



Review

Starch-Based Coatings for Preservation of Fruits and Vegetables

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Received: 30 March 2018; Accepted: 22 April 2018; Published: 24 April 2018



Abstract: Considerable research has focused on the control of the physiological activity of fruits and vegetables in postharvest conditions as well as microbial decay. The use of edible coatings (ECs) carrying active compounds (e.g., antimicrobials) represents an alternative preservation technology since they can modify the internal gas composition by creating a modified atmosphere through the regulation of the gas exchange (oxygen, carbon dioxide, volatiles) while also limiting water transfer. Of the edible polymers able to form coating films, starch exhibits several advantages, such as its ready availability, low cost and good filmogenic capacity, forming colourless and tasteless films with high oxygen barrier capacity. Nevertheless, starch films are highly water sensitive and exhibit limited water vapour barrier properties and mechanical resistance. Different compounds, such as plasticizers, surfactants, lipids or other polymers, have been incorporated to improve the functional properties of starch-based films/coatings. This paper reviews the starch-based ECs used to preserve the main properties of fruits and vegetables in postharvest conditions as well as the different factors affecting the coating efficiency, such as surface properties or incorporation of antifungal compounds. The great variability in the plant products requires specific studies to optimize the formulation of coating forming products.

Keywords: edible coating; starch; antifungal; postharvest; preservation; fruit; vegetable; wettability

1. Introduction

Fruits and vegetables are essential in the human diet due to the health and nutritional benefits associated with their intake. However, they are products with a relatively short postharvest life, since they remain as living tissues up until the time they are used for consumption and are prone to physiological and biochemical changes, which can also have physical or pathological origins [1], leading to important economic losses [2,3]. Fruits and vegetables lose weight during postharvest handling and storage by transpiration, resulting in textural changes and surface shrinkage that affects their shelf life. On the other hand, softening of fruit during storage is also attributed to the deterioration of the cell wall components, mainly pectin, due to the activity of various enzymes.

Postharvest treatments with conventional synthetic waxes and/or chemical fungicides have been used for many years to control postharvest decay and extend fruit shelf life. However, the continuous application of these treatments has led to health and environmental issues, associated with chemical residues, or to the proliferation of resistant pathogenic strains. The increasing restrictions on the use of agrochemicals imposed by many countries and the growing consumer demand for high quality, minimally processed fresh food products have intensified the search for new preservation methods and technologies. The use of edible coatings (ECs) has emerged as an effective and environmentally-friendly alternative to extend their shelf life [4] and protect them from harmful environmental effects. Such films, applied as coatings, can create semipermeable barriers to gases and water vapour, reducing respiration

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and weight loss and maintaining the firmness of the fresh product while providing gloss to the coated products. In addition, coatings are able to act as carriers of a wide variety of functional ingredients, such as antimicrobials, antioxidants, anti-browning agents, nutrients or flavouring and colouring compounds [5,6], enhancing food stability, quality and safety, thus promoting the coatings' functional performance beyond their barrier properties [7]. Non-toxic antifungal compounds, incorporated in edible coatings, can prevent fungal decay, which is one of the main causes for deterioration of fruits and vegetables.

An EC is a thin layer of edible material coated directly on a food surface, applied in liquid form (film-forming solution/dispersion) on the food, usually by immersing or spraying [8]. The film-forming solution or dispersion contains a polymeric material with filmogenic capacity [9]. The film provides a barrier against water vapour and gases and thus lower levels of O_2 and higher levels of O_2 inside the fruit, which helps to control the enzyme activities, contributing to maintain the firmness of the coated product during storage.

The efficiency and stability of ECs depend on their composition. Polysaccharides, including starch, cellulose, pectin, alginates, chitosan and others, are naturally occurring polymers, widely used for this purpose [10], and are compatible with a broad range of functional compounds [11] whose aim is to improve their properties. Starch is a promising polysaccharide for food coating/packaging purposes, when taking into account its filmogenic capacity, ready availability and low cost. The starch world market can mainly be divided into four raw materials: corn, potato, sweet potato and cassava, although the predominant source used to obtain biodegradable plastics has been corn starch. This may be due to corn being the main source of starch produced worldwide (approximately 65%), followed by sweet potato (13%) and cassava (11%) [12].

Starch-based coatings are colourless and have an oil-free appearance, and can be used to increase the shelf life of fruits, vegetables and other products, although due to their hydrophilic nature, they are highly water sensitive and exhibit low water vapour barrier capacity. Other components, such as plasticizers and emulsifiers (or surfactants), may be added to the polymer matrix to improve the flexibility, extensibility and/or the stability of the polymer matrix structure. Blending (with other hydrophobic compounds to limit the hygroscopicity of starch-based materials has become an economical and versatile way to obtain new materials with better properties [13].

This paper reviews the starch-based ECs used to preserve the main properties of fruits and vegetables in postharvest conditions. The different methods used to determine or improve surface properties of coating materials when applied on fruits and vegetables, which greatly affect coating spreading and efficiency, have been analysed. Moreover, recent studies related with the application and/or characterization of mainly starch-based edible coatings, with and without antifungal properties to prevent fruit fungal decay, have been reviewed and their main conclusions summarized.

