Abstract: In recent years, food packaging has evolved from an inert and polluting waste that remains after using the product toward an active item that can be consumed along with the food it contains. Edible films and coatings represent a healthy alternative to classic food packaging. Therefore, a significant number of studies have focused on the development of biodegradable enveloping materials based on biopolymers. Animal and vegetal proteins, starch, and chitosan from different sources have been used to prepare adequate packaging for perishable food. Moreover, these edible layers have the ability to carry different active substances such as essential oils—plant extracts containing polyphenols—which bring them considerable antioxidant and antimicrobial activity. This review presents the latest updates on the use of edible films/coatings with different compositions with a focus on natural compounds from plants, and it also includes an assessment of their mechanical and physicochemical features. The plant compounds are essential in many cases for considerable improvement of the organoleptic qualities of embedded food, since they protect the food from different aggressive pathogens. Moreover, some of these useful compounds can be extracted from waste such as pomace, peels etc., which contributes to the sustainable development of this industry.

Keywords: edible films; edible coatings; starch; chitosan; egg white protein; whey protein essential oils; plant extracts; polyphenols

1. Introduction

The food market represents a large part of the global economy and is growing at an alert pace every year. It is a common fact that food waste reaches around 1.3 billion tons each year [1]. An average consumer from Europe or the USA produces about 95–115 kg/year of food waste, and the average consumer from Africa and South/Southeast Asia produces about 6–11 kg/year [1]. These figures represent the ready-to-eat products and do not include the waste generated in the cultivation of plants, animal breeding, and processing steps. Considering the life cycle of food manufacturing, it can be stated that this industry is the most polluting among anthropogenic activities. The main reason for food waste production in less developed countries is the absence or poor infrastructure and technological means for preservation or distribution [2]. The high carbon footprint, which accelerates global climate...
change and all sorts of environmental burdens, is another negative aspect of producing this particular waste. In addition, human health is highly endangered by food spoilage; about 600 million people per year become ill after consuming contaminated food [3]. Another important aspect is the food packaging, which also largely contributes to worldwide pollution by generating a high amount of waste: plastic, paper, metal, glass, etc. Especially plastics of any type and shape are under the scrutiny of public concern due to their ubiquitous spread in all environmental compartments and contamination of the ecosystems. Microplastic is considered an insidious type of plastic pollution [4]. In 2015, more than 300 million tons of plastic waste was generated, and 79% of this was deposited in landfills [5]. It is also estimated that 46,000 plastic pieces are floating on each square mile of ocean, and because of these materials, 1 million sea birds, 100,000 sea mammals, and a large amount of fish are killed. In addition, some packaging contains harmful compounds (e.g., bisphenol A); therefore, a replacement of them is beneficial for human health.

Consumer habits are difficult to change significantly; therefore, developing strategies for diminishing this trend is indisputably necessary. A not-so-healthy alternative to increase the shelf life of food is to introduce preservatives in their recipes. In some cases, this is an inevitable choice, but in most situations, the types and amounts of preservatives exceed the safety rules. Another important approach for addressing simultaneously the packaging load and the food squandering from related industries is represented by edible films that can substitute for the classical packaging, concomitantly with protecting for a longer time any kind of food from adulteration and subsequent disposal to waste. Edible film alternatives are not new in the food industry; they represent a full and mature technology with highly renowned results in terms of efficiency and versatility. In addition, some of the food industry wastes can be successfully used in film preparation, bringing a supplementary reduction of overall waste. Improvements and innovations in the last decade have expanded both the number of applications and the quality of these products [6]. This review will center on the latest work on new edible films and coatings with advanced features, with an emphasis on the utilization of plant extracts containing polyphenols and some essential oil components for enhancing antimicrobial components.

2. Historical Consideration, Definition, Quality Parameters, and Technical Requirements of Edible Films/Coatings

Edible films/coatings are essentially a food package for food. This type of wrapping was used empirically, without scientific knowledge of their role, in food preservation from a long time ago (being documented for 12 centuries in China). In 1922, the method of waxing fruits was introduced to increase their commercial appeal.

As a definition, edible films or coatings are materials used for encapsulating different foods to preserve their properties and could be consumed alongside the food. Films and coatings are sometimes replaceable terms and fulfill the same role, but they represent different concepts. Films are prefabricated by various methods and used to embed food, while coatings are applied on the food surface using a viscous liquid. From the particularities of edible envelopments appear the incontestable advantages of these materials: eco-friendly since they are consumed and, when it is not the case, biodegradable; the solid waste amount is considerably reduced; the organoleptic properties of food are improved, the nutritional properties are enhanced by adding adjuvants; they bring the possibility of wrapping individual items avoiding bulk packaging issues, antimicrobial properties, and the possibility of using a series of byproducts (e.g., agricultural waste) from different activities.

Edible coating and films are used in the same way as any other packaging to preserve the properties of original food, and for that, they have to present some specific features:

- Generally Recognized as Safe (GRAS) labeling or GRAS/FS (some compounds are safe for use in food industry; however, their concentrations is limited by currently used standards). In this respect, the main goal is to avoid toxic, allergic, and/or non-assimilable components [7–15];
- Comply with good manufacturing practices (GMP) [16,17];
• Acknowledgment about a food additive for which a regulation was issued as a result of public statements (e.g., petitions);
• Adequate mechanical properties for preventing the damaging of food surfaces during manipulation from field to supermarket;
• Adherent to food surface;
• Agreeable taste or tasteless;
• Stability in time and especially avoidance of mold development;
• Reduce water depletion of the enveloped product;
• Maintain an adequate gas transfer, especially for oxygen and carbon dioxide and to avoid the loss of components that are responsible for aroma, flavor, and nutritional value;
• Enhancement of structural properties;
• Appearance—overall presentation of the final product requires attaining classical package performances in terms of design. Otherwise, the product can be rejected by consumers;
• Costs—in order to justify a major change in food industry paradigm, the costs need to be lower than other approaches. In some areas, this technology has already attained maturity and the expenses are considerably lower;
• Application devices/methods—distribution of film/coatings formulations on different products in a consistent, efficient, and competitive manner is mandatory. The apparatus used for film preparation must be similar with the classic apparatus. The method of application must be compatible with current equipment;
• Manufacturing processes have to be easy and economically viable. Maintenance and cleaning of the devices used has to be easy to perform.

The difficulties of complying with the above criteria represent the main reason for the small number of companies in the beginning (around 10 in 1986) comparing with over 600 in 1996 [18]. The tremendous progress in the field accomplished in recent years has led to an important increase of businesses related to edible coatings, and the market for these products was evaluated in 2018 to attain around 727.6 million USD with the CAGR (Compound Annual Growth Rate) projected to be 6.2% [19]. The important players in the field are Tate and Lyle, DuPont, Ashland, Koninklijke DSM N.V., Cargill Inc., Devro Plc, Kerry Inc, Innoteq, Watson, Biofilm Limited, ODF Pharma, Proinec, MonoSol, Umang Pharmatech, ProLiant Inc., American Casein Company, The Solae Company, Cargill, etc. There is still more room for growth of this market, since there is a large part of the world (South America, Africa, South East Asia, and Australia) where these products are scarcely extended [20].

3. Building Blocks of Edible Coatings/Films Formulations

Applying edible films on food is somehow a straightforward task, since numerous biomaterials can be used with good results. Edible films contain four components: basic materials, plasticizers, additives, and solvents.

3.1. Basic Materials

The basic materials generally used belong to three categories of natural products: proteins, polysaccharides, and lipids. They come either from vegetal or animal sources and can be used in the pristine state or after a preliminary modification.

3.1.1. Proteins

Proteins as raw materials are extracted from milk (casein, whey), other animal sources (collagen, gelatin), corn (zein), wheat (gluten), soy, eggs (white egg), sorghum, pea, rice bran, cottonseed, peanut, keratin, etc. Proteins are known as natural polymers composed of amino acids in different proportions that vary from source to source.
Protein film formation occurs through the denaturation of the initial material by heat, solvent utilization, and pH. In general, biopolymers are not thermoplastic and are liquefied only by adding some plasticizers. The absence of plasticizers leads to thermal degradation [21]. Temperatures above the glass transition \( T_g \) (Figure 1) undergo the transformation of proteins by molecules disaggregation, unfolding, dissociation, and straightening; molecules are reuniting through other links while the material becomes soft, elastic, and configurable in any shape. The cooled material acquires improved properties and structure due to the new covalent, hydrogen, ionic links formed. Operational parameters such as temperature, plasticizer (type and concentration), pH, etc. are important in film formation.

Some of the most used proteins are:

Gelatin, a protein obtained through the hydrolysis of collagen and other insoluble proteins, is an important candidate for films/coatings production [7,22–33]. It is mainly formed from proline, hydroxyproline, and glycine. Gelatin is used in different mixtures for preserving meat food and even for improving the recipes. Krishna et al. developed films from fish gelatin by extrusion or casting using 20–25% glycerol as a plasticizer at 110–120 °C [34]. Extruded films were more flexible and thicker compared with those formed by casting. Fang et al. used gelatin coatings for reducing the salt amount in sausages, and as a result, preventing the risk of high salt concentration in food for human health [35]. They prepared coating solutions from gelatin–NaCl solutions (1.5–15%) and applied them on non-salted sausages. The original recipe of this product contain 2% NaCl. In terms of pH, color, texture analysis, and sensory evaluation, the coated and not coated products are the same, but the coated product contain overall 50% less salt.

Formulations containing gelatin–chitosan–extracts from grape seed with or without nisin (a polycyclic antibacterial peptide) [22] were used for coating fresh pork chunks to evaluate their usefulness for preserving the freshness. The results obtained permitted establishing the influence and amount required for each component. Hence, the antioxidant properties increased significantly from simple mixtures to complex ones: 1% chitosan < 1% chitosan + 3% gelatin < 1% chitosan + 3% gelatin + 0.5% grape seed extract. Zhang et al. used a combination of gelatin (3%) with ZnO nanoparticles incorporated in ginger essential oil [31], which were applied on beef patties. Some of the mechanical properties (thickness and elongation) increased, while others (tensile strength and gel strength) decreased. Ginger essential oil reduces the water vapor permeability, and lipid oxidation can be avoided.

Gelatin–turmeric combinations improved the elongation at break (EAB) of some edible films [36]. Liang et al., (2017) [37] obtained a homogeneous film from sturgeon skin-derived gelatin in combination with esculine (a coumarin glucoside) as antimicrobial agents. The films have good tensile strength (TS), good water vapor permeability (WVP), and higher antioxidant activity due to esculine, but with an accentuated yellow color and lower elongation at break (EAB).
Mahdu et al., measured the moisture sorption isotherms of sugarcane jaggery portions coated with different edible layers based on whey, gluten, gelatin, zein, and arabic gum at different concentrations (0.5%, 1.5%, and 2.5%) [38], and they applied different models to the experimental data. The Guggenheim–Anderson–de Boer (GAB) model was the best fitting one, and gelatin and gluten gave the best results regarding moisture prevention.

