

Review

# State of the Art of Antimicrobial Edible Coatings for Food Packaging Applications

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**Abstract:** The interest for the development of new active packaging materials has rapidly increased in the last few years. Antimicrobial active packaging is a potential alternative to protect perishable products during their preparation, storage and distribution to increase their shelf-life by reducing bacterial and fungal growth. This review underlines the most recent trends in the use of new edible coatings enriched with antimicrobial agents to reduce the growth of different microorganisms, such as Gram-negative and Gram-positive bacteria, molds and yeasts. The application of edible biopolymers directly extracted from biomass (proteins, lipids and polysaccharides) or their combinations, by themselves or enriched with natural extracts, essential oils, bacteriocins, metals or enzyme systems, such as lactoperoxidase, have shown interesting properties to reduce the contamination and decomposition of perishable food products, mainly fish, meat, fruits and vegetables. These formulations can be also applied to food products to control gas exchange, moisture permeation and oxidation processes.

**Keywords:** antimicrobial; edible coatings; food packaging

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## 1. Introduction

The search for more natural and healthy food, based on minimally-processed, easily-prepared and ready-to-eat fresh products, has resulted in an increase in consumer requirements for safe and high-quality food [1]. These new social trends joined with changes in the usual procedures to make food processing faster and more efficient have caused a rising interest to obtain food products with a longer shelf-life. These properties are directly related to the development of new improved packaging materials, including active, intelligent and edible systems [2,3].

Food products are perishable by nature, as they can be subjected to degradation by many different environmental effects, including contamination by bacteria and fungi. Therefore, processed food requires protection from spoilage during preparation, storage and distribution to improve shelf-life and quality. Many of these microorganisms, in particular pathogens, could cause severe health problems to consumers, especially if food is handled and distributed under inappropriate conditions. In addition, undesirable reactions can occur to modify odor, flavor, color and textural properties in fresh food [4]. Traditional preservation techniques, such as heat treatment, salting or acidification, have been applied for a long time by the food industry to prevent the growth of spoilage and pathogenic microorganisms in food, but they often result in unacceptable losses in their nutritional value [1]. However, new strategies related to preservation techniques have been demonstrated to be more effective in protecting food without hampering organoleptic and nutritional properties. This is particularly the case for edible antimicrobial films and coatings that have attracted the interest of researchers and the food industry in

the last decade, since they can improve the safety, quality and functionality of food products while inhibiting the growth of undesirable microorganisms during storage, transportation and handling [2].

Different biopolymers, such as proteins, lipids, polysaccharides or their combinations, have been used as carriers to produce edible coatings with antimicrobial properties (Figure 1) [5–8]. They can be directly extracted from biomass and easily processed to get films to be used as coatings to control gas exchange, moisture permeation or oxidation processes in food while also reducing the microorganism's growth. These biopolymer films can be also used to host additives and nutrients to be released at a controlled rate to food, forming the basis of active packaging systems [9]. Other biopolymers, such as chitosan, pectin and pullulan, have been used with sodium benzoate and potassium sorbate to obtain edible coatings to improve the quality and shelf-life of fresh food [10]. The functionality of edible coatings can be expanded by incorporating antimicrobial additives to protect food products from microbial spoilage, extending their shelf-life and safety. Some reports have been recently published with the aim to evaluate and demonstrate the effectiveness against several pathogens of antimicrobial substances incorporated as active additives into edible coatings [5,11–13]. The incorporation of antimicrobial agents into edible coatings and their effectiveness could be improved by using nano-emulsification, which would also reduce essential oils losses by volatilization [14].

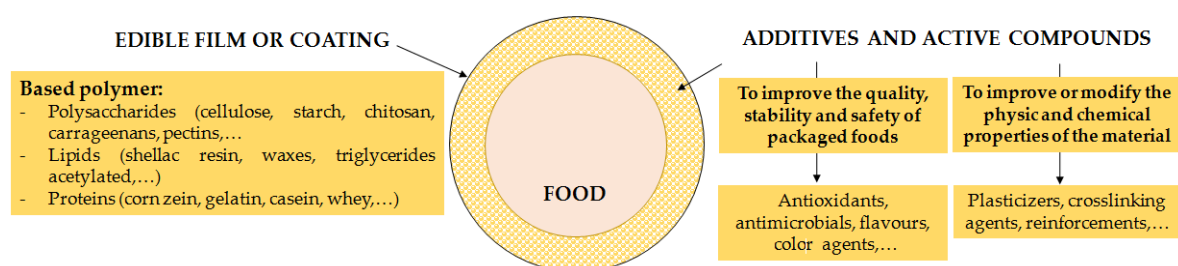


Figure 1. Edible films' and coatings' compositions.

Essential oils derived from plants [14,15], organic acids [16], nisin or natamycin from microbial sources [17], enzymes obtained from animal sources [18], like lysozyme and lactoferrin, and polymers, such as chitosan [19], have been also proposed as active agents against microorganisms in food.

Bioactive edible coatings can be applied to food surfaces by dipping or spraying, and they have been tested in meat, fish, dairy products or minimally-processed fruit and vegetables. Kraśniewska et al. reported that pullulan with 1.0% of oregano essential oil was an effective active material to maintain the safety and quality of fresh Brussels sprouts stored at 16 °C for 14 days [20]. Eggs coated with chitosan and lysozyme-chitosan combinations maintained their internal quality for a long-term storage since the albumen properties (including pH, dry matter, viscosity and relative whipping capacity) were kept as in their original state [21]. Shakila et al. evaluated the antimicrobial activity against *Staphylococcus aureus* (*S. Aureus*), *Aeromonas hydrophila* and *Listeria monocytogenes* (*L. monocytogenes*) of gelatin coatings with different additives (chitosan, clove and pepper) in vacuum-packed fish steaks. Satisfactory results were obtained for coatings with chitosan and clove, which extended shelf-life from four to eight days at 4 °C [6]. Other applications of these antimicrobial coatings in meat products can be also found in the recent literature. For example, Matiacevich et al. studied the effect of alginate-based edible coatings with propionic acid and thyme essential oil on the microbiological growth in chicken breast fillets stored under refrigerated conditions, demonstrating the inhibition of *Salmonella* after seven days [22].

This review focuses on the use of antimicrobial agents in edible coatings, including incorporation methods and the main characteristics of these innovative combinations. An overview of the state of the art of antimicrobial coatings and their use in different food products is presented in the following sections.

## 2. Antimicrobial Agents

A wide variety of antimicrobial agents has been described for their use in coating formulations for packaged fresh food [9]. Some of them are derived from natural sources, and they have been traditionally used as food additives and been awarded with the Generally Recognized as Safe (GRAS) label. Organic acids, enzymes, bacteriocins and plant-derived compounds and by-products, including essential oils, such as thymol or carvacrol, have been proposed for their antimicrobial performance [4]. The selection of the most appropriate antimicrobial agent is an important issue, and some relevant aspects should be considered, particularly the good interaction between the polymer matrix and the active agent, the presence of other functional additives in the formulation, the type and properties of the packaged food and their effectiveness against the target microorganisms. These aspects can result in major modifications of the coating final properties and the antimicrobial activity against the target microorganisms.