2. Requirements of the Coating-Forming Agents to Preserve Fruits and Vegetables

The most important functional properties of edible films and coatings are their barrier properties to water vapour and gases, compound migration, their ability for physical and mechanical protection and their impact on the product appearance (colour and gloss) [1]. The loss of quality in fresh products occurring during postharvest storage is associated with the biochemical and physiological changes in the live tissue, which is greatly affected by mass transfer phenomena, including moisture or oxygen exchanges, flavour loss or undesirable odour absorption [14,15]. Likewise, great losses in post-harvested products are due to microbiological alterations, mainly fungal decay, which shorten their shelf life and increase the risk of foodborne illnesses. Then, one of the main interests in coating design is the inclusion of substances with antimicrobial activity within polymeric matrices.

How effective EC is at protecting fruits and vegetables greatly depends on the product wettability to obtain a uniformly coated surface, which is influenced both by the fruit/vegetable surface properties and by the chemical composition and structure of the coating-forming polymers: the presence of different compounds, such as plasticizers, surfactants, antimicrobials or antioxidants [16]. The possible loss of

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these molecules during coating formation can affect the thickness of the film [3]. The EC effectiveness is also closely related with other factors, such as tensile properties. Mechanical resistance is important for two reasons: to prevent the film/coating fracture and to protect fruits and vegetables from mechanical factors and the physical damage caused by impact, pressure or vibrations during storage.

As regards the mass transfer properties of the coatings, the challenges include decreasing the water vapour permeability values in order to prevent both the moisture loss in the products (weight loss) and any changes in texture, flavour and appearance [17]. Both loss and gain of water are nearly always considered undesirable. The coating must also provide an adequate gas barrier (low oxygen permeability values), since this respiration process accelerates the consumption of sugars and other compounds, thus increasing the ethylene production and causing senescence [14]. In terms of their oxygen barrier properties, starch-based coatings usually stand out compared to other coating materials such as other polysaccharides or proteins [18]. However, in order to prevent anaerobic respiration, moderate barriers with a certain degree of oxygen and carbon dioxide permeability are needed for the respiration of living tissues [19]. In this sense, depending on the different respiration rates of the fruit or vegetable, a different minimum oxygen transfer rate may be needed to avoid unwanted metabolic changes. A sufficient gas barrier could also prevent fruits and vegetables from losing volatile flavour compounds or acquiring foreign odours. The good adherence and extensibility of the coating are key factors for the enhancement of the coating functions while also improving the appearance and attractiveness of the coated fruit or vegetable. Other considerations to take into account when formulating ECs are that some active ingredients might change the organoleptic profile of the coated product, causing undesirable odours or modifications in the functional properties. Some active compounds, such as essential oils, may cause toxicity in plant cells at high concentrations, or lose their functionality when reacting with external factors or food components [20]. Figure 1 shows the main relationships between the coating properties and the quality factors that are preserved in the fruit and vegetable. The interfacial interaction between the coating-forming agents, affected by its surface properties, and the product surface energy determine how effective the product coating is at exerting adequate protection.

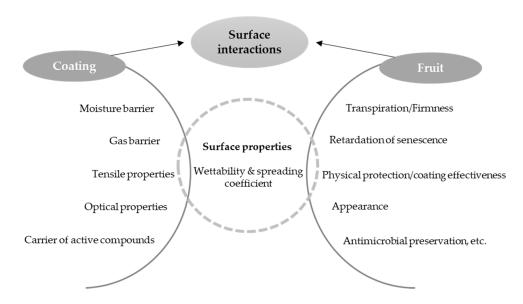


Figure 1. Relationship between coating properties and quality attributes of fruits and vegetables.

Factors Affecting the Coating Spreadability

The wettability of the product by a coating solution is of particular importance, as it is crucial when defining the ability of a coating to wet and spread uniformly on the surface of the fruit or vegetable. Wettability is studied by determining the values of the spreading coefficient (W_s) as a function of the

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works of adhesion (W_a) (Equation (1)) and cohesion (W_c) (Equation (2)). The equilibrium-spreading coefficient can be defined by Equation (3) [21] and can only be negative or zero. The adhesive forces promote the extension of a liquid on a solid surface and the cohesive forces its contraction. Consequently, the wetting behaviour is conditioned by the balance between these forces, and it is important to optimize the coating formulations in terms of biopolymer, plasticizer, surfactant, antimicrobial, antioxidant or other compound concentration in order to promote their spreading coefficient on a determined surface. In practical terms, the closer the W_s values are to zero, the better a surface will be coated.

$$W_a = \gamma_{LV} + \gamma_{SV} - \gamma_{SL} \tag{1}$$

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

$$W_s = W_a - W_c = \gamma_{SV} - \gamma_{LV} - \gamma_{SL} \tag{3}$$

The surface tension of the coating solution, as well as its contact angle (θ) on the target solid surface can be measured and then used to determine the W_s , through the estimation of the vapour-solid-liquid interaction. The contact angle of a liquid drop on the solid surface is defined by the mechanical equilibrium established under the action of the three interfacial tensions: solid-vapour (γ_{SV}), liquid-vapour (γ_{LV}) and solid-liquid (γ_{SL}) (Equation (4)).

