuries, harvesting and consumption of seaweeds were linked to coastal inhabitants, until their scaleup industrial utilization in the 19th century. The first extraction reports of alginic acid extracted from kelps are from the 1880s, by the British chemist Edward C. Stanford [83]. He patented and published a paper with his discovery in 1883 in *The Chemical News and Journal of Physical Science*, entitled 'On Algin: A new substance obtained from some of the commoner species of marine algae'. Later, alginate was firstly used as a food gelling agent to the production of artificial cherries in 1946 [87]. Since then, alginates have been widely used in different industries, such as in textile, pharmaceutical, cosmetic, food and other fields.

Alginate solubility depends on 4 main factors, G and M distribution blocks, solvent pH, ionic strength, and presence of gelling ions. It can go through sol-to-gel transformation by ionotropic gelation, resulted of the addition of multivalent ions that will act as crosslinking agents, e.g., calcium, magnesium, aluminium, manganese, and iron, or by lowering the pH below the pK_a value of the guluronic residue [82,88]. Alginate is greatly sensible to pH changes due to the presence of carboxylic acids along its chain. At pHs < 3.4 (below pK_a of M = 3.38 and G = 3.65), the carboxylic groups are nonionized ($-COOH$) making alginate insoluble, whereas, at a pH > 4.4, these groups become ionized ($-COO^-$), resulting in chain expansion by the electrostatic repulsion of these negative charges, reaching the highest point at pH 7.4 [80]. Alginate affinity for divalent ions decreases in the following order: trivalent cations > Pb^{2+} > Cu^{2+} > Cd^{2+} > Ba^{2+} > Sr^{2+} > Ca^{2+} [68,80]. For alginate gels formation, a three-dimensional network is diaxially formed by the interaction of carbonyl groups from the G blocks with multivalent cations, such as calcium, resulting in the formation of a cavity or binding site, commonly named as "egg-box" conformation or zipping mechanism [68,84]. Preparation, characteristics, and binding mode of alginate with different ions were critically reviewed by Hu et al. [84]. The flexibility of alginate matrices can be increasingly influenced by their segment blocks according to the following order: MG > MM > GG [82]. Besides this interchain interactions, it is known that alginate as an anionic polyelectrolyte can be combined with a cationic polyelectrolyte, such as chitosan via complexation of their opposite charges, complex coacervation, aiming the formation of micro/nanoparticles to encapsulate bioactive substances [89].

3.2. Carrageenans

Carrageenans are linear polysaccharides also obtained from marine sources, the red seaweeds that belong to the *Rhodophyceae* family. They are mainly extracted from *Kappaphycus alvarezii* and *Eucheuma denticulatum*, although other species (e.g., *Chondrus crispus*) from Ireland and England have been used [68]. Historically, this seaweed has been known since 400 A.D., especially in Ireland, as a homemade medicine to treat respiratory diseases, and as a gelling agent. Moreover, the first reports on the use of *Chondrus crispus* to produce carrageenan dates from 1862, by the British chemist Edward C. Stanford, who also worked with alginates back then [68,83]. The name *carrageenan* and its utilization as we know nowadays have been proposed after the 1950s [68].

Obtaining macromolecules from seaweeds has many advantages, since these algae have a fast growing, thus producing large amounts of renewable biomass that are available in abundance to many industrial purposes [90]. Besides, these materials are biodegradable, biocompatible, present low or no toxicity, as well as have many biological properties. These biopolymers are used in the development and formulation to a wide range of

pharmaceutical and food products because of its emulsifying, gelling, and stabilizing properties [91].

Carrageenans are sulfated water-soluble polymers consisting of 1,3-linked- β -D-galactopyranose and 1,4-linked- α -D-galactopyranose alternating units [7]. Industrially, the most employed types are κ -(kappa), ι -(iota) and λ -(lambda) carrageenans, being classified according to their molecular structures, which include the sulfation arrangements as well as the existence or absence of 3,6-anhydro- α -galactopyranose on the 1,4-linked α -galactopyranose unit [92]. Figure 6 shows the main types of carrageenans used for industrial purposes.