Organic acids and their salts are those compounds most commonly used as antimicrobial agents due to their well-known effectiveness and low cost. In addition, many of them are also labelled with the GRAS status, and they are accepted as food preservatives in the current European legislation [16]. Some of them have been traditionally incorporated into coatings, such as lauric, acetic, sorbic, citric, benzoic or propionic acids. Their antimicrobial effect has been related to the increase in the proton concentration thereby decreasing the external pH. In this way, the integrity and permeability of the microbial cell membranes can be altered, as well as some disturbance in the nutrients transport can be observed, causing cell inactivation and death [1]. Jin et al. [23] reported the effect of chitosan and the combination of three organic acids (lactic, levulinic and acetic) on fresh American ginseng roots. The antimicrobial activity of this coating was evaluated in terms of microbial stability by using total aerobic bacteria, yeasts and molds, showing that the microbial loads on coated samples were relatively stable during the 38-week storage period, with populations ranging between 2.2 and 2.9 log CFU·g<sup>-1</sup>. They also concluded that the use of a mixture of these antimicrobials can be more effective than the separate addition of the individual compounds due to synergistic or additive effects, permitting the inhibition or even killing certain microorganisms [24].

The antimicrobial activity of different spices and herbs has been known from ancient times, and they have been traditionally added to food as seasoning additives due to their aromatic properties [25]. Several studies have reported the preservation abilities of plant-derived compounds in food applications, as well as factors influencing their effectiveness. Among them, essential oils and their main components, such as thymol, carvacrol, *p*-cymene and  $\gamma$ -terpinene in *Thymus* [26], are gaining interest due to the presence of phenolic compounds or other hydrophobic components [27]. These phenolic groups are responsible for damage to the cell wall, interaction with and disruption of the cytoplasmic membrane, damage of membrane proteins, leakage of cellular components, coagulation of cytoplasm and depletion of the proton motive force [28]. All of these effects produce death to microorganisms by modification of the structure and composition of the bacteria cell walls [27]. However, their structural diversity and variations in their chemical composition can produce significant differences in their effectiveness against pathogens. For example, the antimicrobial effect of *Mosla chinensis* methanolic extract was evaluated against eight bacterial and nine fungal strains [29]. Results showed that this essential oil, whose main components are carvacrol (57%), *p*-cymene (14%), thymol acetate (13%), thymol (7%) and *c*-terpinene (2%), showed great potential against two Gram-positive bacteria commonly found in many food products, *S. aureus* and *L. monocytogenes*.

In general terms, essential oils are rich in monoterpenes, sesquiterpenes, esters, aldehydes, ketones, acids, flavonoids and polyphenols, which are well known for their antimicrobial performance [30]. Their mechanism of action against bacteria is not clear, since each compound in each essential oil exhibits a unique antimicrobial mechanism, which is specific to a particular range of food and microorganisms [31]. Table 1 shows some antimicrobial agents with antimicrobial activity on edible coatings.

**Table 1.** Antimicrobial activity of some edible films.

Antimicrobial Agent	Matrix	Microorganisms Tested	Reference
Citral and eugenol	Alginate and pectin	Aerobic mesophilic microorganisms, yeast and molds	[32]
Oregano essential oil	Basil seed gum	Aerobic mesophilic microorganisms, yeast and molds	[33]
Oregano essential oil	Mucilage	<i>L. monocytogenes</i> , <i>Salmonella typhimurium</i> , <i>Bacillus cereus</i> , <i>Yersinia enterocolitica</i> , <i>P. aeruginosa</i> , <i>S. aureus</i> , <i>E. coli</i> , <i>E. coli</i> O157:H7	[34]
Lemongrass	Alginate	<i>E.coli</i> , psychrophilic bacteria, molds and yeast	[35]
Clove	Gelatin	Total bacterial counts, <i>pseudomonas</i> , <i>Enterobacteriaceae</i> , lactic acid bacteria	[36]
Orange essential oil	Gelatin	Total viable counts, psychrotrophic bacteria and <i>Enterobacteriaceae</i>	[37]
Oregano, thyme essential oils	Soy protein	<i>E. coli</i> O157:H7, <i>S. aureus</i> , <i>P. aeruginosa</i> and <i>Lactobacillus plantarum</i>	[38]
Carvacrol and cinnamaldehyde	High methoxyl pectin and apple, carrot or hibiscus puree films	<i>L. monocytogenes</i>	[39]

Alparslan et al. added different amounts of essential oils obtained from orange (*Citrus sinensis* (L.) Osbeck) leaves into an edible gelatin coating solution with noticeable effects on the quality and shelf-life of shrimp covered by this edible film coating. The total viable counts, psychrotrophic bacteria counts and *Enterobacteriaceae* were determined [37]. Zhang et al. [40] used cinnamon bark and soybean oils in antimicrobial sodium alginate coatings, showing complete inhibition of *L. monocytogenes*, *Salmonella enterica* (*S. enterica*) and *Escherichia coli* O157:H7 (*E. coli* O157:H7) inoculated on cantaloupes after 15 days of storage. Other essential oils coming from different plants have been also used in edible coatings to improve the shelf-life of food products: garlic [41], basil [42], clove and pepper [6], oregano [20], thyme [22,43] and mandarin [15], among others.

Among natural antimicrobials from animal origin, chitosan has captured the attention of researchers and the food industry for commercial applications. This material offers the possibility of obtaining coatings to cover fresh or processed foods to extend their shelf-life, being an excellent film-forming material showing antifungal and antimicrobial activity due to its polycationic nature [44]. Regarding the mechanism of the antimicrobial activity, different ideas have been reported, although the precise mechanism has not been yet determined. Some authors have drawn theories based on: (i) interactions by electrostatic forces between chitosan amine groups and microbial cell membranes; (ii) the action of chitosan as a chelating agent; (iii) the penetration of low molecular weight chitosan molecules through the cell membrane; or (iv) modifications on cell surfaces that may affect the integrity of the microbial cell membrane interfering with energy metabolism and nutrient transport in bacteria cells [19,45]. Carrión-Granda et al. reported the combined effect of chitosan coatings with oregano and thyme essential oils (0.5%) in modified atmosphere packaging conditions onto peeled shrimp tails. The antimicrobial effect of this coating agent was evaluated for 12 days under chilling, and they further determined the total viable counts, lactic acid bacteria, *Enterobacteriaceae* and total psychrotrophic bacteria. Results showed that antimicrobial activity of the combination of chitosan with thyme was significantly increased in relation to samples treated just with chitosan, since the reduction was about 1 log CFU·g<sup>-1</sup> at the end of the storage period [46].

Other antimicrobials obtained from bacteria, such as nisin, pediocin, natamycin or reuterin, have been also used against target microorganisms. Particularly, bacteriocins are proteinaceous compounds with antimicrobial activity produced by lactic acid bacteria [18]. Ribosomally-synthesized

peptides with antimicrobial activity, such as nisin, have shown their ability to inhibit the growth of Gram-positive and spore-forming bacteria associated with food [1]. Nisin is able to penetrate the cytoplasmic membrane of bacteria causing the leakage of cytoplasmic contents and dissipation of the membrane potential [17]. Nisin also inhibits Gram-negative bacteria when chelating agents, such as EDTA or lysozyme, are present [9]. Nisin has been tested as an antimicrobial additive in coated surfaces in different products, such as dairy foods [47], fruits and vegetables [48,49], meats and fishes [50–53].