Likewise, the interfacial tension can be separated into polar and dispersive components (Equation (5)) and, for pure liquids, the polar and dispersive components are known. If the surface contact angle between those liquids and the solid is obtained, the interaction can be described by Equation (6). This can be used to estimate the dispersive (γ_S^d) and polar (γ_S^p) components of the solid surface tension if at least three

pure liquids are used and the dependent variable $\left(\frac{1+\cos\theta}{2}\frac{\gamma_L}{\sqrt{\gamma_L^d}}\right)$ is plotted vs. the independent variable

$$\left(\sqrt{\frac{\gamma_L^p}{\gamma_L^q}}\right)$$
, from the intercept and slope of the fitted straight line, respectively.

Then, the determination of the solid surface energy or surface tension, which is a controlling factor in the wetting processes, involves the measurement of the contact angle of several standard liquids on the product surface in order to estimate the dispersive and polar contributions of the surface tension.

$$\cos \theta = (\gamma_{SV} - \gamma_{SL})/\gamma_{LV} \tag{4}$$

$$W_a = W_a^d + W_a^p \leftrightarrow W_a = 2 \cdot \left(\sqrt{\gamma_S^d \cdot \gamma_L^d} + \sqrt{\gamma_S^p \cdot \gamma_L^p} \right)$$
 (5)

$$\frac{1+\cos\theta}{2} \cdot \frac{\gamma_L}{\sqrt{\gamma_L^d}} = \sqrt{\gamma_S^p} \cdot \sqrt{\frac{\gamma_L^p}{\gamma_L^d}} + \sqrt{\gamma_S^d}$$
 (6)

The estimation of the critical surface tension (γ_c) can be carried out from the Zisman plots [22] by plotting the cosine of the contact angle of these pure liquids against their surface tension. From the fitted straight line, the intercept with $\cos\theta = 1$ corresponds to the critical surface tension, which is an imaginary point of the γ_{SV} , frequently used to describe the wettability of the solid surface. It represents the value of γ_{LV} of the liquid above which its spreading on the solid surface is complete (Equation (7)).

$$\gamma_C = \lim_{\gamma LV} \text{ as } \theta \to 0$$
 (7)

The reported values of the critical surface tension are generally lower than the solid surface tension values [23]. In Table 1, the surface and the critical surface tension of some fruits and vegetables are summarized. All of the fruit surfaces were found to be of low-energy and had the ability to participate in non-polar interactions, as a consequence of the higher values of the dispersive component of the surface tension.

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Fruit/Vegetable	Polar	Dispersive	Solid Surface	Critical Surface	Reference
Skin	Component	Component	Tension	Tension (γ_c)	
Mango	1.71	24.77	26.48	19.5	[24]
Apple	0.68	27.13	27.81	25.4	
Apple (cv. Golden)	0.68	27.13	27.81	25.4	[25]
Acerola	4.35	23.59	27.94	9.39	[26]
Cajá	2.29	27.86	30.15	23.92	
Mango	1.47	27.57	29.04	22.68	
Pitanga	3.07	23.88	26.95	13.42	
Seriguela	4.59	26.89	31.48	19.62	
Tomato	3.04	25.67	28.71	17.4	[27]
Carrot	0.34	26.13	26.48	24.1	
Strawberry	5.95	22.99	28.94	18.84	[28]
"Fuji" apple	-	-	-	18.7	[29]
Garlic	-	-	-	18.3	[30]
Orange Grapefruit	-	-	-	23.0	[31]

Table 1. Surface properties of fruit and vegetable skins/peels ($mN \cdot m^{-1}$).

How the composition of coating solutions and the addition of surfactants affect the wetting properties of different fruits and vegetables has been evaluated by several authors (Table 2). It has been reported that Tween 80 was effective at reducing the surface tension of different coating solutions, through a reduction in the cohesion forces, which improved the compatibility between the solution and the surface of the fruit skin, enhancing its wettability [25,29]. The authors of [27] found that increasing chitosan and glycerol concentrations in the coating solutions reduced both the W_s and W_a , but increased the W_c . On the other hand, different formulations of galactomannans and glycerol coatings showed good values of W_s when applied on different tropical fruits [26]. Ribeiro [28] obtained good wettability with a coating based on 2% starch and 2% sorbitol as a plasticizer applied on strawberry.

Upon drying, a coating with adequate cohesion and adhesion must be obtained, directly affecting its performance as a preservation agent [17,25]. Coating integrity also is a critical factor which depends on the film flexibility, surface tension and adhesion to the food product. Plasticizer-free matrices are too brittle and rigid because of strong interactions between the polymer chains and are incompatible with irregular surfaces, such as that of some fruits [32].

3. Starch-Based Coatings for the Preservation of Fruits and Vegetables

Starch, the reserve polysaccharide of most plants, can be obtained from different sources: cereals (corn, wheat or rice), legumes (pea) and tubers (cassava or potato) and is one of the most abundant natural polysaccharides used as a food hydrocolloid. This is because of its wide-ranging functionality, relatively low cost and great ability to form transparent, tasteless, odourless films, with very good oxygen barrier properties, which is very useful for food preservation purposes [33,34]. It has a granular structure and is composed of two macromolecules: amylose and amylopectin. Amylose is a linear polymer formed by glucose units linked by α -(1,4) whereas amylopectin is a highly branched polymer of glucose units with ramifications in α -(1,6) [35,36]. However, due to their hydrophilicity starch-based films/coatings exhibit water solubility and poor water vapour barrier properties [10]. It has been particularly studied in the context of the postharvest preservation of a variety of fresh horticultural products, including apples, oranges, strawberries and tomatoes [37].