Casein represents a family of phosphoproteins (αS1, αS2, β, κ) that can be found in different proportions in human or animal milk. A complex film produced from casein, alginate, pectin, glycerol, and probiotic Enterococcus faecium Rp1 was developed by Namratha et al., [39]. This film was tested and showed important antimicrobial and antioxidant properties and good physicochemical features for about 60 days, while E. faecium Rp1 was still viable. Regarding the efficacy of films incorporating probiotics against S. aureus, E. coli, and S. flexneri, there was a low survival rate of these bacteria: 20%, 5%, and 10% respectively. In contrast, in the case of probiotic-free films, the survival rate was about 90% for all three microorganisms. The biopolymer coating for processed apples was performed by Volpe et al. [40] using chitosan-caseinate coatings, which increased the shelf life by four days, and the fruit properties were thoroughly preserved. Pella et al., [24] studied the effect on guava fruits of films based on cassava starch, casein, and gelatin with sorbitol (30% w/w) as a plasticizer. The films were prepared using a factorial design (2³, three central points). The fruits remained unchanged for nine days, which could also be observed from harvest index (HI), titratable acidity (TA), and vitamin C measurements. Vitamin C values were initially 98.04%, and after 9 days, they were 85.42% for non-coated fruit and 98.24% for coated products.

Whey is a by-product of cheese manufacturing that consists of the liquid remaining after casein coagulates at a pH value of 4.6 and around 20 °C. Whey proteins present hydrophobic, charged/polar amino acids evenly distributed along the chain in contrast to casein proteins. Therefore, the whey hydrophobic groups are hidden within molecules. These proteins can be used in different commercial purposes, and “edible films” is an important direction with good results, especially since large amounts of dairy products are used constantly. Whey is much more used in the edible film/coatings field, probably because it is a by-product that in most cases needs disposal, since the global cheese production is 24 million tons/year and consequently 21.6 million tons/year of whey [41]. This by-product is ecologically difficult to manage, so the edible films are a good alternative for its use [42–55]. As with casein, whey has to be molded along with several other adjuvants such as chitosan and alginate [42], whey protein concentrate/wheat cross-linked starch composite film [43], Iprovit Bacterial Milk–Yogurt Starter™—sodium alginate—whey—glycerol [52], the microorganism Bifidobacterium animalis subsp. Lactis BB-12—whey protein isolate—alginate [44], whey—glycerol (30–60% w/w of whey) [45], maltodextrin–Arabic gum—whey [53], green tea/rosemary extract—whey [46], whey—glycerol (5–15% w/w)—rosemary and thyme extracts [47], etc. It was found that the produced films act as a barrier for different microorganisms [42,46,50–52,54] and present improved physicochemical properties [42,43,53,55].

Egg white is a complex mixture of globular proteins [56]: ovomucin, ovotransferrin, ovalbumin, ovomucoid, lysozyme, G2 globulin, G3 globulin, and avidin. The content of the main constituents from total proteins are ovalbumin—54%, ovotransferrin—12%, and ovomucoid—11%. For edible film preparation, the –SH groups, which exist only in ovalbumin and the S–S bonds encountered in lysozyme, ovomucoid, and ovotransferrin are very important in reticulation processes. Mixtures of pullulan (Pu)—egg white (EW)—glycerin (Gly) [57] lead to the formation of edible films with superior properties due to hydrogen bond rearrangements. These changes were assessed by Fourier-transform infrared spectroscopy ( Nicolet 380 FT-IR Spectrometer, Thermo Fisher Scientific, Waltham, MA, USA), XRD (D8 Advance, Bruker AXS, Karlsruhe, Germany), secondary structures, and free amino group analysis. The highest values of the TS (329.48 MPa) and EAB (10.33%) were obtained for the Pu:EW = 1:1 ratio. Increased TS can also be obtained by adding oleic acid probably due to the increased negative charges and exposure to –SH groups [58], while the use of dialdehyde starch increases water solubility and mechanical strength [59]. Alkaline pH (10–12) is required for homogeneous films [60], while
UV radiations increase the reticulation of polymer chains, resulting in a low solubility and improved mechanical strength [61]. Sothornvit et al., [62] prepared three types of edible films from soy, egg white proteins, and whey proteins, respectively. Whey films presented a better water vapor permeability compared with egg white films. In another paper [63], edible films were prepared using egg white–succinylated casein with and without transglutaminase (TGase) treatment. Films treated with TGase (concentrations of 15 U/g protein) showed maximum mechanical strength and lower values of WVP. Another interesting combination for edible film preparation was obtained using egg white and κ-carrageenan [64]. When the percentage of egg white increased, the EAB and light transmission improved to 10.85% and 53.3%, respectively. In addition, in another paper [65], egg white lysozyme was incorporated into zein–glycerin films to obtain food packaging with antimicrobial properties against B. subtilis and L. plantarum. Moreover, the films containing in addition disodium salt of ethylenediaminetetraacetic acid (EDTA) are efficient against E. Coli. Its antimicrobial feature is manifested even at low lysozyme concentrations. Aguilar et al. [66] produced edible films and microparticles by ionic gelation using pectin–alginate blend followed by electrostatically coating with whey or egg white proteins. A thorough characterization of films and particles was made in terms of calcium content, thickness, adsorbed protein, moisture, WVP, structure, and mechanical properties. As the authors suggest, the difficult part of the synthesis was obtaining a homogeneous drying of the films; this aspect require further studies.

3.1.2. Polysaccharides

Polysaccharides represent, besides proteins, another important pillar of the biological kingdom. These compounds are ubiquitous and therefore compatible with the natural products that are supposed to be protected by edible films/coatings. Moreover, the possibility of chemically modifying this natural and biodegradable polysaccharide increases significantly the range of compounds eligible for biocoatings. The main polysaccharides used to produce edible barriers are starch (natural or modified) [67], modified cellulose (carboxy methylcellulose [68], hydroxypropylcellulose [69], hydroxypropylmethylcellulose [70]), inulin (Jerusalem artichoke) [44], sodium alginate (brown seaweed) [71], chitosan (crustacean shells deacetylation) [72], pectin [7,73], carrageenan [64], pullulan [74], gellan [75], xanthan [76], etc.

Starch (Figure 2) is an abundant natural compound that is present in all plant components (stem, seeds, fruits, roots, etc.). It is the plant “battery” since it represents energy storage in chemical form, but also the main energy source for animals and humans (60–70% of the human caloric intake comes from starch). Starch consists of two different polysaccharides: amylose composed of D-glucose residues with α-(1→4) linkages—linear and amylopectin formed and α-(1→4) linkages and approximately 6% α-(1→6) linkages as branches from the parent molecule. Both are comprised of water-insoluble granules with different shape, morphology, and crystallinity for different sources. Starch possesses thermoplastic characteristics in the presence of plasticizers, high temperatures, and mechanical pressing. In this respect, starch is similar to other synthetic polymers; hence, the operation implemented to other polymers can be applied to starch also. Therefore, to give starch a new shape, a thermomechanical process is needed (extrusion, injection, kneading, molding, casting, all in the presence of heat, water, and appropriate plasticizers). A novel and efficient approach [77] to obtain thermoplastic starch consists of using sodium hexametaphosphate (SHMP)/polyvinyl alcohol fibers (PVAF) as cross-linking agents and the following procedure: SHMP and PVAF are mixed in solution, starch and glycerol were added to solution, the mixture was submitted to extrusion, granules were obtained, and granules were molded in films. In this process, SHMP produces cross-linking between starch molecules and PVAF.
films/coatings formulation. There are several advantages to use starch, such as its low cost, large availability, non-toxic nature, and versatility in processing (as flexible as polyethylene and rigid as polystyrene), but it also has disadvantages. For example, raw starch is brittle, too hygroscopic, and has low mechanical properties. To eliminate these drawbacks, plasticizers can be used, and different sources of starch can be taken into consideration.

Colombian native potatoes: pacha negra, mora, and alcarrosa. For all three types of starch, the reduction in gas exchange was found to be approximately 27%. Corn starch, with hydroxypropyl methylcellulose, \textit{Uncaria gambir} extract, and glycerol were used as the main ingredients for edible film preparation [94]. To assess the influence of gambir extract to edible film parameters, an experimental design and statistical analysis were used. It was observed that the optimal operational parameters are a 40% concentration of \textit{Uncaria gambir} and 1000 rpm centrifugation for separation.

Andean blueberries [93] were protected using starch–glycerol films, the starch source being Colombian native potatoes: pacha negra, mora, and alcarrosa. For all three types of starch, the reduction in gas exchange was found to be approximately 27%. Corn starch, with hydroxypropyl methylcellulose, \textit{Uncaria gambir} extract, and glycerol were used as the main ingredients for edible film preparation [94]. To assess the influence of gambir extract to edible film parameters, an experimental design and statistical analysis were used. It was observed that the optimal operational parameters are a 40% concentration of \textit{Uncaria gambir} and 1000 rpm centrifugation for separation.

Other authors prepared films based on arrowroot starch–cranberry powder (0–55%) [95]. Arrowroot (\textit{Maranta arundinacea} L.) was selected due to it containing good starch properties: high amylose content, digestibility, and gelling ability. Cranberry powder was selected due to its large amount of anthocyanins, proanthocyanidins, organic acids, glycosides, flavonoids, phenolic acids, and ascorbic acids, which confer remarkable antioxidant properties to prepared films. Despite their beneficial activity, increasing the cranberry content to over 45% leads to brittle, rough, and irregular films.

In an experimental design [67] with two factors—starch type (cassava, arrowroot, and canna) and starch percentage of (3%, 4%, 5%) (v/v)—the quality of prepared edible films was assessed. Sorbitol was used as a plasticizer. In terms of tensile strength, \textit{Canna edulis} showed higher values, while for elongation, arrowroot gave better results. It is not surprising, since these two parameters are inversely proportional. For another important parameter, WVP, the authors found lower values in case of arrowroot-type starch for all experimental concentrations. Therefore, in this paper, it was shown that the starch type is paramount for the film quality.
Another interesting starch provenience is elephant foot yam (EFY), which is one of the natural rich sources of starch. Nagar et al. used starch from this tropical tuber crop to obtain edible films in conjunction with hydrocolloids—xanthan (XG) and/or agar–agar (AA) [96]. It was found that the WVP values decreased with increasing concentrations of AA and XG. The hydrophilic nature of hydrocolloids facilitates water molecules to bond with polymer chains and form microcavities. Hence, at diminished content of agar–agar (AA) and/or xanthan (XG) the films present higher WVP (suitable for packaging where moisture is needed) and for higher AA and/or XG content the films present reduced WVP (when sealing is required).