### 3. Application of Antimicrobial Edible Coatings

An edible coating material can be defined as a thin layer of the selected formulation, which is applied directly over food in liquid form by using different techniques, such as immersion, spraying, etc. [54]. In addition, an edible film is defined as a packaging material, which is a thin layer placed on or between food components, used as wraps or separation layers [55]. The edibility of films and coatings is only achieved when all components including biopolymers, plasticizers and other additives are food-grade ingredients, while all of the involved processes and equipment should be also acceptable for food processing [56]. The main coating techniques used in these materials are described below.

#### 3.1. Spraying

The interest in the industrial application of sprays in packaging is greatly increasing not only due to the potential cost reduction that the spraying technique may imply, but also by the high quality of the final product. This could be achieved when compared to those obtained by using conventional techniques [57], which mainly involve high temperatures, leading to important losses in volatile antimicrobial agents. The spraying technique offers uniform coating, thickness control and the possibility of multilayer applications [58,59]. Spraying systems do not contaminate the coating solution, allow temperature control and can facilitate automation of continuous production. In this sense, temperature directly affects the permanence of the antimicrobial agents due to their high volatility. In addition, the thickness control is very important to establish the amount of antimicrobial agent necessary to be released [60].

On the other hand, sprayed coatings combine hydrophobic and hydrophilic substances. Indeed, the spray can generate a coating with two solutions, by applying the emulsion solution directly, which is formed before atomization, or by forming a bilayer after two spray pulverizations. The application of a bilayer has the disadvantage of requiring four steps (two spray applications and two drying processes) [61].

By using this technique low viscosity coating solutions can be easily sprayed at high pressures [62]. The drop-size distribution of a sprayed coating solution can be up to 20  $\mu\text{m}$ , whereas electrospraying can produce uniform particles lower than 100 nm from polymer and biopolymer solutions. Furthermore, the formation of polymer coatings by spraying can be also affected by other factors, such as the drying time and temperature [63].

A spray is a collection of moving droplets as the result of atomization processes to break up bulk liquids into droplets [64], including the following considerations:

- Increase in the liquid surface area, which is an important issue in processes where rapid vaporization is required. In fact, in antimicrobial applications, it is important to obtain homogeneous coatings where the additive is available to release quickly to the surrounding environment.
- The formation of an even surface, since the droplets dispersion generates coatings with homogeneous spatial patterns and controlled thicknesses. This is essential to evaluate the kinetics release of the antimicrobial additive.



- Cost reduction, since spraying techniques are usually fast and efficient processes in terms of solvent and material consumption.

Different spraying techniques have been proposed [65]:

- Air-spray atomization: In this case, the fluid emerging from a nozzle at low speed is surrounded by a high-speed stream of compressed air (up to 8 bar). The friction between the liquid and air molecules accelerates and disrupts the fluid stream and causes atomization.
- Pressure (airless) atomization: High pressures (34–340 bar) force the fluid through a small nozzle (spray tip) to emerge as a sheet. The friction between the fluid and the air molecules disrupts the stream, breaking it initially into fragments and ultimately into droplets. The fast-moving, high-pressure liquid stream provides energy enough to overcome the fluid's viscosity and surface tension by forming small droplets.
- Air-assisted airless atomization: This technique combines the features of air spraying and airless techniques. It is based on the principle of the airless atomization with the addition of a concentrated airflow to obtain droplets in a more controlled way.

A variety of factors can affect the droplet size and the liquid stream [66], corresponding to the fluid (surface tension, viscosity, density and temperature) and technique (spray tip size and shape, fluid and air pressure). Heat-sensitive antimicrobial agents are volatile compounds, and they should be preferably incorporated onto food matrices by non-heating methods, such as spray coating. In this sense, Peretto et al. [67] used spraying as an innovative and efficient technique for the application of an edible alginate coating enriched with carvacrol and methyl cinnamate (natural antimicrobials) onto fresh strawberries demonstrating superior performance on firmness, color retention and weight loss reduction in comparison with uncoated samples. Other examples of sprayed-antimicrobial edible coating films include those based on a tapioca starch with green tea extracts that have reduced the growth of aerobic microorganisms and yeasts when applied to fruit-based salads, romaine hearts and pork slices [68].

### 3.2. Dipping

The dipping method has been used to form coatings onto fruits, vegetables and meat, among other food products [69]. Properties such as density, viscosity and surface tension of the coating solution are important to determine the film thickness [5], since dipping techniques are able to form thick coating layers [62]. In this method, a membranous film is formed over the product surface by directly dipping the product into the aqueous coating formulation and further air-drying. This process may be separated into three stages [70]:

- Immersion and dwelling: The substrate is immersed into the precursor solution at a constant speed followed by dwelling to ensure that interaction of the substrate with the coating solution is enough for complete wetting.
- Deposition: A thin layer of the precursor solution is formed on the food surface by deposition. The liquid excess drains from the surface and is removed.
- Evaporation: The solvent excess evaporates from the fluid, forming the thin film.

It is important to highlight that, when using this method, the coating solution must be diluted, and significant residual coating material is produced. The optimal amounts of coating solution cannot be easily controlled by dipping, and a further processing step to dry off surplus solution is needed, requiring extra time and hindering industrial applications of this technique [57].

Different examples of the use of the dipping technique for edible coating processing have been reported. A significant shelf-life extension for oysters was achieved by dipping them into a sodium acetate (10 g/L), solution resulting in a coating with sodium alginate (40 g/L) and further use of modified atmosphere packaging (MAP) conditions (0:75 O<sub>2</sub>:CO<sub>2</sub>) [71]. In addition, other authors

evaluated the dipping method by coating papaya fruit with k-carrageenan [72] and carrots with sodium alginate [73] as packaging strategies to extend the shelf-life of fresh foods.

### 3.3. Spreading

This method, also known as brushing, consists of the controlled spreading of a suspension onto the material surface to be further dried. This method is considered a valid alternative for the preparation of films with dimensions larger than those prepared by casting procedures. The thickness of the coating suspension is controlled by a blade attached to the lower part of the spreading device, and the film drying is held on the support itself, by circulation of hot air. This method can be applied to the production of polysaccharides and protein-based films [74]. Two parameters can be used to characterize the spreading of liquid droplets: the wetting degree and the spreading rate [75]. In this sense, contact angle measurements are commonly used to evaluate the degree of spreading/wettability of a surface by a particular liquid. Spreading is affected by several factors, such as the substrate properties (surface roughness and geometry), system conditions, such as temperature and relative humidity, and liquid properties (viscosity, surface tension and density) [76]. Viscosity was found to have a major effect as it defines the resistance of a liquid to spreading on solid surfaces. Therefore, spreading of highly viscous liquids is more difficult than liquids with low viscosity [77].