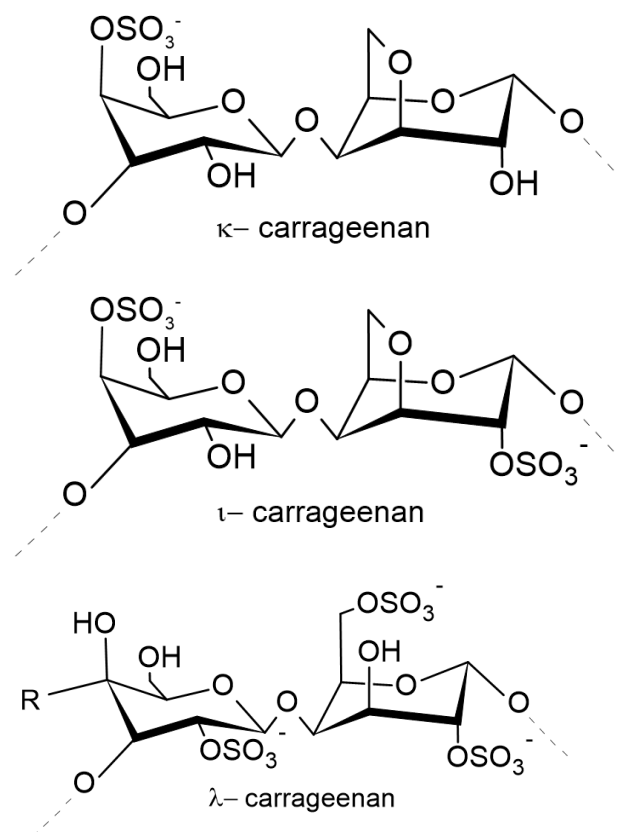


Figure 6. Chemical structures of the three types of carrageenans.

Kappa-carrageenan and iota carrageenan exhibit about 25–30% and 28–30% of sulfate content, respectively [93]. These carrageenans are naturally hydrophilic polymers belonging to one of the major groups of galactans, extracted from cell walls of red seaweeds [68]. This sulfated water-soluble behavior, as well as their chemical structure, which can form a helical assembly, is responsible for its great potential to prepare film-forming solutions, thus having good employment to the design of edible films and coatings [68,93].

To the design of edible packaging, carrageenans have been used to replace casings made from animals' gut for sausage making, and as coatings in solid oily foods, meat, fish and poultry for preventing their superficial dehydration [91]. In a recent study, Tran et al. [49] developed and optimized a composite film based on sodium alginate, kappa-carrageenan, *Momordica cochinchinensis* pulp and glycerol. These authors noted that the optimal film composition was 1.03% of sodium alginate, 0.65% of kappa-carrageenan, 0.4% of *M. cochinchinensis* pulp, and 0.85% of glycerol (*w/v*). Their composite film exhibited good mechanical and physical properties, and low water vapor permeability, which demonstrated potential for food application.

Jancikova et al. [93] studied the addition of 5–20% of lapacho extract (LE) in edible films also containing ι - and κ -carrageenan as matrices. These authors observed that the use

of those different gelling agents in the composite films resulted in significant statistical differences ($p < 0.05$) in their tensile strength and breaking strain properties. Besides this, these authors also evaluated the antioxidant properties of the produced films and noted the best results 20% for the lapacho extract- κ -carrageenan film. In another work, Simona et al. [94] investigated the properties of carrageenan films to which orange essential oil and trehalose were incorporated. Their results showed that the incorporation of orange essential oil with trehalose had a decreasing effect in UV-Vis transmittance for the carrageenan-based films, thus producing a UV protector coating. These authors also observed that the films exhibited resistance to Gram-positive bacteria (*Staphylococcus aureus* subsp. *Aureus*).

3.3. Chitosan

Chitosan is a polysaccharide derived from chitin, which is found in crustaceans shells, in the exoskeleton of insects or produced by microorganisms [95,96]. After cellulose, chitin is the second most abundant polysaccharide in the world, exhibiting a global turnover around 10^{11} tons per year [97]. The earliest works with chitin and chitosan date from the 1850s with Charles Rouget and later in 1890s with Felix Hoppe-Seyler [95]. Since then, chitosan has been used to a wide range of purposes, mainly because of its renewable sources, relevant biological properties (e.g., as antioxidant, antimicrobial, and anticancer activities), as well as to its biodegradability, biocompatibility and nontoxicity [31] (Figure 7).



Figure 7. Diverse biological, functional and technological properties of chitosan.