The protection of Huangguan pear (Pyrus pyrifolia), which is a new species with superior properties (larger, smoother, sweeter) was made through a new approach first reported in the literature: edible films prepared from cross-linked (with adipic acid) cassava starch reinforced by starch nanocrystals (SNCs) [97]. The authors prepared the films for parameter testing, and the same solutions were used for coatings. The study involved also the impact of grading operations previous to coating, since these fruits are very sensitive. The results showed that coatings containing 6% SNC presented the best performance, and the fruits were significantly protected, especially when a graded process occurs after coating.

Chitosan (Figure 3) is a well-known polysaccharide derived from chitin with countless uses due to its properties [98–100]: it is a natural linear polymer that is non-toxic and therefore safe for the food industry, biodegradable, antitumoral, fungistatic, hemostatic, anticholesteremic, antioxidant, antacid, colon targeting, analgesic, etc. Chitosan is mainly insoluble in water, but it becomes soluble in acidic solutions (acetic acid, formic acid, etc.). If the deacetylation degree exceeds 50%, the solubility increases, too. When chitosan is introduced in the acidic media, the chain-linked amino groups become protonated, and the chitosan shifts into a cationic state. Hence, positive charged chitosan acquires antimicrobial properties since it interacts with the negative surface of cell membranes, leading to bacteria deactivation.

![Chitosan Structure](image)

**Figure 3.** Comparison among cellulose, chitin, and chitosan structures.

The diverse features of chitosan make it a good candidate for the preparation of edible coatings and films, solely or in various formulations. Chitosan edible film coatings were also investigated for the protection of food qualities of sweet cherry (Prunus avium L.) [101]. Several parameters of sweet cherry such as its total carbohydrate content, titratable acidity, total soluble solids, water activity, and pH were preserved using chitosan coatings. The authors used four types of chitosan: two prepared by themselves from shrimp wastes with different deacetylation degrees and two commercial ones. The chitosan prepared by reaction with 40% sodium hydroxide at 120 °C for 300 min (CH-1, deacetylation 78.2%, 182 kDa) provided a longer shelf life for studied fruits.

**Pectin** is an important polysaccharide considering the structural role in plant tissues. Pectin’s biological function is to reticulate the cellulose and hemicellulose fibers, producing a more resistant structure. It is found in almost all plants in different concentrations mainly in the middle lamella layer between cells (Figure 4). Pectin is a generic name for a family of polymers which differ in terms of molecular weight, chemical configuration, and types and abundance of monosaccharide subunits:
Pectin is an important polysaccharide considering the structural role in plant tissues. Pectin’s structural role is to reticulate the cellulose and hemicellulose fibers, producing a more resistant biological function. It is found in almost all plants in different concentrations mainly in the middle lamella layer between cells (Figure 4). Pectin is a generic name for a family of polymers which differ in terms of molecular weight, chemical configuration, and types and abundance of monosaccharide subunits: (rhamnogalacturonan I, rhamnogalacturonan II, and homogalacturonan) [102]. In a simply way, pectin is a chain of alpha-1,4-linked galacturonic acid subunits (Figure 5) that have an “accordion-like” conformation which result in molecules with extensible characteristics.

Due to the significant role in plant biology and already being an important ingredient in the food industry, pectin is a good candidate for edible films coatings/preparation. Pectin type E440i (contain different levels of methyl esters on polygalacturonic acid chain) and E440ii (contain amides groups along with methyl esters groups) are largely used in the food industry. Regarding the safety of this compound, the European Food Safety Authority Panel agrees with the fact that this compound lowers the cholesterol level of adults if the daily dose reaches 6 g (for infants, there are no data available).

Due to the significant role in plant biology and already being an important ingredient in the food industry, pectin is a good candidate for edible films coatings/preparation.

Natural sources of pectin with different extraction yield are represented by different agricultural wastes: apple pomace (4.2–19.8%), citrus peel (13.4–37.52%), sugar beet pulp (23–24.87%), tomato waste (7.55–32.6%), mango peel (17.15%), watermelon rinds (19–21%), etc. Extraction is made in hot acidic (pH = 1.5–2) water solution followed by vacuum concentration and precipitation with different agents (mostly alcohols). Microwave, ultrasound, and supercritical-assisted extraction processes are used for pectin separation [105].

Tumbarski et al., used celery-based pectin (1%) alone or in combination with bacteriocin (Bacillus methylotrophicus BM47) for preparing a coating for blackberries preservation [73]. Comparing with a
control (not-coated fruits), the coating preserved all the initial properties of the blackberries (loss of weight, decay, total soluble solids, titratable acidity, pH, organic acids, sugars, total phenolic content, total anthocyanins, and antioxidant activity). Ascorbic acid is one of the most important parameters, and it remains at the same level compared to the initial moment for 16 days (57.5 mg/100 g for the pectin-based coating and 58.8 mg/100 g for pectin and bacteriocin).

Ngo et al., conducted a study in which they used pectin and nanochitosan (in different ratios) and tried to elucidate the influence of each component and ratio between them on the mechanical and barrier properties. The results showed that a ratio of 50:50 obtained the best results: tensile strength of 8.96 MPa, water solubility decrease to 37.5%, water vapor permeability (WVP) to 0.2052 g·mm/m²·day·kPa, and oxygen permeability (OP) to 47.67 cc·mm/m²·day [106].

Yeddes et al., prepared gelatin–chitosan–pectin edible films improved with rosemary essential oil [7]. The preparation process was optimized using a Doehlert matrix (an advanced response surface methodology superior to other design, such as composite central and Box–Behnken) [107]. Hence, the mechanical and textural properties were improved, and the optimal composition was found to be 10% chitosan, 24.3% gelatin, 65.2% glycerol, and 0.5% pectin, while the antioxidant activity was enhanced for the addition of 1.996 mg/g rosemary oil.

3.1.3. Lipids, Waxes, and Resins

Lipids (saturated, monounsaturated, polyunsaturated fat acids) [108], waxes [68] (Figure 6), and resins (wood rosin, shellac) [109] represent another important area of edible coatings/films either as primary components or as plasticizers. Shellac is produced by insects Laccifer lacc a and contains a complex mixture of aliphatic alicyclic hydroxy acid polymers i.e., aleuritic, bucolic, shellolic, and jalaric acids).

![Figure 6. Main types of compounds included in waxes: hydrocarbons (n-alkanes where n = 22–36), fatty alcohols (R length is 12–34 carbon atoms), fatty acids (R length is 12–34 carbon), long-chain esters (R₁ and R₂ = 10–20 carbon atoms in length).](image)

3.2. Plasticizers

Plasticizers choices encompass a large range of molecules: water, xylitol, mannitol, propylene glycol, glycerol, sorbitol, polyethylene glycol, sucrose, polypropylene glycol, triethylene glycol, ethylene glycol, corn syrup, 1,4 butane diol, 1,6 hexane diol, triacetin, glucose, urea, diethanolamine, dibutyl phthalate, tributyrate, tributyl citrate, diethyl tartrate, acetylated monoglycerides, fatty acids (oleic, stearic, lauric, etc.), lactic acid, deep eutectic solvents [110], etc. Some of them are removed and others are introduced in the manufacturing process in accordance with updated food regulations [111,112]. Plasticizers are generally required in a proportion of 10% to 65% as a function of polymer rigidity. They improve the process of polymer formation, and for a final product (films or coatings), these molecules allow a larger temperature range for their use, impart flexibility, lower the brittleness, and increase the hardness of the film. In addition, the main role of...
these compounds is to diminish the intermolecular forces between polymer chains (Figure 7), which increases the free volume and movement of the polymeric chains.

![Figure 7. Activity of plasticizer between protein chains.](image)

As for the classification of these molecules, there are two types of plasticizers [113]: internal and external, which is terminology taken from polymer science. Internal plasticizers (co-polymers or compounds that interact with polymer chain) increase the steric hindrance of chains followed by increased free volume and flexibility. As a consequence, the glass transition temperature ($T_g$) and elastic modulus (EM) decrease, while the whole polymer become softer. External plasticizers do not react with polymer chains; they act on protein chains by solvation and lubrication, and also as agents to lower the glass transition temperature and increase free volume. The most important features of plasticizers are compatibility, efficiency, and permanence. Compatibility means that plasticizers have a similar chemical structure, and they are also low volatility, non-harmful, and free of aromas. Efficiency is attained if the plasticizer fulfills its role at lower concentration and presents a higher diffusion capacity into the polymer matrix. We encounter in this matter a delicate balance between diffusivity and volatility; therefore, the size of the molecules involved is crucial. The permanence of plasticizers is also related to their molecule size, but likewise, polarity and hydrogen bonds play an important role. The correlation of plasticizers amount and type with $T_g$ variation is less easy to obtain for natural polymers, since biopolymer chains are less mobile due to chain interactions, hydrogen bonding, ionic interactions, and S–S bonds. Plasticizers generally bring superior mechanical properties to the mixture, but their hygroscopic properties (for most of them) increase the water vapor permeability and decrease the blocking properties of coatings for gases, moisture, and aroma compounds.

3.3. Additives

Additives play many important roles in the film/coatings matrix and include various compounds such as antioxidants, nutraceuticals, pharmaceuticals, antimicrobials, nutrients, flavors, fragrances, colors, probiotics, emulsifiers, cross-linking agents, browning inhibitors [114–116], etc.

Antimicrobial molecules are used in order to prevent the decaying of foodstuffs. Commonly used agents are benzoic acid, potassium sorbate, propionic acid, sodium benzoate, clove bud oil, sorbic acid, etc. The antimicrobial properties come from the ability of undissociated organic acids molecules to penetrate bacterial walls and disrupt the cell’s development. Other antimicrobial components of natural sources are grapefruit seed extracts, allyl isothiocyanate, hinokitiol, nisin, pediocin, natamycin, and reuterin (Figure 8).
Antioxidants and antibrowning agents retard discoloration, rancidity [53], and the degradation of food. The most commonly used are ascorbic acid, scorbyl palmitate, butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT), citric acid, propyl gallate, tertiary butylhydroquinone (TBHQ), tocopherols, Maillard reaction products, rosemary, and sage extracts. Maillard compounds (hundreds of them) result from a sequence of non-enzymatic reactions (Figure 9), which start from sugars and an amino group (typically from an amino acid or protein) through room temperature to approximately 200 °C. These compounds are very good antioxidants, but they are usually associated with cooking processes (some of them) such as the flavors, aroma, and brown color released by baked food. Essential oil contains mainly terpenic compounds that are responsible for the antibacterial and antioxidant properties of films: monoterpenes (limonene, tricyclene, α-thujene, α-pinene, camphene, sabinene, β-pinene, β-myrcene, 3-carene, α-terpinene, α-phellandrene, p-cymene, v-terpinene, α-terpinolene), oxygenated monoterpenes (1,8-cineole (Z)-sabinenehydrate, linalool, d-fenchylalcohol, α-campholenal, camphor, borneol, 4-terpineol, α-terpineol, bornylacetate), sesquiterpene hydrocarbon (α-copaene, aromadendrene α, (E)-caryophyllene, alpha-amorphene, γ-cadinene, δ-cadinene), oxygenated sesquiterpene (caryophyllene oxide, α-eudesmol) [12,117–126].
These molecules comply with the Code of Federal Regulations; they are GRAS, GRAS/FS, GMP, and are very effective, but their use is subject to controversy in the food industry, and replacements are needed (except for the natural product, and even for them, the use is allowed only after a careful evaluation of effects).