As an example of antimicrobial spread surfaces, polyethylene (PE) was coated with chitosan, where the polymer surface was previously corona-treated to enhance the chitosan adhesion. The antimicrobial activity against Gram-positive (*L. monocytogenes*) or Gram-negative (*E. coli*, *Salmonella*) bacteria was proven in uncoated and chitosan-coated PE films [78]. Active packaging films containing partially purified antibacterial peptide solutions (ppABP) produced by *Bacillus licheniformis* Me1 were developed by using low density polyethylene (LDPE) and cellulose films by spread coating, showing a remarkable antibacterial activity. The release study of ppABP from the coated films showed that ppABP was released from LDPE films as soon as they are in contact with water, while a more gradual release of the coated ppABP was observed in the case of cellulose films [79].

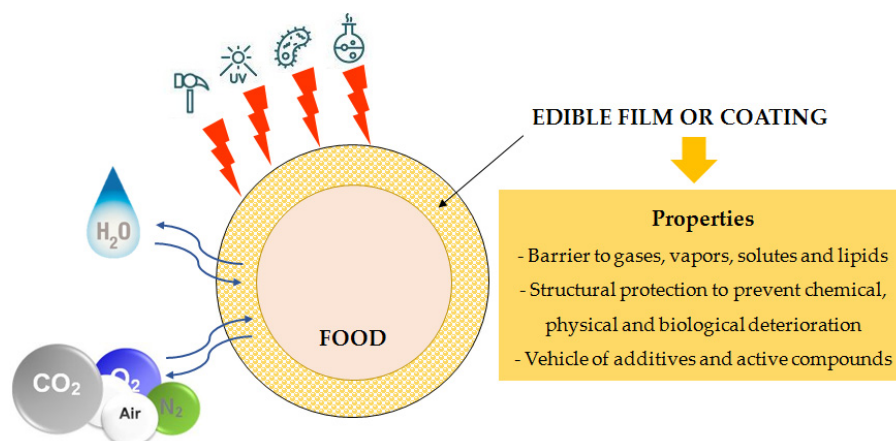
In summary, although coating application methods are very diverse, their selection depends on the desired product, coating thickness, solution rheology and the drying technique.

## 4. Properties of Antimicrobial Coatings for Food Packaging Applications

Antimicrobial edible films and coatings have demonstrated their ability to protect foodstuff against spoilage and to decrease the risk of pathogens growth by controlling the diffusion and gradual release of embedded antimicrobial agents onto the food surface [80]. The selection of the most adequate antimicrobial agents is based on the consideration of their effectiveness against the target microorganisms, as well as their possible interactions with the film-forming polymers and the packaged food. These interactions can modify the antimicrobial activity and their own film characteristics, constituting key factors for the development of antimicrobial films and coatings [9]. The most common functions of edible films and coatings in food packaging are described in Figure 2.

Different physical tests to determine the mechanical and barrier properties of edible coatings and films have been reported. Quasi-static tension or puncture tests are applied to edible films to determine mechanical parameters, such as the elastic modulus, tensile strength and strain at break [81]. Water vapor permeability (WVP) of these films is determined in accordance with the ASTM E-96 static method. The resistance of films to water is critical for the potential application of these films since much food is stored in aqueous solutions. Sometimes, high solubility in water is desired, particularly when the film or coating will be consumed simultaneously with food. On the other hand, as a consequence of the generally poor barrier to water vapor and low mechanical strength of biopolymers, some edible films and coatings have still limited applications in food packaging [44]. In this sense, the use of polysaccharides is limited by their water solubility and high WVP. Blending with different biopolymers or addition of hydrophobic materials such as oils or waxes can be useful to overcome this shortcoming [82]. Chemical modification of the biopolymer structure, in particular by

crosslinking [83], has been also proposed for such purposes [84,85]. Gutierrez et al. used this method to modify starch by cross-linking with sodium trimetaphosphate [86]. Edible films showed hydrophilic characteristics and some increase in the degradation temperature. In addition, the films developed in their studies were actually edible and, consequently, considered safe by the U.S. Food and Drug administration (FDA). Schmid et al. carried out swelling studies to demonstrate the improvement of the structural stability of whey protein isolate-based coatings. Results demonstrated that the denaturation degree had a significant influence on the cross-linking density, consequently a direct proportion of the number of disulfide bonds in the WPI-based network [87].



**Figure 2.** Main functions of edible films and coatings in food packaging applications.

Physico-chemical properties (thickness, WVP, puncture strength, tensile strength and elongation at break) of chitosan films enriched with oregano essential oil were evaluated [88]. The results of these tests showed that this combination resulted in increased thickness, higher elasticity, reduced puncture and tensile strength and lower WVP compared to pure chitosan films and chitosan films with surfactant.

Antimicrobial properties of edible coatings can be evaluated by using different types of microorganisms and different testing methods, including the film disk agar diffusion assay, the enumeration by plate count of microbial population and the film surface inoculation test [9]. These methods have been applied for the *in vitro* evaluation of the antimicrobial films performance. However, when coatings are applied onto food, their effectiveness is evaluated through the enumeration of indigenous or inoculated microbial population during food storage. For example, bologna-ham slices covered with chitosan films with 1% oregano essential oil under storage for five days at 10 °C absorbed 60 ppm of the essential oil and were effective against *L. monocytogenes* and *E. coli* O157:H7. The application of pure chitosan films reduced the pathogen counts on meat products from 1–3 logs, and chitosan films enriched with 1 and 2% oregano essential oil were sufficient for a four log reduction of *L. monocytogenes* and *E. coli* O157:H7.

## 5. Food Packaging Applications

This section is focused on the main studies reported in the scientific literature of antimicrobial coatings for food packaging purposes. In general terms, due to their chemical characteristics and their high degradation rates, antimicrobial coatings have been widely studied for fish and meat products, while some studies have been also reported with fruits and vegetables. Table 2 summarizes recent studies reported in the literature regarding the applicability of new antimicrobial coatings for food products.



**Table 2.** Antimicrobial coatings developed for fish and meat products.

Food	Product	Matrix	Antimicrobial Agent	Ref	
Fish products	Sliced fresh <i>Channa argus</i>	Chitosan and polyethyleneimine	Thyme essential oil	[89]	
	Fish sausages	Chitosan and warm-water fish gelatin	Shrimp concentrate from <i>Litopenaeus vannamei</i> cooking juice	[90]	
	Trout fillets	Chitosan	Lactoperoxidase enzyme	[91]	
		Whey protein	Lactoperoxidase enzyme	[92]	
	Fresh Indian salmon	Gelatin from waste of <i>Nemipterus japonicus</i>	Garlic ( <i>Allium sativum</i> ) Lime ( <i>Citrus aurantifolia</i> )	[93]	
	Silvery pomfret	Chitosan	Gallic acid	[94]	
	Rainbow trout	Soy, whey, egg, wheat gluten, corn, collagen and fish proteins	–	[95]	
	Gilthead seabream fillets	Methylcellulose	<i>Satureja thymbra</i> (L.) essential oil	[96]	
	Fresh silver carp fillets	Methylcellulose	<i>Pimpinella affinis</i> essential oil	[97]	
	Surimi	Zein	Iron chelator	[98]	
Meat products	Cooked cured chicken breasts	k-carrageenan and chitosan	Mustard extract	[99]	
	Fresh chicken breasts	k-carrageenan and chitosan	Mustard extract	[100]	
	Pork meat	Sodium Alginate	Thyme and propionic acid	[101]	
		Oleic acid as part of starch	Lactic acid, nisin and lauric arginate	[22]	
	Dry-cured ham	Chitosan	Clove oil and/or ethylenediaminetetraacetate	[50]	
		Propylene glycol, xanthan gum and carrageenan with propylene glycol alginate	–	[102]	
		Ham	Soybean meal and xanthan	Lactoperoxidase	[103]
		Frankfurters and ham	Sodium alginate	Ethanol	[104]
Roast beef	Chitosan	Lauric arginate ester, lactic and levulinic acids	[105]		