Chitin is composed by (1,4)- β -2-acetamido-2-deoxy-D-glucopyranose units and low amounts of (1,4)- β -2-amino-2-deoxy-D-glucopyranose units, whereas chitosan is composed of N-acetyl-D-glucosamine (A-units) and deacetylated D-glucosamine (D-units) units [98]. The main difference is that chitosan is deacetylated (Figure 8), which makes it soluble in mild acidic solutions, whereas chitin is insoluble in many organic and inorganic solvents [99]. The presence of hydroxyl and amine groups in chitosan chains allows the occurrence of chemical changes under pronation, which can influence significantly its biological, mechanical, and physical-chemical properties [46,96]. The NH_2 groups located at C-2 of rings on the D-glucosamine repeating units promote a polycationic behavior of the chitosan chains [46]. This polycationic characteristic allows the formation of electrostatic interactions between chitosan and other polyanionic biopolymers, such as sodium alginate, and some

amphiphilic proteins. The formation of such complexes is used to encapsulate bioactive substances and to form micro- or nanoparticles by complex coacervation [100–102]. The protonated amino groups are responsible for chitosan interaction with negative cell membranes of microorganisms, altering its permeability and metabolism, chelate trace metals and microorganism's spores, thus leading to cell death and inhibiting microbial growth [31].

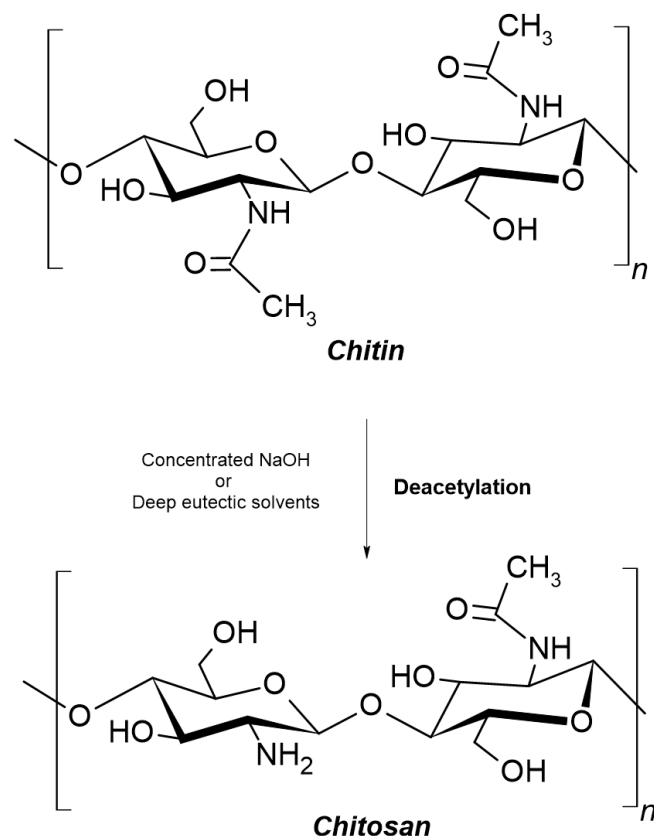


Figure 8. Deacetylation of chitin to chitosan.

Due to its physicochemical and biological properties, chitosan has been widely exploited to the design of different food packaging. Besides, chitosan-based films and coatings are environmentally friendly materials because they are biodegradable in many landsites and aquatic environments [103,104]. In a recent work, Hiremani et al. [105] studied the physicochemical and biological properties of a bioactive film of chitosan/white turmeric starch prepared by the solvent casting technique. These authors observed that the produced films exhibited good tensile properties, a smooth surface, as well as a fine miscibility, water resistance and UV barrier behavior. They found that the chitosan/white turmeric starch blended films were more than 60% degraded within 15 days, confirming its well-reported biodegradability. In another study, Sady et al. [106] incorporated chokeberry pomace extracts into chitosan-based films for potential food application. Their films showed good antioxidant properties, as well as enhanced UV–vis light and water vapor barrier, and reduced oxygen permeability.