To better control the weight loss during postharvest handling and storage caused by transpiration, one of the alternatives is to incorporate hydrophobic substances into the coating formulation. The authors of [38] reported a moisture loss restriction in pea starch-guar gum coatings with oleic acid and shellac, as well as a decrease in the respiration rates of treated fruit. Similar observations were described by [39] in oranges coated with polysaccharide-based coatings. In the same way, different plasticizers, especially sorbitol, had a significant effect on the delay in the change of weight and firmness of tomatoes coated with a non-conventional starch [40].

Table 2. Recent studies into the effect of different components and concentrations on the surface properties of coatings and their spreading coefficient (W_s) on fruits.

Polymer Matrix	Additives/Surfactants	Fruit/Vegetable	Main Results	Reference
Chitosan (0.5, 1.0, 1.5% w/v)	Glycerol (0.5, 1.0, 1.5% v/v) Aloe vera liquid fraction (0.5% v/v)) Tween 80 (0, 0.1, 0.2% w/v)	Blueberry	Coatings with 0.5% (w/v) chitosan + 0.5% (w/v) glycerol + 0.1% (w/v) Tween 80 + 0.5% (v/v) $A. \ vera$ liquid fraction presented the best W_s values (close to zero), to uniformly coat blueberry surface.	[41]
Galactomannans (seeds of <i>A. pavonina</i> and <i>C. pulcherrima</i>) (0.5, 1.0, 1.5% w/v)	Collagen (0.5, 1.0, 1.5% w/v) Glycerol (0, 0.5, 1.0, 1.5% v/v)	Mango Apple	The best W_s values for mango were obtained with blends of 0.5% of galactomannan from A . pavonina, 1.5% of collagen and 1.5% of glycerol ($W_s = -29.07 \text{ mN·m}^{-1}$). Blends of 0.5% of galactomannan from C . pulcherrima, 1.5% of glycerol-free collagen were the best for apples ($W_s = -42.79 \text{ mN·m}^{-1}$).	[24]
Galactomannans (seeds of <i>A. pavonina</i> and <i>C. pulcherrima</i>) (0.5, 1.0, 1.5% w/v)	Glycerol (1.0, 1.5, 2.0% v/v)	Acerola Cajá Mango Pitanga Seriguela	The wettability values ranged from -36 to -26 mN·m ⁻¹ . For the galactomannan from <i>A. pavonina</i> , the best W_s values were obtained for acerola (0.5% galactomannan and 1.0% glycerol) and seriguela (0.5% of galactomannan and 1.5% of glycerol). For mango, pitanga and cajá, coating solutions with 0.5%, 1.0% and 1.5% of galactomannan exhibited good W_s values at the different concentrations of glycerol used. When the galactomannan from <i>C. pulcherrima</i> was used, the best W_s value was obtained with solutions containing 0.5% of galactomannan, except for mango where the best W_s value was obtained with 1.5% of galactomannan.	[26]
Policaju (1.5 and 3.0% w/v)	Sorbitol (0.4% w/w) Tween 80 (0 and 0.1% w/v)	Apple	The best W_s values were found with the addition of 0.1% (w/v) Tween 80 (W_s : -29 and -26 mN·m ⁻¹ , for 1.5% and 3.0% (w/v) policaju, respectively). The addition of Tween 80 to the solution reduced the surface tension of the liquid through a reduction in the cohesion forces, thus enhancing W_s and improving the compatibility between the solution and the surface of the fruit skin.	[25]
Chitosan (1.0, 1.5, 2.0% w/v)	Glycerol (0.25 mL/g chitosan) Sorbitol (0.5 mL/g chitosan) Tween 80 (0.02–0.1% w/v)	Tomato Carrot	The increase in the concentration of chitosan and plasticizers reduced the values of the wettability and adhesion coefficients. The optimum W_s values were experimentally obtained with solutions of 1.5% (w/v) of chitosan and 0.1% of Tween 80 (w/w) as surfactant agent (W_s : -23 and -30 mN·m $^{-1}$, respectively, for tomato and carrot).	[27]
Starch (2% w/v) Carrageenan (0.3% w/v) Chitosan (1% w/v)	Sorbitol (2% w/v) Glycerol (0.75% w/v) Tween 80 (0.01–0.1% w/v). Tween 80 (0.01–0.1% w/v).	Strawberry	The addition of 2% sorbitol improved the wettability of the starch coating; however, the high surface tension of the carrageenan coatings led to high contact angles. For each polysaccharide-based coating, the best wettability was obtained for the following compositions: 2% starch and 2% sorbitol, 0.3% carrageenan, 0.75% glycerol and 0.02% Tween 80 or 1% chitosan and 0.1% Tween 80.	[28]

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Starch-based coating formulations (2 wt % starch) with 2% added sorbitol exhibited good wettability properties on the surface of strawberries. However, they were more permeable to O_2 , which was associated with the high concentration of plasticizer in the dry coating. In fact, plasticizers are used to decrease the intermolecular attractions between adjacent polymeric chains, which in turn facilitates the molecular mobility and diffusion of the gas molecules through the polymer network [28,42].