### 5.1. Fish Products

In general terms, fish products are susceptible to fast degradation mainly due to their high content in lipids, moisture losses and deterioration of the sensory and chemical quality of their muscles. As a result, coatings based on proteins, polysaccharides and lipid materials have been recently applied as antimicrobial active packaging systems to extend the shelf-life of seafood by the reduction of pathogenic microorganisms at their surface [106]. In this context, important results have been reported to control the proliferation of *L. monocytogenes* on cold smoked salmon [107]. Neetoo et al. [108] also found that alginate coatings supplemented with 2.4% sodium lactate and 0.25% sodium diacetate solutions significantly delayed the growth of *L. monocytogenes* during a 30-day storage at 4 °C on cold-smoked salmon slices and fillets. As is seen in Table 2, changes in the quality of *Channa argus*, treated with thyme essential oil (1% v/v) added to chitosan (2% w/v) and poly(ethylene imine) (PEI) (1% v/v) edible compounds, were used as film-forming substrates, and they were studied for 10 days of storage at 4 °C [90]. Results underlined the beneficial effect of the addition of PEI or thyme essential oil in combination with the chitosan biopolymer matrix, reducing the total viable bacterial growth

and increasing by 4–5 days the shelf-life of the packaged product. In a different study, *S. aureus* or lactic acid bacteria were not detected in batches of fish sausages during chilled storage for at least 15 days when they were coated with chitosan (1% w/v) and warm-water fish gelatin (1% w/v). Shrimp (*Penaeus* spp.) cooking juice was also used as an antimicrobial agent at two different concentrations (1% and 2% w/v) [90]. In this study, both formulations showed antimicrobial activity against some fish spoilers and pathogenic organisms during chilled storage of Alaska pollock sausages. Enterobacteria counts in sausages wrapped in films of chitosan-gelatin-shrimp concentrate remained below the detection limit during storage (45 days), whereas lactic acid bacteria and *S. aureus* were not detected.

Chitosan was used as an edible antimicrobial coating of rainbow trout for storage at  $4 \pm 1$  °C for 16 days [91]. In this case, lactoperoxidase (LPO) is one of the most important enzymes used as a natural antimicrobial agent, since it has a broad antimicrobial spectrum, showing bactericidal effect on Gram-negative bacteria, a bacteriostatic effect on Gram-positive bacteria, antifungal and antiviral activities [107]. LPO (5% v/v) was coated onto a chitosan matrix (1.5% w/v), showing significant reduction in *Shewanella putrefaciens*, *Pseudomonas fluorescens* and psychrotrophic and mesophilic bacteria. Other combinations of LPO with glucose oxidase, D-(+)-glucose and potassium thiocyanate (1.00, 0.35, 108.70 and 1.09 weight ratio) were suggested, while chitosan concentration was always 1.5% (w/v). Reductions in growth rate for *S. putrefaciens*, *P. fluorescens*, psychrotrophic and mesophilic bacteria at 4 °C were observed, in particular for formulations of chitosan with LPO, since samples did not reach the control values ( $6\text{--}7 \log \text{CFU}\cdot\text{g}^{-1}$ ) in psychrotrophic and mesophilic bacteria after 12 and 16 storage days. Similar studies incorporated LPO at concentrations up to 7.5% (v/v) in whey protein coatings in rainbow trout preservation under refrigeration temperatures for 16 days [92]. Results suggested the bactericidal capacity of LPO, which is related to its high content in glucose-oxidase that produces oxidizing products and, consequently, decelerating the growth of certain microorganisms in fish products. Other studies used chitosan as supporting material for the development of antimicrobial coatings for fresh Indian salmon (*Eleutheronema tetradactylum*) fillets [93]. In this case, combinations formed by gelatin extracted from the processing waste of *Nemipterus japonicus* collected from a commercial surimi processing plant (10 g in 60 mL of distilled water) with 1.5 mL of chitosan solution and concentrations of 30%, 40% and 50% v/v of lime and garlic natural extracts were used as the coating material. These combinations resulted in acceptable protection to salmon up to 12 and 16 days, respectively. The garlic extract showed better antimicrobial activity than lime since lower total plate counts were reported in this study. Finally, gallic acid (0.206%, w/v) was added to the chitosan matrix (2%, w/v) as an edible coating for silvery pomfret (*Pampus argenteus*) stored at 4 °C for 15 days with a reduction of the microbial growth with lower total psychrotrophic count and total plate count values [94].

Rainbow trout is one of the most economically important freshwater-cultured fish species, and some authors have proposed different antimicrobial coatings to preserve their quality and nutrition facts against the microbial growth. Table 2 shows different coatings obtained from protein sources for such purpose. Thus, smoked rainbow trout (*Oncorhynchus mykiss*) was protected with different protein-based edible coatings, in particular soy protein isolates, whey protein isolates, egg white powder proteins, wheat gluten, corn proteins, gelatin, collagen and proteins from two different fish species (rainbow trout and Atlantic mackerel) in combination with vacuum packaging and refrigeration for a period up to six weeks [95]. It was reported that the formation of acetic acid solutions reduced pH to 3–4, and the formed ethyl alcohol showed some antimicrobial effect. The highest reductions in microbiological activity were obtained for samples coated with soy protein isolates, and this result was related to its high content in isoflavones, which reduced the growth of mesophilic aerobic bacteria. In addition, coatings of wheat gluten, gelatin, collagen and proteins obtained from rainbow trout and Atlantic mackerel fish also inhibited to some degree the bacterial growth. Zein has been also studied as edible coating with the incorporation of a polymeric iron chelator molecule, based on hexadentate 3-hydroxypyridinone, to preserve commercially-manufactured fish balls [98]. This product is a hot-water bath cooked surimi product, which is popular in many countries, but it is

highly perishable because of its high moisture content and abundance of fish muscle. The addition of 1 mg/mL of the iron chelator has been described as a potential antimicrobial coating since the inhibition zone of the four tested strains (two Gram-positive bacteria, *B. subtilis* and *S. aureus*, and two Gram-negative bacteria, *E. coli* and *Salmonella* spp.) significantly increased by using these edible coating films.