Chitosan-based films plasticized with 30% of glycerol were used to coat strawberries by Pavinatto et al. [107]. These authors found that the produced film had a bactericidal behavior against gram-positive and gram-negative bacteria and a fungal resistance, which protected the strawberries from microbial degradation without altering its sensorial properties. Chitosan and curcumin grafted with 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-oxidized cellulose nanofiber were used to prepare nanocomposite films by casting [108]. These films presented increased crystallinity due to the addition of nanofibers, as well as oxidation resistance and an improved UV blocking property, although the mechanical strength and water vapor permeability decreased. In another recent study from

Qiao et al. [109], the physicochemical and structural behavior of chitosan films prepared with different solvents were investigated. These authors noted changes in the crystallinity of the chitosan films, which presented an amorphous structure in lactic and citric acids, whereas a partial crystalline structure was present when the films were prepared with hydrochloric and acetic acids. Their observations highlight the importance to choose a proper solvent to the design of chitosan-based films aiming target applications. Moreover, chitosan films can be affected not only by the solvent used and pH of the media, but also by chitosan molecular weight, deacetylation degree, type and content of plasticizer, film matrix and composites.

3.4. Starch

Starch is a natural polysaccharide obtained from different renewable vegetable sources and their byproducts. This biopolymer has gained great attention due to its diverse industrial and biotechnological applications, abundance, world-wide availability, low-cost, biodegradability, edibility, and good processability in conventional plastic processing equipments [110–112]. Cereals and tubers, e.g., maize, cassava, wheat and potato, are the main sources of the worldwide industrial production of starch, although other vegetable sources and their residues such as sorghum, rice, sweet potato, yams, barley, sago, and some nonconventional fruits, tubers, legumes and pseudo-cereals can also be exploited [113–115]. This diverse botanical sources as well as many environmental factors can lead to differences in the molecular structure, composition, shape, and size of starch granules, which will significantly influence its processing properties [111,112].

Native starch is a heterogeneous material, chemically composed by two polyglucans, amylopectin and amylose, forming semicrystalline granules [111,115–117], as can be visualized in Figure 9. The first is the major component, comprised by a highly branched short chains of (1,4)- α -linked *D*-glucosyl units connected by (1,6)- α -linkages. The other is amylose, which has linear longer chains and is a minor component (up to 35%) in starches [115,116]. Generally, starches can contain around 20–25% of amylose and 75–80% of amylopectin [111]. Some mutant plants may have high content of amylose, i.e., amylo-maize or Hylon, whereas others may have little or no amylose, e.g., waxy corn and waxy potato [115]. Amylose has a molecular weight 10 times higher than conventional synthetic petroleum-based polymers, e.g., polyethylene and polypropylene, while has a much lower molecular weight than amylopectin, which can reach millions [110].

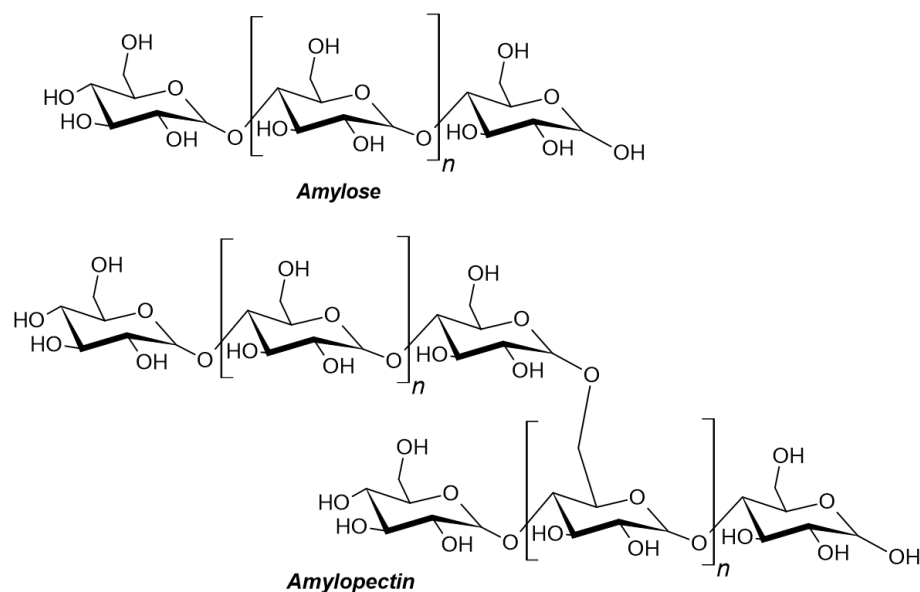


Figure 9. Chemical structure of amylose and amylopectin as the main molecules constituting starches.