Nutrients, flavors, and colorants such as calcium lactate [127], vitamin E [128], and beta carotene [129] can be embedded in coatings/films and bring an important improvement of food quality protection.

Cross-linking agents (formaldehyde [130], glutaraldehyde [131], transglutaminase [63], citric acid [132], genipin [133], lactic acid [52], tannic acid [134], ionized calcium, or UV radiation) enhance the reticulation of biopolymers, resulting in significant changes of the properties: decreased solubility, increased mechanical resistance, etc.

Emulsifiers are surface-active agents that allow a good dispersion of precursors in order to obtain and stabilize protein/lipid or polysaccharide/lipid composites and improve their adherence to food surfaces. Adding emulsifiers, the hydrophilic–lipophilic balance of mixtures can be tailored according to the required lower water vapor permeability (WVP). Common emulsifiers used in the food industry are acetylated monoglycerides, ammonium lauryl sulfate, ethylene glycol monostearate, glycerol monostearate, oleic acid, linoleic acid, potassium oleate, propylene glycol monostearate, sodium alkyl sulfate, sodium oleate, sorbitan monostearate (Spans), lecithin, stearic acid, sucrose stearate, polyoxyethylene (20), sorbitan monolaurate (Tween 20), etc.
3.4. Solvents

Solvents are the media for dispersing all films/coatings components and need to be adapted to the chemical nature of these ingredients. Aqueous solutions of ethanol with different concentrations are generally used, since they can provide an adequate solvation power, a lower microbial growth, and a rapid drying process after application. Examples of alcohol-soluble proteins are wheat gluten and fish myofibrillar proteins. The hydration of protein facilitates their solubilization. Rarely, acetone or isopropanol can be used, but more often, acidic solutions (acetic and lactic acid) at lower pH (2.5–3) are employed for some proteins or chitosan (insoluble in neutral solutions). Other compounds such as cotton seed proteins require highly alkaline solutions (pH = 8–12), temperature 20–60 °C, and solvent content in the 10–50% range.

3.5. Plant Extracts

The whole picture of films/coatings composition is far from complete, and the quest for new improvements is ongoing. In recent years, the utilization of plant extracts in edible formulations has increased at a rapid pace. The reason for this is the complex composition of extracts that mimic the properties of different additives already in use. Moreover, the high selection basis of plants and solvents, development of advanced extraction, separation and purification techniques, and/or analytical methods allow the selection and design of customized fractions from initial concentrate. A special case is Centrifugal Partition Chromatography (Figure 10), which avoids the use of solid phase separation columns (which are expensive and prone to degrade sensitive compounds) with a rotor of special construction that allows the simultaneous utilization of a liquid stationary phase and a liquid mobile phase [105,135–137]. Table 1 presents various applications of plant extracts for a wide range of foods.

| Product   | Preserved Plant Extract Film/Coating Base References |
|-----------|-----------------------------------------------------|
| Fish      | Red radish anthocyanins extract gelatin/gellan gum [3] |
| Rosemary  | essential oil—enriched films Rosemary essential oil glycerol, gelatin, chitosan, pectin [7] |
| Trout fillet | Zataria multiflora Boiss essential oil Alginate coarse/nanoemulsions [9] |

**Figure 10.** Centrifugal Partition Chromatography: a preparative scale method for plant extract fractionation.
Table 1. Application of plant extracts in food preservation.

| Product Preserved | Plant Extract | Film/Coating Base | References |
|-------------------|---------------|-------------------|------------|
| Fish | Red radish anthocyanins extract | gelatin/gellan gum | [3] |
| Rosemary essential oil-enriched films | Rosemary essential oil | glycerol, gelatin, chitosan, pectin | [7] |
| Trout fillet | Zataria multiflora Boiss essential oil | Alginate coarse/nanoemulsions | [9] |
| Table grapes (Vitis vinifera L.) | Thymus vulgaris essential oil | Pullulan and polymeric nanocapsules containing essential oil | [12] |
| Edible films | Nitrile and garlic essential oil | Gelatin-chitosan | [28] |
| Meat | Ginger essential oil | Microemulsion nanofilms: Tilapia fish skin gelatin and ZnO nanoparticles | [31] |
| Edible films against food spoilers and foodborne pathogens | Oregano, clove, tea tree, coriander, mastic thyme, laured, rosemary, and sage essential oils | Whey protein isolate | [50] |
| Edible films | Yerba mate extract | Cassava starch | [86] |
| Edible films | - | Water chestnut starch-chitosan | [88] |
| Edible films | Thyme essential oil/apple skin polyphenols | Acai puree, pectin | [116] |
| Lamb meat | Thyme and garlic essential oils | Alginato | [117] |
| Tomato | Sage essential oil | Aloe vera gel | [118] |
| In vitro and in the food model (polenta) | Caraway and juniper essential oils | - | [119] |
| Ready-to-cook barbecue chicken | Ginger essential oil and citric acid | Cellulose nanofibers coating | [120] |
| Papaya (Carica papaya L.) | Mentha essential oils | Chitosan | [122] |
| Rainbow trout fillets | Lemon and sage essential oil | Quinoa | [125] |
| Pork loin | Oregano essential oil, novorotrol nanoemulsion | Pectin | [130] |
| Rainbow trout fillets | Ferula angulata essential oil | Chitosan | [139] |
| Soybean oil | Hyssop (Hyssopus officinalis L.) extract | biopolymer Nanoemulsions of Lepidium perfoliatum and Orchis mascula | [140] |
| Beef | Myristica fragrans essential oil | Agar | [141] |
| Rainbow trout | Lemon verbena extract/essential oil | Chitosan | [142] |
| Turkey meat | Zataria Multiflora Boiss and Bunium persicum Boiss essential oils | Chitosan | [143] |
| Antibacterial films | Rosmarinus officinalis L. extract | Methylcellulose | [144] |
| Cucumber | Cinnamon essential oil and cinnamaldehyde | chitosan | [145] |
| Goa fruit | Cinnamon essential oil | Gum Arabic, oleic acid | [146] |
| Papaya | Clove essential oil | Manise starch | [147] |
| Beef | Cumin essential oil | Shahri Baland seed mucilage | [148] |
| Edible film | Clove essential oil | Millet starch | [149] |
| Pork chops | Free or nano-encapsulated Paulownia Tomentosa essential oil | Chitosan | [150] |
| Tambakoi (Colossoma macropomum) fillets | Clove essential oil | Chitosan | [151] |
| Apple, tomato, and persimmon | Lechiitin-encapsulated thyme essential oil | Starch-gellan | [152] |
| Edible films | Cupuassu (Theobroma grandiflorum) Puree | Combining, pectin, and chitosan nanoparticles | [153] |
| Edible films | Extracts of green apple (Granny Smith) skin | Methylcellulose | [154] |
| Edible films | Oxidized ferulic acid | Yuba, carboxymethyl cellulose, beeswax, sodium pyrophosphate | [155] |
| Chicken breast fillets | Ginger (Zingiber officinale) essential oil | nano emulsion-based edible coating containing | [156] |
| Gaava (Psidium guajava L.) | Ruta graveolens Essential Oil | Chitosan | [157] |
| Mango cultivar Tommy Atkins | Mentha piperitae L. essential oil | Chitosan | [158] |
| Banana | Clove essential oil titratable | Cassava starch, polyvinyl polychloride | [159] |
| edible films | Microencapsulation of Thai rice grass extract | Carboxymethyl cellulose | [160] |
| edible films | Aloe vera gel | Banana starch-chitosan | [161] |
| Meat products | Asparagus racemosus | Calcium alginate edible film | [162] |
| Edible films | Macadamia skin (Macadamia tetraphylla), banana peel, blueberry ash extracts | Pea starch-guar gum BioComposite | [163] |
| Nile tilapia fillets | Pomegranate peel extract as edible coating | Chitosan | [164] |
| Edible films | Basil essential oil | Fish skin gelatin, palm oil, different surfactants | [165] |
| Tomatoes | Oregano (Lippia graveolens) essential oil | Pectin | [166] |
### Table 1. Cont.

| Product Preserved          | Plant Extract                  | Film/Coating Base               | References |
|----------------------------|--------------------------------|---------------------------------|------------|
| Kashar cheese              | Orange essential oil          | Egg white protein               | [167]      |
| Ground beef patties        | Caesalpinia decapetala and Caesalpinia spinosa (Tara) extracts | gelatine                        | [168]      |
| Edible films               | Ginger essential oil nanoemulsion | Gelatin, montmorillonite      | [169]      |
| Edible films               | Hydrolysable chestnut tannin  | Pigskin gelatin films           | [170]      |
| Organic leafy greens in sealed plastic bags | Carvacrol and cinnamaldehyde | Apple, carrot, and hibiscus    | [171]      |
| Sliced bread               | Clove bud (Syzygium aromaticum/origanum (Origanum vulgare) nanoemulsions | Methylcellulose               | [172]      |
| Apples                     | Satureja hortensis L. extracts | Pullulan                        | [173]      |
| Edible films               | Grape pomace extract          | Chitosan                        | [174]      |
| Edible films               | Ginja extract cherry           | Methylcellulose                 | [175]      |
| Edible films               | Extracts of chives (Allium schoenoprasum), sage (Salvia prionitis, Lamiaceae), European elderberry (Sambucus nigra, Caprifoliaceae), dandelion (Taraxacum officinale) | Atelocollagen   | [176]      |
| Edible films               | Red raspberry extract          | Soy protein isolate             | [177]      |
| Ham                        | Carvacrol and cinnamaldehyde  | Pectin-based apple, carrot, and hibiscus | [178]      |
| Edible films               | Zataria multiflora Bois essential oil /grape seed extract | Chitosan                     | [179]      |

## 4. Preparation and Characterization of Edible Films/Coatings

### 4.1. Techniques for Preparing Edible Films/Coatings

The preparation phase has to follow some previously tested formulations for obtaining products with the desired properties (strength, solubility, flavor, texture, strength, compatibility between components, film stability, etc.). When new components or different concentrations of additives are introduced, the features of coating/films mixtures can change significantly. In order to attain the optimal configuration with the minimum number of trials, some advanced statistical methods (response surface methodology) are implemented. Challenges encountered in preparing and using films/coatings generally are as follows.