Methylcellulose is another polysaccharide widely used as an antimicrobial edible coating matrix. In this context, the effectiveness of the *Satureja thymbra* L. essential oil was evaluated as an antimicrobial coating of gilthead seabream fillets stored at 0 °C with a shelf life extension between 25% and 35% when fillets were coated with a combination of the essential oil (2%, v/v) and methylcellulose (1.5%, w/v), which was attributed to its high content in carvacrol [96]. In a different study, methylcellulose matrix (3%) was enriched with the *Pimpinella affinis* essential oil (1.5%, v/v) to be used as coating of fresh silver carp fillets for refrigerated storage (4 ± 1 °C) over a period of 16 days, showing the inhibition of total bacteria growth and psychrophilic counts [97].

## 5.2. Meat Products

Meat is quite easily contaminated by some pathogens, in particular by *L. monocytogenes*. In fact, one of the main challenges in the meat industry is to avoid the re-contamination by this microorganism in ready-to-eat products [109]. It was reported that the possibilities of contamination in meat products in the U.S. and Canada ranged from 0.4%–71% and 0%–21%, respectively [110]. The meat industry is prone to propose new strategies to eliminate this problem and the consequent economical losses through the development of innovative antimicrobial edible coatings supported in biopolymer matrices. For example, mustard extracts at 0.5% (w/v) were added to k-carrageenan- (0.2%, w/v) and chitosan-based (2%, w/v) coatings prepared using 1.5% malic or acetic acid [99]. These formulations resulted in the reduction of the viability of five different strains of *L. monocytogenes* on inoculated vacuum-packed, cooked and cured roast chicken slices at 4 °C due to their ability to hydrolyze sinigrin (a glucosinolate present in mustard extracts) with the formation of allyl isothiocyanate. These authors expanded the applicability of this coating to *Campylobacter* bacteria. The importance of this microorganism is due to its prevalence since it has been reported as the second responsible of food-borne illnesses with 145,350 and 845,024 cases per year in Canada and the U.S., respectively [111]. Then, the mustard extract in concentrations up to 300 mg/g was added to k-carrageenan- (0.2%, w/v) and chitosan-based (2%, w/v) coatings prepared using acetic acid (1%, v/v), and the effect against *Campylobacter jejuni* (*C. jejuni*) on vacuum-packaged fresh chicken breasts stored at 4 °C was studied [100]. They concluded that coatings containing 200–300 mg/g of mustard extract reduced the population of *C. jejuni* on chicken breasts compared to those coated with only k-carrageenan/chitosan.

Other antimicrobial agents have been proposed to obtain new bio-based edible coatings active against *L. monocytogenes*. In this context, thyme (*Thymus vulgaris* L.) and propionic acid (both at 0.5%, w/w) have been reported as effective antimicrobial agents when they were added into edible coatings based on sodium alginate and sorbitol, both at 1% w/w. In fact, these active agents improved the shelf life and safety of fresh chicken against *E. coli* and *L. innocua* [101].

A mixture of lactic acid, nisin and lauric arginate was added at different concentrations to an oleic acid nanoemulsion as part of a waxy starch-based edible coating (4%, w/v) to reduce the growth of *Brochothrix thermosphacta*, *L. monocytogenes* and *Micrococcus luteus* in pork meat [22]. The optimum combination of antimicrobial agents was 17.5 mg/mL of lactic acid, 3.75 mg/mL of nisin and 0.0625 mg/mL of lauric arginate showing the highest antimicrobial effect.

Raw meat shows high water activity and plenty of nutrients to allow bacterial growth, resulting in major losses on quality at cold temperatures, so the use of active edible films is a promising alternative to improve the meat quality. However, deeper research is necessary since most of these films did not improve the meat shelf-life significantly. Another alternative is based on the use of bioactive edible coatings based on chitosan (2% w/v) with clove oil (0.05% v/v) and/or ethylene-diamine-tetra-acetate (10 mM) to reduce *E. coli* and *S. aureus* growth on refrigerated lean pork slices at 4 °C [50].

Dry-cured ham is another major product of the meat sector, and contamination results in a serious problem, particularly in the main producers of this foodstuff (Spain and the U.S.) [112]. *Tyrophagus putrescentiae* infestation in cured ham has been reduced by coating with xanthan gum with 10%–20% propylene glycol and carrageenan/propylene glycol alginate [102]. The contamination by *Salmonella* has been reduced on ham slices by the application of an antimicrobial coating based on defatted soybean meal (26.6%) and lactoperoxidase (81 U/mg) extracted from bovine milk (22.3%) blended with xanthan (13.2%), glycerol (6.0%) and water (31.9%) [103]. Low levels of *L. monocytogenes* on Frankfurt sausages from pork meat and ham slices were achieved when samples were coated with Na-alginate edible films (1.5%, w/v) immersed in three different Greek alcoholic beverages, namely “tsipouro” (41% v/v ethanol), “raki” or “tsikoudia” (39.6% v/v ethanol) used as antimicrobial agents and stored under chilling conditions [104]. However, the same study revealed that the inadequate reheating after storage may not be enough to cancel the risks from *L. monocytogenes* populations on roast beef samples. Nevertheless, these risks were reduced under chilling temperatures for 30 days by approximately 0.9–0.3 log CFU/cm<sup>2</sup> when coated with 5% (w/w) chitosan in an acidic solution containing 2% (v/v) of acetic, lactic and levulinic acids and 20% (v/v) of lauric arginate ester [105].

In conclusion, these studies demonstrated the effectiveness of the new antimicrobial edible coatings against pathogens inoculated on different meat products, such as fresh chicken and pork, as well as on ready-to-eat meat products, such as dry ham, cooked ham and roast beef.

### 5.3. Fruits and Vegetables

Fruits and vegetables are characterized by their high nutritional value, particularly being rich in vitamins, minerals and fibers. However, their high water activity promotes fast degradation since they are constituted by living tissues susceptible to enzymatic browning, off-flavors development, texture breakdown and microbial contamination [113]. These undesirable properties, as well as the continuous increase in the consumer’s requirements have attracted the food industry’s interest in improving protection of fresh fruits and vegetables. Some alternatives have been proposed to reduce spoilage and microbiological contamination, increasing their shelf-life. Research efforts in this field have resulted in the development of new antimicrobial edible coatings. Table 3 summarizes the most relevant studies reported in the literature on the applicability of new antimicrobial coatings for fruits and vegetables.

**Table 3.** Antimicrobial coatings developed for fruits and vegetables.

Food Applicability	Product	Matrix	Antimicrobial Agent	Ref.
Fruits	Watermelon ( <i>C. lanatus</i> )	Sodium-alginate, pectin, and calcium lactate	Trans-cinnamaldehyde	[114]
	Cantaloupe melon	Chitosan and pectin	Trans-cinnamaldehyde	[115]
	Persimmon	Pectin, citric acid and calcium chloride	Nisin	[116]
	Strawberries	Pectin, pullulan, and chitosan	Sodium benzoate, Potassium sorbate	[117]
		Pectin and calcium chloride	Eugenol, Citral, Ascorbic acid	[118]
	Raspberries	Pectin and calcium chloride	Eugenol, Citral, Ascorbic acid	[119]
	<i>Arbutus unedo</i> L. fruit	Sodium alginate	Citral, Eugenol	[10]
	Pineapple	Sodium alginate	Lemongrass essential oil	[120]
	Fuji apples	Sodium alginate	Lemongrass essential oil	[35]
	Blueberry	Chitosan	Carvacrol, Cinnamaldehyde, <i>Trans</i> -cinnamaldehyde	[121]
Avocado	Gum arabic, aloe vera and chitosan	Thyme oil	[122]	

Table 3. Cont.