A representative summary of recent studies highlighting the positive effect of active edible coatings on the shelf life of fruit and vegetables is shown in Table 3, giving an overview of other compounds commonly used to improve the properties of starch-based coatings. Different results can be obtained and these not only depend on the kind of fruit or vegetable but also on the coating composition. In general, the incorporation of lipids into starch or polysaccharide-based coatings contribute to the reduction in the amount of water lost in the coated product and can also affect the gas exchanges, depending on the lipid ratio and its physical state. Solid lipids generally offer better resistance to the mass transfer of water or gas molecules than liquid lipids [43]. The lipid-polymer ratio is also an important factor, since lipids provoke interruptions in the polymer matrix which have a great impact on the coating performance. In this sense, the adequate compatibilization of lipids and polymers and the final size distribution of lipid particles in the film also affect the coating's functional properties [44]. As concerns starch coatings, the amylose-amylopectin ratio affected their functionality due to the different structure of the films generated with high-amylose starch, where linear amylose, with regions of helical conformation, exhibited better packed domains in the matrix that is more crystalline in nature [36]. The authors of [37] found that coatings made with high-amylose starch better preserved the weight losses and firmness of strawberries for longer periods than coatings of medium-amylose starch. Likewise, these authors observed that coatings plasticized with sorbitol exhibited a better water vapour barrier capacity than those containing glycerol. In this sense, the authors of [45] report the use of amylose-only starch obtained from transgenic plants to obtain starch materials with improved properties.

In general, coatings help to better retain different active compounds incorporated into their formulation, such as antimicrobials, on the product surface for longer storage times, thus enhancing their effectiveness. The authors of [11] observed a better retention of potassium sorbate and antifungal activity during the refrigerated storage of apple, tomato and cucumber coated with starch-based formulations.

In fresh-cut products, the effects of starch-based coatings can also be beneficial for the maintenance of quality and safety during storage. Coatings with a starch:protein ratio of 15:85, with an added 6% (v/w) of pink pepper phenolic compounds, prevented enzymatic browning in fresh-cut apples for 12 days [46] and cassava starch coatings decreased the respiration rate, preserving the mechanical properties and colour characteristics, of fresh-cut mango that was pre-treated with citric acid (0.5% w/v)and peracetic acid $(0.05\% \ w/v)$ [47]. In general, a good performance could be achieved with starch-based coatings in different fruits or vegetables, forming edible barriers at a competitive cost. However, other components, such as plasticizers, surfactants, lipids or other more hydrophobic polymers, must be incorporated into the coating-forming formulation in order to obtain a good adherence on the target product surface, an adequate mechanical resistance of the film and optimal barrier properties against water vapour or gas. Likewise, taking into account the different physiology and surface properties of the plant products, an optimal formulation must be developed for the different products; this must include active compounds permitting the control of fungal or bacterial decay which limits the product shelf life. In this sense, new approaches must make use of compounds that are innocuous for human health and effective at controlling microbial growth or physiological processes. The following sections analyse the new tendencies in this field.

Table 3. Starch-based coatings applied to fruits and vegetables (polymer and additives concentration in the coating-forming dispersion, except when indicated with respect to the total polymer).