- Using essential oil in the formulation due to the low miscibility in protein or polysaccharides solution. For this problem, an adequate emulsifier is used in an appropriate concentration;
- The casting method is not so easy to apply at the industrial level compared with the extrusion method. In this respect, maintaining the required concentration of essential oil in the film is an aspect that requires further studies;
- Obtaining a complex material at a low price with high resistance to surrounding factors;
- Choosing the right material for film preparation from a wide range of options.

Film/coatings are formed as a result of a multitude of chemical and physical processes blended in a manner that assures a suitable output. An important aspect is to figure out in what order the ingredients are added, which include the temperature regime and pH of the solution. The process of mixing usually starts with water and surfactants followed by the main ingredients (proteins, polysaccharides, lipids) and active ingredients (antimicrobial, antioxidants, etc.). Finally, water is added for adjusting components’ concentrations, viscosity, pH, etc.

Different techniques are used for the efficient production of edible barriers. To a certain extent, the preparation of films and coatings follows the same path: obtaining the complex mixture, homogenization, degassing, and stabilization. In the case of films, the first step is to process the liquid by casting (the liquid is poured on solid surfaces), lamination, or extrusion. The second step is drying carefully until the material reaches a certain values of humidity, and the final step is application on food. In the case of coatings, the liquid is dispersed on food surfaces in different ways: spraying, dipping, brushing, panning, fluidized bed, foaming, etc. The process parameters are thoroughly controlled in order to obtain the desired products and are related with the food characteristics (temperature, shape,
size, diameter variation, hygroscopicity, surface tension) or to the coating liquid itself (solvent type, composition, viscosity, surface tension, etc.). The food particle size and shape are the main factors that decide the choice of method (Figure 11).

![Figure 11. Influence of material size on coating selection techniques.](image)

All these materials combined give a final product with enhanced properties. There are also coatings in a multi-component or composite manner, which are based on complementary or synergistic approaches. Usually, there is some association between hydrophobic and hydrophilic structures. These are prepared by two methods: (1) superposing two layers—hydrophobic on top of a hydrophilic layer or (2) emulsion-type composite lipids dispersed in a hydrophilic support. Layer-by-layer deposition (LbL) permits better control over the features of edible coating; especially, its thickness and is more adapted to automated manufacturing [180]. The ionic self-assembly method is based on successive layers of positively charged polymers (chitosan) with negatively charged polymers (polyanionic cellulose, pectin, etc.).

### 4.2. Characterization of Edible Films/Coatings

The characterization of edible films/coatings is a complex process, since it addresses a large range of features and is intended to give a thorough overview of the product's qualities. The diversification of characterization methods is necessary since in some cases, introducing different ingredients in certain parameters can lead to antagonistic variations: for example, when the tensile strength (TS) increases, the elongation at break (EAB) decreases.

After preparation, the product's properties are thoroughly evaluated by measuring the physicochemical and biological parameters, such as water vapor permeability (WVP), elastic modulus (EM), elongation at break (EAB), microstructure, optical properties, antioxidant and antimicrobial activity, etc. Adequate values of these parameters assure to some extent the preservation of the food's original properties.

#### 4.2.1. Wettability of Coatings Formulations on Food Surface

An important aspect of edible coatings is their compatibility in terms of wettability [181,182] between the formulation and the food surface. Mismatch between coating formulation and surface results in inconsistencies in film properties (thickness and uniformity). In most cases, the adhesion of coatings to the food surface is inadequate due to chemical differences, which generally increase when...
the coated object consist of slices of fresh cutted fruits. The fast adsorption of the liquid included in formulations on fruit surfaces prevents the formation of an edible barrier. Improving the wettability requires the surface tension of the solid surface to be similar to that of liquid, and matching those usually requires surfactants such as Tween 80. When a liquid is spread on a solid surface (Figure 12), the spreading coefficient ($W_s$) is close to zero, meaning that the liquid is optimal for covering. The wetting capacity of the liquid depends on the right balance between the adhesion coefficient $W_a$ (accounting for forces that expand the liquid on the surface) and the cohesion coefficient $W_c$ (accounting for forces that keep liquid contracted).

$$W_s = W_a - W_c = \gamma_{SV} - \gamma_{LV} - \gamma_{SL}$$

$$W_a = \gamma_{LV}(1 + \cos \theta)$$

$$W_c = 2\gamma_{LV}$$

The contact angle results from interfacial tensions: $\gamma_{SV}$ (solid–vapor), $\gamma_{LV}$ (liquid–vapor), $\gamma_{SL}$ (solid–liquid). Values of $\theta < 90^\circ$ show that the liquid is wetting the surface, while values above $90^\circ$ show the opposite. When $\theta$ is near zero, the liquid wettability is at its maximum. Contact angle measurements are influenced by factors such as droplet size, temperature (maintained up to 8 °C), surface roughness, and surface impurities. Generally, $W_c$ can be obtained from the contact angle and surface tension values. The contact angle is determined by the sessile drop method [183–186] (a droplet on the food surface is measured by an optical system equipped with a high-performance video camera and a software) or by the immersion method [187] (using a tensiometer).

An important parameter in the physicochemistry of surfaces is surface tension, which stands as the amount of energy required to increase the surface per area unit (J m$^{-2}$). Surface tension ($\gamma_{LV}$) is determined using the De Noüy platinum ring method.

Surface tension is a measure of adhesion between liquid and solid and is calculated from the contact angle measurements of a standard liquid on the surface. Another important parameter is the critical surface tension [152,181] of the food to be coated. This parameter is obtained using the extrapolation of Zisman plot [152] ($\cos \theta$ against the surface tension of different liquids on the studied surface) until intercept of the curve at $\cos \theta = 1$.

Therefore, for a good compatibility (liquid–solid), two conditions must be fulfilled: wettability and adhesion.

Sapper et al., [152] evaluated the influence of coating-forming liquids composition on the wettability/spreading coefficient, contact angle, and surface tension values. In addition, the interaction of these liquids with persimmon, tomato, and apple surface fruits was assessed. The liquid coatings formulations used were obtained from different compounds: starch–gellan, starch–gellan with emulsified essential oil, and starch–gellan with lecithin-encapsulated essential oil. To these formulations were added different concentrations of Tween 85. It was found that in the absence of essential oil in the liquid coatings, Tween 85 has a beneficial effect, improving the spreading coefficient (values almost zero). The addition
of essential oil also shows a good impact on surface tension, but not with Tween 85 in the formulations. As can be seen from Figure 13, the contact angle decreases at higher Tween 85 concentration, which shows the beneficial effect of surfactant. The influence of the food surface nature is emphasized by the lower values of contact angles of persimmon compared with apple and tomato.

![Graph](https://example.com/graph.png)

**Figure 13.** Variation of $\theta$ with Tween 85 concentration for three coating mixtures.
Ortiz et al. [183] observed that in a formulation containing chitosan, glycerol (10%), and Tween 20, the \( W_s \) coefficient increases from \(-28.24\) to \(-20.63\) when the Tween 20 content increases from 5\% to 15\%. Gelatin-based films prepared with mint essential oil (0\%, 0.06\%, 0.13\%, 0.25\%, 0.38\%, 0.50\%) present a significant improvement of coating properties in terms of hydrophobicity (the contact angle of the water on the coating surface increases from 49.0° to 63.1°).

4.2.2. Film/Coating Thickness, Mechanical, and Gas BARRIER properties

Film/coating thicknesses generally have a large variability in a range from 2 to about 3000 \( \mu \)m [188] depending on application/preparation techniques and the composition of formulation. Film thicknesses are measured with digital micrometers in several repetitions and at different positions.

The incorporation of clove essential oil (0.8\% \( \text{w/v} \)) increases the thickness from 43 to 51 \( \mu \)m. This is attributed to the higher free volume of the film [159]. The same situation was observed by Han et al. [189] when cinnamon essential oil was added to into sodium alginate/carboxymethyl cellulose. Another beneficial effect of essential oil is that film rigidity decreases and the mechanical properties are improved [124]. However, higher values of thickness often lead to higher surface roughness [97], which may impair the gas barrier properties.

Organic acids (citric, lactic, malic, or tartaric) play a major role in increasing the thickness of a complex mixture containing nisin incorporated in soy protein [190]. Organic acids were introduced in different concentrations (0–2.5\%), and the thickness reaches a maximum within that interval. Without malic acid in the composition, the thickness is 25 \( \mu \)m, and with 1.6\% organic acid, the thickness reaches 35 \( \mu \)m.

4.2.3. Mechanical Properties

These properties are important, since they assure the physicochemical integrity of the protected food by film/coatings and are generally measured using a texture analyzer. Figure 14 shows three mechanical parameters that are taken into consideration for the characterization of films/coatings in terms of resistance at stress: tensile strength (\( TS \)), elongation at break (\( EAB \)), and modulus of elasticity (\( EM \)). Figure 14 represents a typical curve of these parameters’ evolution (the shape of the curve can change significantly with the quality or type of materials). The range of mechanical parameters out of those already mentioned are larger, including compression, puncture, tearing strength, burst, abrasion, etc., but these parameters are not the object of this paper.

![Mechanical parameters considered for the evaluation of edible films/coatings.](image-url)
Tensile strength can be calculated using the below equation, and the results are expressed in MPa.

\[ TS = \frac{F_{\text{max}}}{LW} \]  

(4)

where \( F_{\text{max}} \) is the maximum tensile force at rupture (N), \( L \) is the thickness of the film (mm), and \( W \) is the width of the film (mm). Elongation at break can be expressed as:

\[ E_{\text{AB}} = \frac{DL}{L} \times 100 \]  

(5)

where \( L \) is the initial length of the filmsample (mm), and \( \Delta L \) is the difference between the final and initial length of sample (mm).

The values of these mechanical indicators are categorized in quality ranges such as inferior, marginal, good, and superior. In this way, the users of films/coatings can objectively assess the compatibility of a coating with the food that needs to be protected. Table 2 presents some values obtained in different studies and with different materials that show the influence of certain ingredients on mechanical strength.

4.2.4. Barrier Parameters

Water vapor permeability (WVP) is a parameter that directly affects the freshness of the product embedded in films/coatings, since the water depletion significantly changes their organoleptic properties. The usual determination method for WVP is gravimetrically according to ASTM E96M-10 at 75% relative humidity (RH) (or other RH if required):

\[ WVP = \frac{w x}{A \Delta p} \]  

(6)

where \( x \) is the film thickness (m), \( A \) is the permeation area (m\(^2\)), \( \Delta p \) is the difference in vapor pressure across the film (Pa), and \( w \) is the weight of water gained by the film in the capsule per hour (g h\(^{-1}\)).