The amylose/amylopectin ratio is influenced by the starch origin and, consequently, leads to different crystallinity degrees. The technological properties of starches depend on their crystallinity, which can vary from 20% to 45% [110,111,117,118]. Mainly because of the presence of strong intermolecular hydrogen bonds, starches are not solubilized in cold water. In hot water, these bonds are weakened by the interaction between water molecules and hydroxyl groups of amylose and amylopectin, which leads to partial starch solubilization [111]. When in solution, amylose can crystallize, forming left-handed double helices, which are packed in, either A- or B-type allomorphs [115]. This phenomenon is called retrogradation. The temperature, solvent and starch/solvent ratio used for solubilizing starch are significant factors that can influence its gelatinization, a relevant process in the production of starch-based films and coatings [111]. Different physicochemical reactions occur during the thermal processing of starches, such as water diffusion, granule swelling, gelatinization, decomposition, melting and crystallization, though the gelatinization is among the most important [110]. Physicochemical techniques, e.g., X-ray diffraction, polarized microscopy, nuclear magnetic resonance, and differential scanning calorimetry are commonly used to investigate the gelatinization process of starch [110].

Starch-based films can be prepared by using a wet or dry process. In the first, the films are made by film-forming dispersion using the casting technique. In the second, by thermoplastic or thermal processing using extruders, thermoplastic starch is produced [110,111,117]. Starches form suspensions with good film-forming capability. Similar to other natural polymer films, the starch-based films exhibit several advantages. Starch films are tasteless, colorless and odorless. They present good oxygen barrier properties, nutritional value, edibility and others [105,118]. However, due to their hydrophilic nature, starch films have some disadvantages, mainly related to their low water vapor barrier capacity and mechanical properties [114,117].

To overcome these limitations, some plasticizers and other hydrophobic substances (e.g., polyols, as well as the incorporation of other natural polysaccharides and proteins to form composite films) can be added to improve the water vapor barrier and mechanical properties [77,111,117,119]. Glycerol is among the most used and best plasticizer for starch-based films. Recently, some ionic liquids or deep eutectic solvents have been used to this end [77]. These alternative plasticizers have been used because they interact with hydroxyl groups of starch chains, exhibit high thermal stability, and low density, vapor pressure, toxicity and viscosity at room temperature [77,79]. Table 3 shows some starch-based films and coatings.

Table 3. Some starch-based films and coatings.

Matrix	Biopolymer Component	Additives	Reference
Potato, or corn, or wheat starches	-	1-ethyl-3-methylimidazolium acetate (1.5 wt%)	[77]
White turmeric (<i>Curcuma zedoaria</i>) starch	Chitosan	Glycerol	[105]
Purple yam starch	Chitosan	Glycerol	[53]
Corn or cassava starches	Chitosan	Lactic acid (1%, v/v), Glycerol (0.9 g 100 mL ⁻¹)	[120]
Potato starch	Chitosan	Citric acid (5–20%)	[121]
Arrowroot starch	-	Blackberry pulp (0–40%)	[122]
Babassu starch	-	Glycerol, sorbitol, glucose, or urea	[78]
Pea starch	Guar gum	Glycerol (25%)	[123]
Wheat starch	-	Glycerol (0–50%)	[124]
Rice starch	Glutelin	Sorbitol (40%)	[125]
Cassava starch	Yam starch	Glycerol (20%)	[126]

3.5. Pea Protein

Plant-derived proteins have gained remarkable attention of food manufactures and consumers in the search for natural food resources and alternative materials for vegetarian,

vegan, and food allergy diet restrictions. Protein-based film packaging exhibits extraordinary mechanical and barrier properties, especially to oxygen and carbon dioxide gases, when compared to polysaccharides [7,38]. Besides being an eco-friendly material, these films can nutritionally improve food quality and preservation. Moreover, the amphiphilic attribute of proteins contributes to their utilization as emulsifiers by stabilizing oil/water interface due to changes in interfacial tension [39]. Nowadays, a great variety of plant proteins have been used in food industry and packaging, such as from soybean, wheat, corn, sunflower, and pea [40].