Polymers	Additives	Fruit/Vegetable	Properties Evaluated in Coated Product	Main Results	Reference
Pea starch (2.5% w/v) Guar gum (0.3% w/v)	* Glycerol (25% w/w) * Shellac (40% w/w) * Oleic acid (1% w/w) Tween 20 * With respect to the polymer	Orange	Weight loss, firmness, respiration rate, ethylene production, colour, acetaldehyde and ethanol concentrations (fruit juice), peel pitting index, fruit decay, stem-end rind breakdown, overall visual acceptability and sensory evaluation.	The incorporation of lipid compounds into pea starch-guar gum coatings reduced fruit respiration rate, ethylene production, weight and firmness loss, peel pitting, and fruit decay rate index.	[38]
Mango kernel starch $(4\% w/v)$	Glycerol (2% w/v) Sorbitol (2% w/v)	Tomato	Weight loss, firmness, total soluble solids, total titratable acidity, ascorbic acid, fruit decay and sensory evaluation.	The formulations containing sorbitol were the most effective at maintaining the overall quality of the tomato fruit during storage	[40]
Corn starch (2% w/w)	* Glycerol (ratio 0.15) Aloe vera (ratio A. vera:starch 1:3) * With respect to the polymer	Tomato	Fruit appearance and weight loss.	Coating retarded weight loss in the fruits stored first at 10 °C and 85% RH for 7 days, and subsequently at 25 °C and 85% RH for 7 days. After 14 days of storage, the tomatoes without coatings exhibited a weight loss that was 84 times greater than the coated ones.	[48]
Cassava starch (2% w/v)	Cinnamon essential oil $(0.01\% \ w/v)$	Guava	Weight loss, firmness, total and soluble pectin and pectin methylesterase.	The treatment with starch and essential oil reduced mass loss and better preserved greenness for the 8 days of storage, compared to the control.	[49]
Cassava starch (2.0% <i>w/v</i>) Chitosan (0.5, 1.0, 1.5, 2.0% <i>w/v</i>)	Mixture of <i>Lippia gracilis</i> Schauer genotypes in an EOs solution (EOM) $(0, 1.0, 2.0, 3.0\% \ v/v)$ Glycerol $(0.64\% \ w/v)$ Glycerol $(1.28\% \ w/v)$	Guava	Firmness, colour, pH, titratable acidity, total soluble solids	The incorporation of 1% or 3% EOM into edible chitosan-cassava starch coatings delayed the ripening process, reduced browning and inhibited colour development to a greater extent than only chitosan or cassava starch coatings.	[50]
Gelatin ($10\% \ w/v$) Corn starch (native, waxy or modified waxy) (3.0 or $5.0\% \ w/w$) (Starch:gelatin blends 1:1)	Sorbitol (ratio polymer: plasticiser 1:0.1)	Grape	Weight loss and sensory evaluation.	Improved appearance was observed in coated grapes after 21 storage days under refrigerated conditions, with a reduced weight loss compared to the control group. Sensory evaluation showed that coatings did not affect the acceptability scores.	[51]
Corn starch (2% w/v) Arabic gum (2% w/v)	Glycerol (10% w/w) Sorbitol (10% w/w)	Green banana	Weight loss, firmness, colour	Coated fruits lose about 30% less weight than the uncoated fruits. The coating application was effective at maintaining the firmness of banana and slowed down the ripening process.	[52]
Pea starch $(4\% w/v)$ Potato starch $(4\% w/v)$ Guar gum $(1\% w/v)$	Glycerol (ratio (polymer:glycerol 2:1) Potassium sorbate (KS) (1% w/v)	Apple Tomato Cucumber	Coatings weight and thickness, KS residual surface concentration, yeast and mould count	All the coatings better retained KS concentration on the fruit surface during refrigerated storage to provide effective antifungal activity.	[11]

 Table 3. Cont.

Polymers	Additives	Fruit/Vegetable	Properties Evaluated in Coated Product	Main Results	Reference
Rice starch (1.0, 1.5 and 2.0% w/v)	Glycerol (0.4, 0.5 and 0.6 mL) Coconut oil Tea leaf extract	Tomato	Weight loss, total soluble solids, titratable acidity, ascorbic acid content, colour and microbial count.	Starch coating, with added glycerol, lipids and antioxidant compounds, delayed the change in colour of tomatoes, which can be directly attributed to the antioxidant activity of the green tea extract. It was also found to form a rigid and continuous fruit coating that was able to extend the ripening period of tomatoes in storage at room temperature.	[53]
Cassava starch (1.0, 2.0 and $3.0\% \ w/v$)	Potassium sorbate (0, 0.05 and $0.1\% \ w/v)$	Strawberry	Firmness, colour, sensory evaluation, coating integrity and respiration rate.	Cassava starch coatings, with or without potassium sorbate, did not cause changes in strawberries' mechanical properties, colour or sensory acceptance. Coatings showed good integrity for 2% and 3% starch, reducing the strawberries' respiration rate.	[54]
Starch (2% w/v) Carrageenan (0.3% w/v) Chitosan (1.0% w/v)	Sorbitol (2% w/v) Glycerol (0.75% w/v) Tween 80 (0.01–0.1% w/v) Calcium chloride	Strawberry	Weight loss, firmness, total soluble solids, colour and total microbial count	Starch coatings were less effective at reducing loss of firmness than chitosan and carrageenan films, which better reduced the fruit weight loss. The minimum firmness loss was obtained with carrageenan and calcium chloride coatings.	[28]
Starches with medium amylose content (MAS) (corn and potato starch); Starches with high amylose content (HAS) (corn starch, genetically modified and acorn starch product (HAP) (2% w/v)	Glycerol and sorbitol $(0, 1.0\%$, and $2\% w/v)$	Strawberry	Coating's water vapour permeability (sliced carrots), weight loss, firmness, anthocyanin content, surface colour, reducing and total sugar content and titratable acidity, soluble, insoluble and total solids and microbiological assays.	Starch coatings with the higher amylose content reduced fruit weight losses and retained fruit firmness for longer periods than coatings with medium amylose content starches. Both sorbitol and glycerol contributed both to reducing weight loss and to maintaining texture and surface colour of fruits. Coatings with sorbitol exhibited better water vapour barrier capacity than those containing glycerol. Sorbitol at 2% w/v was the most effective plasticizer option.	[37]
Rice starch/Fish protein (3 g/mL of total solids) (15/85, 50/50 and $85/15 w/w$)	Pink pepper phenolic compounds (4.0, 6.0 and 8.0% v/w) Glycerol (25 g/g of total solids w/w)	Fresh-cut apples	Colour, browning index, firmness, mass loss, total soluble solids, pH and acidity.	The starch/protein blend (15/85) with 6% (v/w) pink pepper phenolic compounds better preserved fresh-cut apples for 12 days, especially in terms of the inhibition of enzymatic browning.	[46]
Cassava starch (1.0% w/v)	Glycerol (1.0% w/v)	Fresh-cut mango (Pre-treated with $0.5\% \ w/v$ citric acid and $0.05\% \ w/v$ peracetic acid)	Weight loss, respiration rate, firmness, β -carotene content, colour, sensory evaluation and microbiological assays.	Cassava starch coatings, combined with citric acid dipping, promoted a decrease in the respiration rate, the better preservation of mechanical properties and colour characteristics and great sensory acceptance. The use of glycerol in the coating formulation promoted a greater weight loss, impairing fruit texture and increasing carotenogenesis.	[47]

RH: relative humidity; EO: essential oil; EOM: essential oils mixture.