The water permeability has to be restricted in both directions, since the key of food preservation is to keep the food as it is. In general, the main ingredients of film coatings are proteins and polysaccharides with hydrophilic character, and in order to lower the WVP, some lipid products are used [151,162,191]. Other hydrophobic additives such as vitamin E decrease the WVP [192]. In addition, the introduction of chitosan biguanidine (Figure 15) into a ternary system composed of carboxymethyl cellulose/sodium alginate/chitosan biguanidine (CMC/A/CBg) in a progressive manner decreases the WVP from 330 to 178 (g/m\(^2\)/day) [193]. This occurs due to the higher reticulation and the cross-linking reactions among CMC, CBg, and alginate.

![Figure 15. Compounds used for edible films with lower water vapor permeability (WVP).](image-url)
The main barrier in water depletion is the hydrophobic substances, and the useful indicator for selecting them is presented in Figure 16.

![Figure 16. Substances used for obtaining lower values of WVP ranked in terms of hydrophobicity.](image)

Oxygen and CO$_2$ Permeability

Oxygen and CO$_2$ are important gases that affect the integrity of any food during storage. Usually, these parameters are measured using ASTM D 3985–02 (2002) or the new permeability meters provided by different producers. Edible films/coatings preserve the initial state of food by restricting the access of these gases. For example, vitamin C, which is an important dietary component of some foods, is depleted inside product if the coating is too permissive to O$_2$ or even CO$_2$ at higher concentrations. On the other hand, films with a high permeability of oxygen favor the production of ethylene and the fruits ripen quickly, while films with too low permeability allow fermentation [115]. In addition, it is important to acknowledge that water and oxygen permeability in most cases are inversely related; hence, edible films must be prepared in order to fulfill both features [128,194]. Low O$_2$ permeability allows preventing discoloration, which is an important organoleptic property for consumers. Some polymers used for edible films show lower O$_2$ permeability compared with classic polymers such as low density polyethylene (LDPE). Films based on additives such as chitosan, alginate, carrageenan, and pectin present good gas barrier properties. Some functional groups attached to biopolymer chains induce lower permeability for gases: –OH, –CN, –Cl, –F and –COOCH$_3$ (from higher to lower) [13,16,81,124,154]. Table 2 presents some of the permeation characteristics of the different formulations used to prepare edible films.
Table 2. Examples of formulations where mechanical and barrier parameters are changing as a function of composition.

| Formulation                                                                 | Composition  | Thickness (µm) | Tensile Strength (MPa) | EAB (%) | EM (MPa) | WVP $10^{10}$ (g m⁻² s⁻¹ Pa⁻¹) | CO₂ $10^{9}$ (cm³/m² s Pa) | O₂ 10¹⁰ (cm³/m² s Pa) | Moisture Content (%) | References |
|-----------------------------------------------------------------------------|--------------|----------------|------------------------|---------|----------|--------------------------------|---------------------------|------------------------|---------------------|------------|
| Fish skin gelatin (FSG), zinc oxide nanoparticle (ZnONP), ginger essential oils (GEO) different concentrations, Tween 20, glycerol | 0%           | 90.59          | 18.1344                | 78.2348 | -        | 6.42                           | -                         | -                      | -                   | [31]       |
|                                                                             | 10%          | 104.9          | 15.5879                | 101.364 | -        | 6.31                           | -                         | -                      | -                   |            |
|                                                                             | 20%          | 112.34         | 14.4574                | 111.028 | -        | 5.94                           | -                         | -                      | -                   |            |
|                                                                             | 40%          | 122.96         | 13.0296                | 120.421 | -        | 5.22                           | -                         | -                      | -                   |            |
|                                                                             | 80%          | 134.1          | 10.846                 | 132.463 | -        | 4.93                           | -                         | -                      | -                   |            |
| Fish gelatin, glycerol, water, extrusion and casting                        | Extrusion 20% (110 °C) | 450             | 2.41                   | 282.6   | 99       | 1.51                           | 21.4                      |                        |                     | [34]       |
|                                                                             | Extrusion 20% (120 °C) | 580             | 1.51                   | 256.3   | 118      | 1.97                           | -                         | -                      | -                   |             |
|                                                                             | Extrusion 25% (110 °C) | 410             | 1.92                   | 293.4   | 84       | 2.43                           | 16.1                      |                        | -                   |            |
|                                                                             | Extrusion 25% (110 °C) | 340             | 1.87                   | 237.2   | 132      | 2.92                           | 27.7                      |                        | -                   |            |
|                                                                             | Casting 20%   | 100            | 17.8                   | 27.4    | 482      | 1.91                           | 24.7                      |                        | -                   |            |
|                                                                             | Casting 25%   | 100            | 7.7                    | 49.4    | 259      | 2.5                            | 21.4                      |                        | -                   |            |
| Gelatin from sturgeon skin, glycerol, solution                              | Control      | 57.05          | 26.27                  | 53.83   | -        | 2.71                           | -                         |                        |                     | [37]       |
|                                                                             | 0.3% esculin  | 55.82          | 34.26                  | 52.37   | -        | 2.67                           | -                         | -                      | -                   |            |
|                                                                             | 0.6% esculin  | 57.25          | 35.42                  | 49.55   | 1.72     |                                |                           |                        |                     |            |
|                                                                             | 0.9% esculin  | 56.92          | 35.87                  | 42.86   | 1.32     |                                |                           |                        |                     |            |
| Microparticles containing sunflower oil, alginate, pectin coated in protein | FP + WPC 3.75 | 112.7          | 32.8                   | 9.4     | 6.7      |                                | 10.8                      |                        |                     | [66]       |
|                                                                             | FP + OVA 3.50 | 107.4          | 27.6                   | 13.2    | 8.9      |                                | 11.4                      |                        |                     |            |
|                                                                             | FP3.75       | 81.1           | 28.2                   | 5.9     | 11.5     |                                | 17.1                      |                        |                     |            |
|                                                                             | FP3.50       | 73.4           | 32.6                   | 3       | 11.8     |                                | 17                        |                        |                     |            |
| Starch (3%, 4% and 5%) from cassava, arrowroot, and canna edulis            | Cassava      | -              | 0.54                   | 127.16  | 0.172    |                                | 15.05                     |                        |                     |            |
|                                                                             | 3%           | 60             | 0.093                  | 107.47  | 0.214    |                                | 11.34                     |                        |                     |            |
|                                                                             | 4%           | 90             | 1.102                  | 130.91  | 0.184    |                                | 16.3                      |                        |                     |            |
|                                                                             | Arrowroot    | -              | 1.716                  | 67.85   | -        | -                               | 16.94                     |                        |                     | [67]       |
|                                                                             | 3%           | 700            | 1.633                  | 84.37   | 0.175    |                                | 11.4                      |                        |                     |            |
|                                                                             | 4%           | 85             | 1.827                  | 141.36  | 0.124    |                                | 15.2                      |                        |                     |            |
|                                                                             | Canna edulis | -              | 4.064                  | 61.19   | 0.167    |                                | 45.27                     |                        |                     |            |
|                                                                             | 3%           | 125            | 2.438                  | 52.23   | 0.225    |                                | 43.39                     |                        |                     |            |
|                                                                             | 4%           | 90             | 2.398                  | 42.94   | 0.176    |                                | 20.4                      |                        |                     |            |
| Thermoplastic starch (TPS) reinforced with hexametaphosphate (SHMP) and polyvinyl alcohol fiber (PVAF) | TPS          | 2.02           | 125.66                 | -       | -        | -                               | -                         | -                      | -                   | [77]       |
|                                                                             | 2% PVAF/TPS  | 2.62           | 83.618                 | -       | -        | -                               | -                         | -                      | -                   |            |
|                                                                             | SHMP/PVAF/TPS| 5.75           | 114.02                 | -       | -        | -                               | -                         | -                      | -                   |            |
| Formulation                          | Composition | Thickness (µm) | Tensile Strength (MPa) | EAB (%) | EM (MPa) | WVP 10^10 (g m⁻² s⁻¹ Pa⁻¹) | CO₂ 10¹⁰ (cm³/m s Pa) | O₂ 10¹⁰ (cm³/m s Pa) | Moisture Content (%) | References |
|-------------------------------------|-------------|----------------|------------------------|---------|----------|-----------------------------|-----------------------|---------------------|----------------------|------------|
| Vegetable residue (FVR) and potato skin (P) flours | Fvr/P = 10:0 | Average: 242 | 27 | 31.38 | 3 | 2.45 | - | - | - | [87] |
|                                      | Fvr/P = 8:0 |                | 28 | 30.51 | 3 | 2.48 | - | - | - |          |
|                                      | Fvr/P = 8:4 |                | 84 | 34.49 | 4 | 2.78 | - | - | - |          |
| Native starch, acetylated starch, glycerol (% in br/w) | (10:70:20) | 80.53 | 27.09 | 4.73 | - | 1.41 | 3.04 | 4.13 | 1.41 | [89] |
|                                      | (80:5:15)  | 75.97 | 16.25 | 2.59 | - | 0.88 | 2.66 | 2.08 | 0.88 |          |
|                                      | (15:70:15) | 122.93 | 10.31 | 20.14 | - | 1.31 | 4.13 | 3.31 | 1.31 |          |
| Corn starch, Uncaria gambir Roxb extract, glycerol | Extract concentration |                | | | | | | | |          |
|                                      | 20%         | 110 | 14.78 | - | - | 2.42 | - | - | - | [94] |
|                                      | 30%         | 119 | 15.11 | - | - | 2.46 | - | - | - |          |
|                                      | 40%         | 124 | 15.67 | - | - | 2.5 | - | - | - |          |
| Elephant foot yam starch, hydrocolloids xanthan (XG) and agar–agar (AA) | EFYS 0% | 163 | 15.81 | 23.96 | 54.08 | 1.91 | 1.036 | 23.66 |          |
|                                      | AAG 0.5%    | 186 | 17.3 | 19.75 | 60.81 | 1.2 | 4.49 | 25.3 |          |
|                                      | AAG 1%      | 194 | 17.64 | 15.36 | 63.43 | 1.29 | 7.36 | 25.38 |          |
|                                      | AAG 1.5%    | 197 | 17.85 | 13.62 | 65.08 | 1.16 | 5.39 | 25.62 |          |
|                                      | AAG 2%      | 199 | 20.14 | 13.34 | 58.03 | 1.1 | 1.06 | 26.42 |          |
|                                      | XG 0.5%     | 158 | 19.27 | 21.52 | 56.15 | 2.47 | 7.59 | 25.21 |          |
|                                      | XG 1%       | 173 | 19.1 | 17.4 | 58.28 | 9.09 | 1.09 | 22.83 |          |
|                                      | XG 1.5%     | 186 | 19.34 | 15.36 | 64.03 | 1.94 | 1.19 | 23.17 |          |
|                                      | XG 2%       | 187 | 19.48 | 14.69 | 69.77 | 9.34 | 5.48 | 24.36 |          |
| Cassava starch                       | CS          | 43 | - | - | - | 0.37 | - | - | 18.3 | [159] |
| Casava starch + clove essential oil  | CS + 0.8% EO W/V | 51 | - | - | - | 0.3 | - | - | 16.24 |          |
| Starch–glycerol whey protein (emulsifying) | Control | 3.17 | 48.91 | 1.24 | | | | | | [188] |
|                                      | WP 0.2%     | 2.65 | 35.49 | 0.86 | | | | | |          |
|                                      | WP 0.4%     | 2.02 | 34.66 | 0.7 | | | | | |          |
|                                      | WP 0.6%     | 1.5 | 32.02 | 0.6 | | | | | |          |
|                                      | WP 0.8%     | 1.81 | 31.74 | 0.64 | | | | | |          |
| Starch, sorbitol, mango peel, (NaOH solution up to 100%) | 5%/2%/0% | - | 10.17 | 2.75 | - | - | - | - | - | [195] |
|                                      | 5%/2%/2%    | 15.2 | 5.53 | - | - | - | - | - | - |          |
|                                      | 5%/2%/4%    | 13.45 | 6.69 | - | - | - | - | - | - |          |
| Agar and alginate, glycerol, Larrea nitida extract | Ag/Alg | 27.5 | 27.5 | 992 | 8.47 | 13.95 |          |
|                                      | Ag/Alg/Ln   | 19.5 | 14.5 | 970 | 6.1 | 7.47 |          |
|                                      | Alg         | 22.4 | 17.8 | 793 | 8.3 | 1.77 |          |
|                                      | Alg/Ln      | 10.3 | 10.6 | 784 | 6.04 | 4.16 |          |
|                                      | Alg/Alg/Ln  | 24 | 23.7 | 635 | 7.83 | 1.89 |          |
|                                      | Ag/Alg/Ln   | 12.9 | 21.4 | 477 | 6.06 | 3.76 |          |
| Formulation | Composition | Thickness (µm) | Tensile Strength (MPa) | EAB (%) | EM (MPa) | WVP $10^{10}$ (g m⁻² s⁻¹ Pa⁻¹) | CO₂ $10^{9}$ (cm³/m² s Pa) | O₂ 1010 (cm³/m² s Pa) | Moisture Content (%) | References |
|-------------|-------------|----------------|------------------------|---------|---------|-------------------------------|-----------------------------|------------------------|----------------------|------------|
| Zein protein, polyphenols, and/or essential oil | Zein | 115.4 | 10.73 | 3.69 | 648.28 | - | - | - | - | [197] |
| | Zein + Gallic acid | 115.66 | 8.59 | 3.52 | 428.5 | - | - | - | - | |
| | Zein + Vanillic acid | 124.98 | 6.99 | 2.75 | 445.49 | - | - | - | - | |
| | Zein + Carvacrol | 129.7 | 4.68 | 8.76 | 226.82 | - | - | - | - | |
| | Zein + Eugenol | 134.63 | 7.56 | 7.83 | 344.05 | - | - | - | - | |
| | Zein + Citral | 157.8 | 4.32 | 1.21 | 412.16 | - | - | - | - | |
| Sodium alginate/pullulan/capsaicin | SA/Pul/Cap–0% | 32 | 46.34 | 4.7 | 1.18 | - | - | - | 21.69 | |
| | SA/Pul/Cap–2% | 33 | 53 | 3.58 | 1.93 | - | - | - | 19.4 | |
| | SA/Pul/Cap–4% | 35 | 54.1 | 3.22 | - | 1.99 | - | - | 17.4 | [198] |
| | SA/Pul/Cap–6% | 36 | 54.41 | 3.16 | - | 2 | - | - | 16.72 | |
| | SA/Pul/Cap–8% | 38 | 55.25 | 3.08 | - | 3.06 | - | - | 15.2 | |
| Chitosan, starch, thyme extract | CHS | - | 9.5 | 98 | 17 | 9.6 | - | - | 6.6 | - | [199] |
| | CHS:TE | - | 8.2 | 47 | 51 | 8.8 | - | - | 4.3 | - | |
| Chitosan, lactoperoxidase with or without iodine | Chitosan | - | 388.73 | 20.39 | 7.89 | - | - | - | - | |
| | Chitosan/LPOS | - | 574.42 | 18.31 | 5.61 | - | - | - | - | |
| HDM, high methylester pectin; L232, polymer for industrial seed coating; Noil, nanocomposite of pectin and neem oil; Nwax, nanocomposite of pectin and carnauba wax. HDM pectin | HDM pectin | 76.67 | 28 | 1.08 | 1990.29 | - | - | - | - | [200] |
| | Nwax10 | 104.23 | 28.99 | 3.23 | 885.53 | - | - | - | - | |
| | Nwax20 | 112.7 | 29.02 | 3.17 | 721.86 | - | - | - | - | |
| | Nwax30 | 96.98 | 29.49 | 3.84 | 668.47 | - | - | - | - | |
| | Nwax40 | 112.14 | 29.86 | 3.38 | 525.99 | - | - | - | - | |
| | Nwax50 | 101.34 | 30.5 | 3.8 | 497.74 | - | - | - | - | |
| | Nwax60 | 123.28 | 30.28 | 4.28 | 453.19 | - | - | - | - | |
| Chia mucilage (CM) hydrocolloid glycerol (25%, 50%, and 75% w/w) | CM25 | - | 0.054 | 0.054 | 0.131 | - | - | - | 18.18 | [201] |
| | CM50 | - | 0.056 | 0.056 | 0.325 | - | - | - | 32 | |
| | CM75 | 0.06 | 0.06 | 0.442 | - | - | - | - | 41.88 | |
4.2.5. Other Characterization Methods of Edible Films/Coatings