Food Applicability	Product	Matrix	Antimicrobial Agent	Ref.
Vegetables	Pepper	Chitosan	Lemongrass essential oil	[123]
		Pullulan	Leather bergenia leaves ethanolic extracts	[124]
		Pullulan	<i>Satureja hortensis</i> aqueous or ethanolic extracts	[125]
	Pumpkin	Xanthan gum, guar and chitosan	–	[126]
		Starch	Carvacrol	[127]
		Zein	Benzoic acid	[128]
	Cherry tomatoes	Zein	Cinnamon, Mustard essential oil, commercial wax	[129]
	Fresh cut broccoli	Chitosan	Bioactive compounds and essential oils	[130]
	Cauliflower florets	Maltodextrins and methylcellulose	Lactic acid, Citrus extract, Lemongrass essential oil	[131]
	Green beans	Modified chitosan	Mandarin essential oil	[132]
Rucola	Modified chitosan	Lemon, mandarin, oregano or clove essential oils	[133]	

Fresh-cut watermelon (*Citrullus lanatus*) was coated with three different solutions: sodium alginate, pectin and calcium lactate, and stored at 4 °C for 15 days. Glycerol was used as the plasticizer in all cases. Sodium alginate was added to the solution in three different concentrations (0.5, 1 and 2 g/100 g). Finally, *trans*-cinnamaldehyde encapsulated powder (2 g/100 g) was added to the solution as the active antimicrobial agent. Watermelon samples coated with sodium alginate showed the lowest counts on psychrotrophic and coliform microorganisms while also decreasing the growth of yeasts and molds [114]. Fresh-cut cantaloupe (*Cucumis melo* L.) was coated with chitosan (0.5, 1, 2 g/100 g, pectin (0.5, 1, 2 g/100 g), encapsulated in *trans*-cinnamaldehyde (1, 2, 3 g /100 g) and stored under chilling conditions at 4 °C [115]. The authors concluded that the highest concentration of the antimicrobial agent (3 g/100 g) was more effective (4.44 log cycles reduction) against the aerobic population than the uncoated sample by Day 15. However, coating with chitosan with no other active agents did not show significant inhibition for these microorganisms. Pectin biopolymers were also used to coat fresh-cut persimmon (*Diospyros kaki*) [116]. The edible coating was elaborated from a base solution of apple pectin at 10 g/kg emulsified with oleic acid and Tween80, whereas glycerol was added as the plasticizer. As anti-browning agents, 10 g/kg citric acid and 10 g/kg calcium chloride (CaCl<sub>2</sub>) were added into the coating solution. Finally, nisin was added as the antimicrobial agent at 500 international units (IU) per mL. Persimmons were peeled, cut and dipped into the coating solution for 3 min. After dipping, fruit pieces were removed and dried before being placed on polypropylene trays and sealed with polypropylene-polyethylene terephthalate film. After eight days of incubation, the inhibition of the mesophilic aerobic bacteria growth was observed while coating also reduced the populations of *E. coli*, *Salmonella enteritidis* and *L. monocytogenes*.

Coating strategies are very common in strawberry (*Fragaria ananassa*) processing as a consequence of their short post-harvest life, high metabolism and microbial decay [134]. Edible active coatings based on pectin, pullulan and chitosan with sodium benzoate and potassium sorbate have been reported as potential antimicrobial coatings for strawberries [117]. Microbiological analyses showed that the application of coatings reduced the total aerobic counts, molds and yeasts growth, with chitosan offering the best results in microbial growth tests. Edible coatings based on sodium alginate (1%, w/v) and pectin (2%, w/v) enriched with essential oils (citral at 0.15% and 0.3%, w/v, and eugenol at 0.1% and 0.2%, w/v) also showed their antimicrobial effect against aerobic mesophilic and psychrophilic bacteria, molds and yeasts [118]. Similar results were reported for raspberry coated with the same



formulations [119]. Edible coatings enriched with citral and eugenol were widely effective in reducing microbial spoilage. For example, essential oils or their constituents in alginate matrices are able to reduce microbial spoilage in fresh-cut pineapple [120] and fuji apple [35].

Other biopolymers used for coating fruit products have been also used as essential oil carriers in antimicrobial fruit active packaging. In this sense, chitosan has been reported as a coating for blueberries with the addition of three compounds with antimicrobial properties, carvacrol, cinnamaldehyde and *trans*-cinnamaldehyde (0.5%, w/v) [121]. Chitosan with this essential oils mixture was the most effective coating against mesophilic aerobic bacteria and also helped to reduce populations of bacteria and yeasts/molds. Antifungal effects against *Colletotrichum gloeosporioides* of gum arabic (10%, w/v), aloe vera (2%, w/v) and chitosan (1%, w/v) by themselves or in combination with thyme oil (1%, w/v) were studied on avocado fruit [122]. This study recommends the formulation of chitosan with thyme oil (3:1, v/v) as a potential antifungal coating for avocado storage.

Pepper has been widely coated onto different matrices since it is highly susceptible to chilling injury at temperatures below 7 °C [135]. However, at chilling temperatures, there is some enhancement in the rupture of the pepper surface and the consequent increase in susceptibility to contamination by different microorganisms, in particular by *Colletotrichum capsici*, the major causal agent of anthracnose. The addition of lemongrass essential oil at 0.5% and 1.0% (w/w) into 0.5 and 1.0% (w/w) of chitosan solution has been successful to control anthracnose in bell pepper [123]. In fact, the fungal growth was effectively controlled by 0.5% and 1.0% (w/w) lemongrass essential oil, whereas the application of 1.0% (w/w) of chitosan with 0.5% (w/w) of the lemongrass essential oil was effective as antimicrobial coating for bell peppers stored at room temperature for 21 days.

*Bergenia crassifolia* is another natural source to be considered in the protection of pepper. The antimicrobial effect of ethanolic extracts from bergenia leaves has been proven in pullulan coatings at different concentrations (0.4%, 1%, 2%, 5%, 10%, 20%, w/v) [124]. Samples coated with the antimicrobial solutions showed reductions in microbial growth by 1 log CFU/g when compared to control materials. A different coating for pepper to prevent the growth of Gram-positive and Gram-negative bacteria and *Penicillium expansum* has been recently described in the literature [125]. In this case, pullulan solution was prepared by dissolving pure pullulan (10%, w/v) and glycerol (5%, w/v) in distilled water. Then, aqueous or ethanol extracts of *Satureja hortensis* (20%, w/v) were added into the coating solution. Results showed that the formulation with the aqueous *Satureja hortensis* extract was more effective than ethanol against the growth of Gram-positive, Gram-negative bacteria and *Penicillium expansum*.