4. Antifungal Coatings for Fruit Preservation

During storage, fruits are often subjected to different levels of microbial decay, mainly due to phytopathogenic fungi, which usually infect the host through wounds sustained during harvesting, handling and/or processing [55]. Fungal disease is mainly controlled chemically, but the use of synthetic fungicides is limited due to undesirable aspects, including the toxicological hazard to human health and slow degradation periods, which could lead to environmental problems [56]. The negative public perception of industrially-synthesized food antimicrobials has generated interest in the use of more naturally occurring compounds.

As reported by [1], there are three categories of antifungal agents that can be incorporated into ECs: (a) synthetic food preservatives or GRAS (Generally recognized as safe) compounds with antimicrobial activity, including some organic and inorganic acids and their salts (benzoates, sorbates, carbonates, propionates, etc.) and parabens (ethyl and methyl parabens) and their salts, (b) natural compounds, such as essential oils (EOs) or plant extracts (thyme oil, carvacrol, cinnamon, cinnamaldehyde, citral, eugenol, lemongrass, oregano, rosemary, etc.); and (c) antimicrobial antagonists (yeasts or bacteria).

The use of antifungal compounds, such as organic acids and various plant extracts or EOs, are among those mostly antimicrobial substances studied as a possible means of controlling the growth of phytopathogens in fruits and vegetables during postharvest shelf life [57]. There are a great number of in vitro and in vivo (inoculated fruits and vegetables) studies dealing with this topic, in which different active compounds have been tested against different fungi through their direct application or incorporated into film-forming formulations. In Table 4, different studies analysing the in vitro antifungal activity of different natural extracts or essential oils are summarized. Likewise, Table 5 shows several in vitro studies on the antifungal effect of these kinds of products incorporated into different polymer matrices to obtain active films. Studies into the application of these kinds of coatings to fruits or vegetables so as to extend their shelf-life are shown in Table 6, where the main findings of the authors are remarked on.

The antifungal activity of some plant extracts, such as *Aloe vera*, can be attributed to the presence of bioactive compounds, such as quinones and phenol compounds (flavonoids), which can be more or less active against different fungi. Moringa plant extracts had a significant effect on the growth rate of *C. gloeosporiodes*, *A. alternata* and *L. theobromae*, a fact that can be closely linked to the high concentration of phenolic compounds in the tissue [58]. *B. cinerea* and *P. expansum* were better inhibited than *A. niger* in the presence of pulp or liquid fractions of *A. vera* [41]. *Aloe vera* gel was also effective at controlling the growth of several fungi, exhibiting the greatest efficacy against *F. oxysporum*. When incorporated into corn starch matrices in a *Aloe vera* solids–starch ratio of up to 1:1, using glycerol as plasticizer, these coatings were effective at controlling fungal decay in cherry tomatoes and were a natural, non-toxic alternative to synthetic fungicides [48].

EOs extracted from plants are rich sources of bioactive compounds (terpenoids, phenolic compounds) which have been recognized as antifungal agents [55]. In order to achieve effective antimicrobial activity, high concentrations of essential oils are generally needed. The incorporation of essential oils into biopolymer matrices (active coatings) can be a useful strategy to improve coating functionality in terms of the enhancement of the antimicrobial properties of the coatings, while reducing the matrix's hygroscopic character. Likewise, the incorporation of EOs into the coating matrix allows for a reduction in the cost and minimizes their intense aroma perception [59]. The authors of [60] observed an enhanced in vitro antifungal activity against *B. cinerea* of chitosan films after the incorporation of lemon essential oil, although a decrease in the inhibition capacity was reported after two days of storage. This decrease was explained by considering the changes in the availability of the antimicrobials, controlled by the rate of diffusion of the active compounds into the agar medium throughout the storage period and the progressive evaporation of volatiles.

Edible composite coatings based on hydroxypropyl methylcellulose, hydrophobic components (beeswax and shellac) and food preservatives with antifungal properties (potassium sorbate, sodium benzoate and sodium propionate), were effective at reducing the incidence of *P. digitatum* and *P. italicum*

during the long-term cold storage of mandarins [61]. Lemongrass essential oil has been observed to be effective as a treatment for the control of anthracnose (*C. capsici*) of bell pepper in vitro, due to the presence of numerous secondary compounds. This was attributed to the ability of the oil to penetrate through the cell membrane, resulting in deteriorative biological processes. However, the antimicrobial activity of the EO was higher in the in vitro tests than that observed in in vivo applications on bell pepper, which was attributed to the inability of the oil to adhere to the bell pepper surface [62].