Color

Color is an important organoleptic parameter, and at first glance, the success of coating it is represented by final product color. Color is measured usually with a Portable Colorimeter Reader. Evaluations are made on $L^*$ (luminosity), $a^*$ (+red, −green), and $b^*$ (+yellow, −blue), which represent the color parameters of the CIELab scale. Generally, color parameters were measured in five points on an edible film surface. Hue (qualitative attribute) and Chroma (quantitative attribute) were calculated following Equations (7) and (8), respectively [159]:

$$
Hue = \tan^{-1}\left(\frac{b^*}{a^*}\right) \quad (7)
$$

$$
Chroma = \sqrt{a^{*2} + b^{*2}} \quad (8)
$$

These values can be used to calculate the browning index [195], which is an efficient parameter for monitoring product freshness:

$$
BI = \frac{[100(X - 0.31)]}{0.172} \quad (9)
$$

$$
X = \frac{(a^* + 1.75L^*)}{(5.645L^* + a^* - 3.02b^*)} \quad (10)
$$

Increasing the plant extract in mixtures formed from starch and mango peel extract causes the color to turn yellow, as indicated by Hue and Chroma, and this is attributed to the higher concentrations of carotenoid and flavonoid compounds [195]. Alves et al., [159] found that little changes in color are observed by introducing essential oil in combination. Values of $a^*$ and $b^*$ are near zero, which gives the film a gray color.

Thermogravimetric Analysis

Similar to any polymers, biopolymers are sensitive to temperature; hence, thermostability represents an important parameter. In fact, thermogravimetric analysis stands as the thermal fingerprint of edible films/coatings. The thermogravimetric curves (TG—thermogravimetric analysis and DTG—differential thermal analysis) usually used in the film characterization of the films are recorded on a thermogravimetric analyzer under different atmospheres (more often nitrogen). The samples were heated from near ambient temperature to 500–800 °C at $x$ °C/min. A typical thermogram is presented in Figure 17. From these curves, the moment of water desorption, plasticizer volatilization, and at higher temperature, the polymer decomposition can be specified [202]. It is an important tool to assess the differences between formulations in a series or when the basal formulation is preserved and only some ingredients are added [96,124,200,203,204].

Film Morphology

The morphology of composite edible films is examined using a scanning electron microscope (SEM) under 500 magnifications or higher. It is a very useful technique for taking a closer look at the surface of edible barriers in order to observe any cracks, breaks, openings on the surfaces, or particular morphologies that can appear by mixing several ingredients [33,93,198,201,205–208]. The goal is to obtain a homogeneous surface without defects.
FTIR Spectroscopy

Fourier-transform infrared spectroscopy (FTIR) is a very useful method to record the infrared spectrum of prepared films. An FTIR spectrometer collects high-definition data in a large range of spectra, and the operating parameters are attenuated total reflection (ATR) mode, absorption or transmission mode, wavenumber ranging from 4000 to 400 cm\(^{-1}\) (room temperature, \(x = 110–120\) number of accumulated scans, resolution of 4 cm\(^{-1}\)). Fourier-transform infrared spectroscopy (FTIR) is used to identify the absorption bands that result from the vibrations of functional groups presented in macromolecules. Usually in the case of polysaccharides but also in the case of all compounds containing \(-\text{OH}\) groups, a strong band at approximately 3300 cm\(^{-1}\) is observed. If water is trapped in the film structure, a band can appear at 1750–1540 cm\(^{-1}\). Polysaccharides adsorb strongly in the 1200–900 cm\(^{-1}\) region due to C–O–C glycosidic linkage [91,177,209–211]. In chitosan and proteins, stronger bands appear at approximately 1651 cm\(^{-1}\), 1547 cm\(^{-1}\), and 1320 cm\(^{-1}\) corresponding to C=O stretching (amide I), N–H bending (amide II), and to C–N stretching (amide III) [174,177,212]. The positions of these bands are subject to shift due to new bond formation in the frame of complex mixtures used in edible films/coatings. This technique is very useful to assess the differences between formulations if some ingredients are changed, as observed by Zhang et al. [97].

X-ray Diffraction

X-ray diffraction is a characterization technique for different materials that permits the identification of crystal structures and interatomic distances. That is possible, since the X-ray wavelength is similar to the distances between atoms. The analysis can be applied using an X-ray diffractometer, which is usually provided with a tube, a copper anode, and a detector operating at 40–45 kV and 30–40 mA, and the range of \(\theta\) and speed are selected according to applications. Diffractograms of edible films (single or multi-component) reveal a combination between amorphous and crystalline areas (Figure 18) with high variations depending on some factors, including the biopolymer type and sources, plasticizers, film drying regime, moisture content, etc. Chitosan diffractograms present semi-crystalline features with two diffraction peaks (2\(\theta\) = 11° and 20°) [213]. In addition, a partially crystalline pattern was observed in a large number of finished films/coatings, which suggests that is the main feature of these materials [57,95,193,204,214]. Usually by mixing different ingredients, the diffraction bands existing in diffractograms sustain some attenuation, diminishing, scattering, and broadening, which are all clear indications of the crystallinity lowering.