Pea, *Pisum sativum* L. ssp. *sativum*, is a notable grain legume that belongs to Fabaceae (or Leguminosae) and Papilionoideae botanical families, such as soybean [127]. It is known by several common names such as common pea, green pea, garden pea, field pea, yellow field pea, spring pea, English pea, and Austrian winter pea (*Pisum sativum* L. ssp. *sativum* var. *arvense*) [128]. Pea is a cool-season pulse crops largely cultivated for protein and feed purposes, being one of the most important food harvests, reaching around 11 Mt per year worldwide [127]. The major producers in 2014 were Canada and USA in the Americas, France in the European Union, Russia in Eastern Europe, and Australia in Oceania regions [128]. Nowadays, the major producing regions, in ascending order, are Asia, Europe, Africa, Americas, and Oceania, reaching more than 21 million tons of green pea production worldwide in 2019 [129].

Up to 30% of proteins can be found in peas, which contains 65–80% of globulins and up to 20% of albumins [39]. Moreover, peas exhibit a good-balanced amino acid profile, highlighting higher amounts of lysine, around 6%, and tryptophan than cereal grains. By contrast, peas are deficient in methionine and cysteine, in relation to cereal grains [130–132]. Five proteins fractions can be found in peas. Besides globulins and albumins, prolamins, glutelin are found in peas [128,133]. Albumins are water-soluble metabolic and enzymatic proteins involved in cytosolic functions. Unlikely, globulins are salt-soluble proteins, which are used on seeds germination to provide nutrients for plant growing [130].

The major storage proteins found in pea seeds are globulins, which consists of legumin (11S, M_w ~380 kDa) and vicilin (7S, M_w ~60 kDa) and convicilin (7S). Legumin is a hexameric molecule containing six subunits held together by noncovalent interactions, that contains 4–5 acidic (α) (40 kDa) and 5–6 basic (β) (20 kDa) polypeptides from several gene families' precursors, linked by disulfide bonds. Vicilin is a trimeric protein containing three subunits each 50 kDa in size, that lacks cysteine residues and cannot form disulfide bonds. Convicilin, a third major storage protein in peas, has a subunit of ~71 kDa and a M_w in its native form of 290 kDa [130,134,135]. The variations in content, composition, and structure of both globulins, vicilin and legumin, reflect in their nutritional and functional properties, e.g., higher globulins content may result in better emulsification properties [39].

The pea proteins, as well as other proteins in seeds are considered bioactive proteins, as they can contribute to wellness and health. Nevertheless, the food applications of pea proteins are related to their nutritional value, distinguishable functional properties, low allergenicity, non-GMO status, and biological activities, such as reducing the risk of cardiovascular disease and lowering blood pressure effect [132]. The extraction methods used to obtain pea proteins have a significant impact on its functionalities. A diverse number of methods have been used to extract plant proteins, which can be classified as conventional (solvent and alkali-based), physical (ultrasound, microwave, high-pressure and pulse electric field-assisted extraction), and green extraction technologies [136].

For pea protein isolation, two processes (dry or wet extractions) can be used. The dry process is based on air classification, in which the pea seeds are milled into a flour, and separated in light and heavy fractions, protein and starch-rich respectively, by the application of a spiral air stream. For the wet process, the proteins are extracted according to their solubility in solutions, alkaline or saline, and further isolated by dialysis, micellar precipitation, or ultrafiltration [128,137]. Recent studies have found that the methods used for pea protein recovery influence its composition and gelling properties. Ultrafiltration and dialysis let to higher albumin fraction, whereas alkaline-isoelectric and micellar precip-

itation result in the retention of globulins [137]. In that work, gels formed by pea proteins with higher compressive strength resulted from micellar precipitation and ultrafiltration extractions. When considering the isolated protein yield and protein solubility, the best results can be achieved by salt extraction-dialysis, followed by alkali extraction/isoelectric precipitation and micellar precipitation, as found by Stone et al. [138]. In that same work, the authors found that salt-extracted isolates displayed the highest oil holding capacities and lowest water holding capacities. Additionally, pea protein isolate (PPI) is commonly obtained using isoelectric precipitation with pH around 4.5 followed by membrane separation, e.g., ultrafiltration or diafiltration, to increase protein concentration [130]. Thus, the technological understanding involving pea protein extraction and composition helps to achieve their best functional physicochemical properties and meticulously design of formulated products.

Generally, functional properties of proteins can be categorized as related to: (a) hydration (absorption of water and lipids, solubility, wettability, and thickening); (b) structure and rheological behavior (adhesiveness, viscosity, elasticity, gelation and aggregation); (c) protein surface (emulsification, foaming, whipping and formation of protein-lipid-based films) [131]. From these, solubility, water- and lipid-binding capacities, rheological, emulsifying and gel forming properties, can be cited as the most significant properties for application of pea proteins in food products.