The development of different antimicrobial edible coatings for fresh-cut pumpkins (*Cucurbita moschata*) has increased in recent years [136]. Coatings based on different concentrations of xanthan gum, guar and chitosan reduced the growth of *Salmonella* ssp. [126]. Edible coatings with starch and carvacrol in minimally-processed pumpkin reduced the contamination by *E. coli*, *S. enterica* serotype *Typhimurium*, *Aeromonas hydrophila* and *S. aureus* [127]. Zein-based coatings with benzoic acid as the antimicrobial agent have been successfully tested on the quality of sliced pumpkin samples [128]. No mold growth was observed, and a final decrease in the total counts of mesophilic aerobic bacteria around the 1.0 log level was observed for coated samples. Zein has been also used as an edible matrix for coating cherry tomatoes [129]. The objective of this study was to investigate the effectiveness of zein-based coatings with propylene glycol (10%, w/v for both) in reducing populations of *S. enterica* serovar *Typhimurium* and preserving quality of cherry tomatoes. Then, a range of 5%–20% of cinnamon or mustard essential oils was added to the zein solution. On the other hand, a commercial wax formulation was also used in a different batch as an antibacterial agent. All of them were used to control the *S. enterica* dissemination in cherry tomatoes stored at 10 °C up to three weeks. As a result, the population of *S. Typhimurium* was reduced by 4.6 and 2.8 log CFU/g by the zein coatings with 20% cinnamon and 20% mustard oil, respectively. The same coating reduced populations of *S. Typhimurium* to levels below the detection limit. However, no antimicrobial activity was observed on the fruit coated with the commercial wax.

The antimicrobial properties of chitosan coatings (1%, v/v) enriched with four bioactive compounds (bee pollen, ethanol extract of propolis, pomegranate dried extract and resveratrol) and seven essential oils (tea tree, rosemary, clove, lemon, oregano, calendula and aloe vera) at different concentrations against mesophilic and psychrotrophic bacteria, *E. coli* and *L. monocytogenes* were studied on minimally-processed broccoli [130]. In vitro assays performed in tea tree, rosemary, pollen and propolis showed a remarkable inhibitory effects on *E. coli* and *L. monocytogenes*. Regarding *in vivo* analyses, in general terms, rosemary showed no significant effect on the reduction of the bacterial population. Chitosan coating with tea tree exerted a bacteriostatic effect on mesophilic and psychrotrophic bacteria counts with reductions around two order log lower than control sample up to seven days of storage. Chitosan coating with resveratrol or pomegranate produced a relevant reduction in mesophilic and psychrotrophic bacteria counts. Broccoli samples coated with chitosan and propolis showed a significant reduction in pathogen counts ( $1.0\text{--}2.0 \log \text{CFU}\cdot\text{g}^{-1}$ ), up to five days. When pollen was added to chitosan, a significant inhibitory effect in mesophilic and psychrotrophic bacteria counts ( $2.0\text{--}2.5 \log \text{CFU}\cdot\text{g}^{-1}$ ), compared to control samples, was observed. In a different work, cauliflower florets coated with maltodextrins (7.5 g/L) and methylcellulose (2.5 g/L) using lactic acid, citrus extract and lemongrass essential oil as antimicrobial agents at concentrations ranging from 0–34 mg/L were studied [131]. Complete inhibition of *L. innocua* after seven days of storage at 4 °C was reported. The same microorganism was reduced on green beans (*Phaseolus vulgaris* L.) coated with modified chitosan (3% N-palmitoyl chitosan, degree of palmitoylation 47%) containing 0.05% w/w nanoemulsion of mandarin essential oil [132]. Finally, modified chitosan (0.05% w/w) in 1% (v/v) lactic acid solution, enriched with 0.1% w/w of nanoemulsified lemon essential oil, was successfully used as antimicrobial coating of rucola during storage at 4 °C for three days and at 8 °C during 21 days [133]. A reduction of the initial microbial load around two log was obtained. After three days of storage under chilling conditions, the microbial load in coated samples remained constant with respect to Day 0, whereas the control showed a significant increase of about one log.

## 6. Market Analysis

The value of the global packaging market is expected to reach 910 billion euros by 2018. In particular, the worldwide consumption of flexible packaging is expected to reach 225 billion euros by 2020. In fact, the food industry covers more than two thirds of the current consumption of flexible packaging worldwide [137]. Particularly, novel edible coatings are emerging based on bio-based polymers with additives with specific functional properties to improve the shelf life of food products. In addition, coated food brings distinctive possibilities for the development of new products, processing improvement and general quality of the packaged food [11]. It has been observed that North America captured the highest market share in the global food coating ingredients market in 2015; this is majorly due to the increase in the confectionary market of the zone [138]. Europe accounted for the largest market of food coating ingredients for bakery and confectionery products in 2014 [139]. The market trend makes a rapid growth at a compound annual growth rate (CAGR) of 5.9% from 2014–2019 expectable; due to the changes in consumer lifestyles, increasing consumer disposable income and new developments to improve the organoleptic properties, to extend the shelf-life of food products and to provide the consumers safety [140].

## 7. Conclusions

This review underlines the most recent trends in the use of new edible coatings enriched with antimicrobial agents to reduce the growth of different microorganisms, such as Gram-negative and Gram-positive bacteria, molds and yeasts. The use of antimicrobials obtained from natural sources is one of the consequences of the rising consumer interest for healthy foods free of chemical additives. Among them, it is worth noting that organic acids and their salts (lauric, acetic, sorbic, citric, benzoic or propionic acids), spices and herb-derived compounds (essential oils and their main components),

chitosan and natural antimicrobials obtained from bacteria, such as nisin, pediocin, natamycin or reuterin, have been recently proposed as antimicrobial agents for edible coating formulations.

The main coating techniques in food packaging are spraying, dipping or spreading. In this context, spraying attracts the industrial interest in packaging in contrast to dipping or spreading mainly due to two different factors: firstly, the potential cost reduction by applying this technique; secondly, the high quality of the final product that could be achieved when compared to those products obtained by using conventional techniques. However, coatings have still limited applications in food packaging due to their poor barrier to water vapor and low mechanical properties. Blending with different biopolymers, the addition of hydrophobic materials such as oils or waxes or chemical modification of the biopolymers structure have been proposed to overcome these drawbacks.

Meat and fish products, fruits and vegetables are the most susceptible food products to be coated with antimicrobial edible films. Chitosan, gelatin, methylcellulose, soy, whey, egg, wheat gluten, corn and collagen have been recently reported as coatings of fish products. Regarding meat, k-carrageenan, chitosan, sodium alginate, xanthan gum and soybean meal are the main matrices. On the other hand, fruits have been coated with sodium alginate, pectin, chitosan, pullulan and gum arabic, whereas chitosan, pullulan, xanthan gum, agar, zein, maltodextrins and methylcellulose were used as coatings of vegetables.

In conclusion, the current situation reviewed in this study underlines the necessity of focusing future research on the selection of the appropriate antimicrobial agents and the most adequate polymer matrices, to ensure good interactions among them and effectiveness against the target microorganisms. Their applicability to the food packaging industry needs further and deeper studies, since some of them showed high impact on the organoleptic characteristics of food products. It will be also necessary to study in detail the possible interactions between the coating films and the packaged food. As a general conclusion of this review, antimicrobial edible coatings are ready to suppose an effective alternative in active packaging materials to improve the safety of processed food products for commercial purposes.

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