Figure 17. Typical thermal analysis of edible films.
Antioxidant and Antimicrobial Activity

For the evaluation of antioxidant activity, some global parameters measured through colorimetric methods are used:

- Total phenolic contents (TPC) (Folin–Ciocalteu reagent) data are expressed in equivalents of mg of gallic acid for 100 g of film (mg GAE/100 g film) [78,118–120,155,156,215–228].
- Total flavonoids (aluminum chloride reagent) data are expressed in mg of quercetin equivalents for 100 g of film (mg QE/100 g film) [46,140,163,188,216,217,229–233].
- DPPH method (1,1-diphenyl-2-picrylhydrazyl radical reagent–DPPH) data are expressed in mg of ascorbic acid equivalents for 100 g of film (AAE/100 g of film) [30,32,37,47,72,160,162,163,169,199,233–239].
- ABTS (2,20-azino-bis (3-ethylbenzothiazoline-6-sulphonic acid reagent–ABTS) data are expressed in mg of ascorbic acid equivalents for 100 g of film (AAE/100 g of film) [32,47,166,174,211,235,239,240].

Antibacterial activity is performed by the assessment of the zone of inhibition assay on a solid medium placed in Petri dishes and containing specific media for each microorganism tested [74,125,129,141,171,176,179,187,213,238,241–259].

Strawberries, as one of the most highly consumed and also perishable fruits, require special attention in terms of edible coatings. Composite films obtained using chitosan–banana starch–glycerol–whey protein film formulation. Different amounts of nanoemulsion (0–0.8%) were introduced into the films to evaluate their antioxidant and antibacterial activity as well as mechanical properties, and it was found that 0.8% of nanoemulsion was the optimal value.

Egg white–sorbitol (3% v/w)–orange essential oil (2% v/v) films were prepared for the preservation of ‘kashar cheese’ for 30 days at 4 °C [167]. The orange essential oil composition consists mainly of limonene (84.2%), sabinen (3.2%), myrecene (1.2%), etc., which are known for their antimicrobial activity. Three cheese samples were artificially contaminated with dangerous bacteria E. coli, L. monocytogenes, and S. aureus. Two of them were covered with prepared films (with and without essential oil), while one remained as the control. It was found that in the essential oil-treated sample, the bacterial growth was inhibited rapidly (7 days for L. monocytogenes and S. aureus and 15 days for E. coli), while for the sample without essential oil, the growth decreased more slowly. As expected, the bacterial growth remained high in the unprotected sample.

For Turkish ‘lor cheese’ protection, Kavas et al. [261] used an edible film based on egg white proteins. The testing of the prepared coating solutions was performed by contaminating lor cheese with E. coli O157: H7 (ATCC 43895), L. monocytogenes (ATCC 19118), and S. aureus (ATCC 6538): ‘lor cheese’ (50 g), immersion into inoculum (15 min), first immersion into coating solution (90 s), hold in air (3 min),...
second immersion into coating solution (60 s), drying (10 °C/4–5 h), and storage at 4 °C for 30 days. The coating thickness was higher for samples with balm oil (2%), and therefore, the WVP was lower. The presence of bactericidal compounds in coatings stopped the development of the microorganisms from day one, and the optimal concentration of essential oil was 2%.

Garlic and oregano oils were used as adjuvants to “achira” starch (Canna indica L.) coatings for double cream cheese protection during storage at 5 °C for 42 days [262]. Comparing with control, the two types of films were suitable to preserve the product and eliminate the presence of pathogenic microorganisms. Oregano oil-based films convey lower hardness to cheese samples compared with garlic oil treatment and control.

Mango fruits were preserved from getting spoiled by using starch solutions (2%) from the pulp of banana, soursop, and stenospermocarpic mango (a fruit smaller than normal mango) [263]. The starch extends the shelf life of fruits up to 15 days with no signs of losing the organoleptic characteristics.

Millet starch-based edible films incorporating clove oil (60% eugenol as the main ingredient) in different amounts (1%, 2%, and 3%) have bactericidal activity toward different microorganisms: Syzygium aromaticum, Staphylococcus aureus, Escherichia coli, Pseudomonas aeruginosa, Enterobacter sp., and B. cereus [149]. A Box–Behnken experimental design (BBD) was used to optimize the preparation process of edible coating for protecting ‘Cripps pink’ apples [224]. The independent variables were percent of starch, carrageenan, sucrose fatty acid ester, and glycerol, while the dependent variables were thickness, WVP, solubility, TS, and EAB. Response surface methodology gave the optimum solution for preserving apple qualities: starch 2.5% carrageenan 1.5%, sucrose fatty acid ester 2%, and glycerol 1.5%.

The antioxidant activity of starch films was also improved using microencapsulated ascorbic acid [264], embedding carvacrol and citral [184], the addition of nystose from Bacillus subtilis natto [265], Aloe vera gel [161], turmeric [36], thyme extracts [199], natural plant extracts (epigallocatechin gallate, blueberry ash, fruit extract, macadamia peel extract, and banana peel extract [163, 266].

Gelatin films containing chitosan nanoparticles enriched with tea polyphenols were proven to be suitable for food protection, especially in the context of an innovative electrospray preparation method [23]. This technique is suitable for the better incorporation of polyphenols in chitosan, which is known as a good carrier for different compounds. The preparation of chitosan microcapsules was followed by incorporation in gelatin and film preparation. The gelatin used was provided by grass carp skin, which is one of the main by-products of fish processing in China. Ferulago angulata essential oil (1-3%) and chitosan (approximately 450 kDa) were used as coatings for preserving trout during 16 days storage at 4 °C [139]. The formulation with 3% essential oil presented superior properties.

Aloe vera gel [142, 161, 260, 267, 268] represents an important ingredient in chitosan-based formulations, since this extract has multiple benefits: anti-inflammatory, detoxifying, lowering cholesterol, improving metabolism, anti-ulceration, improving oral health, fighting cold and cough, etc. Therefore, besides edible coatings, aloe vera gel is both food and medicine, which is probably due to the content of glycosides aloin A and B (15-40%) [269].

Other adjuvants for improving the properties of chitosan edible coatings were also used. Pickering emulsions formed by chitosan–cellulose nanocrystals–oleic acid (10 or 20 g oleic acid/kg emulsion) were used for ‘Bartlett’ pears ripening prevention [270]. Chitosan–lemon verbena extract/essential oil for trout (Oncorhynchus mykiss) preservation [142].

Mango fruit was protected against fungi (Phomopsis sp. RP257 and Pestalotiopsis sp.) using a system composed of chitosan and lactoperoxidase [242].

Strawberries were protected by a mixture of chitosan–chitosan nanoparticles–propolis extract (10–30%) [217]. Chitosan nanoeumulsions prepared by high-energy or low-energy methods represent an advanced edible coating for food [129]. In addition, it was found that chitosan–beeswax–carboxymethylcellulose are very efficient coatings for Kinnow mandarin fruits [68]. An interesting approach [27] was to produce coatings from mixtures of gelatin–furcellaran–pu-erh
tea extract–green tea extract (GTE). Furcellaran is a naturally occurring polysaccharide that can be extracted from red seaweeds (Rhodophyceae). Jamróz et al. evaluated the coating properties: physical (color, swelling, thickness, water content), antioxidant (TPC, DPPH, ABTS), antimicrobial (Staphylococcus aureus, Escherichia coli, Henseniaspora uvarum and Candida albicans). Tensile strength increased from 9.62 to 24.14 when GTE was introduced, and a significant inhibition zone of 25 mm appeared against S. aureus. Moreover, it can be considered an intelligent film due to the capability of color changing at different pH values (no color at pH = 3 and orange at pH = 12). This feature is important for fish preservation tests.

5. Statistical Analysis. Design of Experiments (DOE)

In order to manage complex mixtures and processes for obtaining edible films/coatings, it is necessary to optimize the process, and this task require several steps:

Screening–where the factors that influence significantly the preparation are identified (modality of completion: fractional factorial [107])

Improvement–a process through which the near optimum parameters are founded by changing the factor settings (modality of completion: Box/simplex or steepest ascent approach)

Optimization–where optimal settings are founded (modality of completion: response surface designs such as CCD or Box–Behnken [126]).

Response surface design stands for an advanced design of experiments (DOE). Several types of software are used for fulfilling these tasks: JMP, Minitab, Design Expert, etc. These programs have implemented the methodology to perform the laborious statistical analysis required. Basically, these procedures offer information of the effect of input variables/factors (temperature, pH, composition etc.) to output variables/response (thickness, TS, WVP, EM, EAB, etc.) and also the reciprocal effects of independent variables (taken two at a time) on the final product quality (Figure 19). Factors can take only a few potential values called factor levels. Response surface methodologies are divided in two categories:

- Central composite designs;
- Box–Behnken designs.

![Response surface plots](image_url)

**Figure 19.** Example of response surface plots emphasizing the influence of independent variables (factors) interaction on the output variables (response).
Central composite designs are used because they provide a uniform precision of estimates effect, unlike Box–Behnken designs, which are less precise but need fewer design points and consequently are less expensive to run using an equal number of factors such as CCD.

Box–Behnken designs can use up to 3 levels per factor, while CCD can use up to 5.

For predicting the experimental output, a second-order polynomial model is usually used.

\[
\hat{Y} = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_{i<j}^{k} \beta_{ij} X_i X_j
\]

(11)

where \(Y\) is the predicted value of the response; \(\beta_0, \beta_i, \beta_{ii}, \text{and} \beta_{ij}\) are the regression coefficients; \(k\) is the number of independent parameters; and \(X_i\) and \(X_j\) are the coded levels of the experimental conditions.

6. Conclusions

Edible films/coatings are efficient ways to prevent the spoilage of different foods. Important achievements in this industry are exponentially increased by the number of ingredients and their combinations, which can be used and tailored to adapt to any kind of edible protection for fast/slow perishable food. The investigations pursued by different research teams aim to optimize the formulation compositions. Particular attention is attributed to mechanical properties, which require a delicate harmonization between resistance and elasticity. In addition, antioxidant and antimicrobial features are mandatory for these ecological envelopments; hence, numerous studies report the utilization of diverse types of essential oils and plant extracts. The possibility of obtaining an edible layer from other ready-to-use waste (shrimp shell, fruits peels, pomace, etc.) significantly increases the practice of food protection in this manner.

Experimental designs are extensively used to obtain the best result from complex edible coatings and to carefully assess the beneficial or detrimental role of each component. Along with matrix characteristics, application methods represent an important challenge, especially when the transition from lab scale to industrial scale is required.

Another challenge in achieving effective edible layers on food is the increasingly frequent introduction of different types of nanoparticles (silver, ZnO, silica, etc.), which, besides beneficial effects, can bring new issues regarding consumer protections. Regulatory requirements for the presence of nanoparticles in this field may become a difficult task for researchers in the future.

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