Beyond processing factors, environmental conditions, e.g., temperature, pH, ionic strength, and type of solvent, can significantly affect the functional properties of pea proteins, especially its solubility. The solubilization of pea protein isolate is deeply influenced by pH variation, presenting minimum solubility between pH 4–6 [131]. In another work, at neutral pH, pea protein exhibited the highest surface hydrophobicity value, low surface, and low solubility, resulting in a lower emulsion capacity when compared to chickpea, fava bean and lentil proteins produced by isoelectric precipitation and salt extraction [139]. To overcome these issues, the use of polysaccharides forming stable complexes with proteins have gained interest. It prevents proteins aggregation and precipitation and improve functionalities. Furthermore, protein-polysaccharides plays important roles controlling food systems stability and forming film/coating and composite packaging materials.

Differently from polysaccharides, proteins-based films exhibited better mechanical and optical properties, as well as good barrier against gases permeability, especially O₂ and CO₂. The use of protein in food packaging provides a wide range of interactions and chemical reactions with other substances, through covalent (peptide and disulfide) and noncovalent (ionic, hydrogen and van der Waals) bonds [38]. They can be prepared as a mono or multilayer food packaging and blended with other polymers as a composite film. Some of the most used proteins in edible films are collagen, zein, gelatin, soy protein, milk protein and pea protein [140].

3.6. Improvements by the Incorporation of Natural Bioactive Substances into Edible Films

The incorporation of bioactive agents into packaging films is a growing strategy that aims to extend shelf-life, to increase lipid and microbial stability and safety when it is applied to food products. Furthermore, the use of active packaging to extend the food shelf-life and safety is an emerging and dynamic technology in food processing, since this coating can actively interact with the food surface and the environment based on the intrinsic property of their constitutional biopolymers or incorporated additives [141]. Moreover, using natural bioactive compounds have become a good alternative for traditional packaging and replace synthetic additives commonly used, e.g., butylated hydroxytoluene (BHT) [142]. Besides this, the addition of bioactive substances has been associated to microbial and physical improvements in film properties as well as health beneficial activities, such as antioxidant and probiotic, to consumers intake [143]. Some uses of different bioactive substances on chitosan-based films are listed in Table 4.

Natural bioactive materials can be obtained from plants, animals, marine organisms, and microorganisms using biotechnological methods [38]. Plant-based bioactive substances

are widely found in nature and is mainly secondary metabolites that presents inherent biological functions. They can be obtained from renewable plant parts, such as, leaves, flowers, fruits, seeds, grains, roots, and other vegetable portions, as extracts, resins, essential oils, or vegetable oils and fats. In these materials, a wide number of natural phytochemicals, such as phenolic acids, carotenoids, flavonoids, vitamins, isoflavones, indoles, saponins, monoterpenes, bioactive peptides, phytosterols, essential fatty acids, and others can be categorized as bioactive compounds [144].

Table 4. Incorporation of bioactive substances in chitosan-based films.

Additives	Biological Properties	Main Results	Reference
Maqui berry extracts	Antimicrobial and antioxidant	<ul style="list-style-type: none"> – Effective against seven bacteria species. – Higher antioxidant activity. 	[145]
Grape seed extract and <i>Ziziphora clinopodioides</i> essential oil		<ul style="list-style-type: none"> – Improvement of total phenolic content, antibacterial and antioxidant activities, thickness, and water vapor barrier property. 	[146]
Propolis extract		<ul style="list-style-type: none"> – Inhibition all tested bacteria on contact surface. – Tensile strength, elongation at break, total phenolic content and antioxidant activity increased. 	[147]
Apple peel polyphenols		<ul style="list-style-type: none"> – Improvement of the film physical properties – Antioxidant and antimicrobial activities were significantly increased. 	[148]
<i>Zataria multiflora</i> Boiss essential oil		<ul style="list-style-type: none"> – Gelatin-nanochitosan films containing <i>Zataria multiflora</i> Boiss. essential oil delayed the growth of bacteria in chicken meat. 	[149]
<i>Zataria multiflora</i> Boiss essential oil and grape seed extract	Antioxidant	<ul style="list-style-type: none"> – Wettability of the surface, total phenol and antioxidant activity increased. 	[150]
Green tea and black tea extracts		<ul style="list-style-type: none"> – Green tea films presented stronger antioxidant activity than that of black tea films in all food simulants. 	[151]
Carvacrol	Antibacterial activity	<ul style="list-style-type: none"> – Incorporation of carvacrol was not effective against bacterial activity. 	[5]

Natural bioactive substances present many advantages for food preservation when added into packaging. Some disadvantages may persist, such as off-flavors, thermosensitivity and early loss of functionality, which may take place during processing and storage. Furthermore, the bioactive efficiency decreases when exposed to oxidative conditions of the surrounding environment (air, humidity, heat, light, oxygen, and others) [152]. Thus, encapsulation of bioactive substance onto biopolymeric matrices, followed by incorporation of these active particles in the food packaging is an effective strategy, which can improve durability and maintenance of biological properties [153,154]. Some examples of the incorporation of lipid materials onto different hydrocolloid matrices are listed in Table 5.

Table 5. Incorporation of lipids onto different hydrocolloid matrixes.

Hydrocolloids	Lipids	Form	Application	Reference
Chitosan and sodium alginate	Açaí pulp oil	Microcapsules by complex formation	Potential application to biobased packaging	[101]
Gelatin and gum Arabic	Methyl oleate	Microcapsules by complex coacervation	Functional lipids and lipophilic food ingredients	[155]
Gluten and gelatin plasticized with glycerol or sorbitol	Fatty acids	Incorporated in edible films	Biobased packaging	[156]
Mesquite seed gum	Palm fruit oil	Emulsion edible film	Biobased packaging	[63]
Chitosan/xanthan or chitosan/pectin	Palm oil	Microcapsules by complex formation	Yogurt preparation	[157]
Chitosan/sodium tripolyphosphate or chitosan/carboxymethyl cellulose	Palm oil and β -carotene	Microcapsules by complex formation	Food systems under gastrointestinal simulant conditions	[158]
Chitosan plasticized with glycerol	Olive oil	Emulsion film	Biobased packaging	[159]

Furthermore, vegetable oils are a feasible source of essential fatty acids and sterols, particularly, alpha-linolenic acid and linoleic fatty acids and phytosterols, which are related to health benefit effects in reducing the risks of heart diseases, displaying antioxidant activity and others associated to their incorporation in food products [160]. Besides, they are obtained from renewable sources, edible, effortlessly available, generally display low cost, are nontoxic, nondepletable and nonvolatile. Lipid materials can be used to promote a desirable smooth effect, improve water, and gas barrier, as well as enhance cohesivity when incorporated in biobased films [38,141].

The functionality of these bioactive materials may be different, as each component introduces a wide range of properties to the matrix. Besides, functionality depends not only on intrinsic characteristic of each component, but significantly on their compatibility and cohesivity. Different approaches to this can be pursued, e.g., incorporation of the encapsulated particles on the external surface of films or in the interface between the film and the food and dispersed among the film [38]. It is expected that these incorporated active compounds will be released from the package or absorb substances from food and surrounding environment, as well as make changes in food composition and sensorial characteristics [141].

4. Conclusions

Research on novel biobased food packaging has been growing over the last decade mainly boosted because of concern with the global environmental plastic pollution. Innovative strategies to develop biodegradable and sustainable packaging materials based on natural resources are welcome to reduce the use of plastics. The present review highlighted a historical timeline, functionality and application trends of alginate, carrageenan, chitosan, starch and pea protein, as natural biopolymers for the design of food packaging. Using these polymers from agroindustrial residues to this end are also an interesting strategy to close the wheel of a circular and sustainable economy. These materials have valuable potential in the chemistry and packaging industries, especially due to its renewability, biodegradability, compatibility, as well as good technological and functional properties. Therefore, many challenges related to its processability, proper ways of disposal and legislations need to be overcome to increase their industrial application. It is known that a full replacement of plastics for biobased packaging is not quite easy and is far from being a reality in many countries, though any degree of substitution is welcome to achieve a significant reduction of environmental pollution. This replacement is a challenge to be

overcome by packaging industries, especially because there is lack of legislation about it, the adequacy of industrial processing facilities is costly and the disposal of packaging residues is not well known by consumers or not available to all, especially in low-income countries. Despite those difficulties, replacing petroleum-based plastics should be top priority of all food packaging industry around the world.

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