Biological control is also a promising alternative to unpopular synthetic fungicides, and research into postharvest biocontrol has increased in recent decades [63,64]. The main characteristics of an ideal biocontrol agent (BCA) are related to its biosafety, activity in a range of environments and against a variety of pathogens, and ease of management and use [65]. Extensive research has been developed to understand the mechanisms by which BCAs exert their action against pathogens. Nevertheless, in many cases, the suggested modes of action whereby antagonists wield their biocontrol effect have not been totally elucidated. Competition for nutrients and space between the pathogen and the antagonist is considered to be the main mode of action, but other mechanisms, such as parasitism or the production of secondary metabolites, have also been reported. Several microorganisms, which can act as microbial antagonists, have received considerable attention as controlling agents of diseases in fruits. Table 7 shows some examples of BCA applied to the preservation of different fruits.

The combined application of BCAs and edible coatings or films offers many possibilities, both because of the wide variety of matrices which can be used and their potential benefits for the survival and retention of the antagonists. In this sense, the coating-forming formulations should contain components which not only allow for coating formation, but are also compatible with the cells and provide them with an adequate substrate for nutrition and growth [64]. Marín [66] evaluated the effect of different coating-forming systems containing *C. sake* CPA-1 as BCA, based on different biopolymers (corn starch, hydroxypropylmethylcellulose, sodium caseinate, or pea protein) with and without surfactants, on the adherence, viability and survival of *C. sake* cells, as well as on their biocontrol efficacy against *B. cinerea* infections of coated grapes. Taking into account the relative increase in the survival and efficacy of *C. sake*, and the cost of ingredients, sodium caseinate or corn starch were the most suitable coatings with which to obtain formulated biocontrol *C. sake* products. Likewise, the authors of [67] developed dried formulations of *C. sake* CPA-1 based on starch derivatives, which exhibit good stability at a very low moisture content. These formulates can be used as antifungal coating agents when applied on different fruits that are susceptible to *B. cinerea* infections, after their dispersion in water at the adequate concentration.

Table 4. Recent studies into natural antifungal compounds with potential use in fruits and vegetables.

	Direct Application							
Antifungal Component	Fungus Tested	Antifungal Test	Result	Reference				
Moringa leaf (LE) and seed (SE) extracts (methanolic and ethanolic extraction methods)	Colletotrichum gloeosporiodes Alternaria alternata Lasiodiplodia theobromae	Mycelial plugs (3 mm diameter) placed on PDA plates. Evaluation: Radial growth inhibition.	Ethanolic LE and SE were less effective at reducing the GR of <i>C. gloeosporioides</i> and <i>A. alternata</i> . Methanolic extracts were the least effective against the isolates, except for <i>L. theobromae</i> which was relatively inhibited (5%) after ten days.	[58]				
Aloe vera gel	Fusarium oxysporum Alternaria alternate Colletotrichum gloeosporioides Bipolaris spicifera Curvularia hawaiiensis Botryotinia fuckeliana	Mycelial plugs (8 mm diameter) placed on PDA plates. Evaluation: Radial growth inhibition.	The GR of the fungi decreased when <i>A. vera</i> was present in the medium. <i>C. gloesporoides</i> showed the lowest MGI values (7 days), and the greatest inhibition occurred for <i>F. oxysporum</i> .	[48]				
Basil and thyme essential oil	Aspergillus niger Botrytis cinerea Rhizopus stolonifer	Spore suspension (0.1 mL of 10 ⁴ spores/mL) on PDA plates. Filter paper circle (2.5 cm diameter) embedded with different amounts of oils (3.5, 7.0, and 14 mg/plate)	A notable inhibition halo was detected for basil and thyme oil from 14 mg of oil per plate and no effect was observed for smaller amounts.	[44]				
Aloe vera extract (pulp and liquid fractions)	Botrytis cinerea Penicillium expansum Aspergillus niger	Spore suspension (10^4 spores/mL sterile water), using sterile well microplates. Fungal growth was monitored spectrophotometrically at 530 nm by measuring optical density.	B. cinerea and P. expansum presented greater growth inhibition in the presence of A. vera extracts than A. niger. With 0.5% of A. vera liquid and pulp, an inhibition of around 80% was observed for B. cinerea. With 100% of each A. vera fraction, growth was not observed over time (72 h).	[41]				
Lemongrass essential oil	Colletotrichum capsici	Mycelial plugs (2 mm diameter) placed on PDA plates. Evaluation: Radial growth inhibition.	Lemongrass essential oil was an effective treatment for the in vitro control of anthracnose.	[62]				
Food preservatives: Ammonium carbonate, ammonium bicarbonate, potassium carbonate, potassium bicarbonate, potassium silicate, potassium sorbate, sodium carbonate, sodium bicarbonate, sodium acetate, sodium diacetate, sodium benzoate, sodium formate, sodium propionate, sodium methylparaben, sodium ethylparaben	Monilinia fructicola	Mycelial plugs (5 mm diameter) placed on PDA plates. Evaluation: Radial growth inhibition.	Almost all the agents completely inhibited the radial growth of the fungus at various concentrations. Ammonium carbonate, ammonium bicarbonate and sodium bicarbonate were the most effective agents, as they completely inhibited the mycelial growth of <i>M. fructicola</i> on PDA at all concentrations tested.	[4]				

GR: Growth rate; PDA: potato dextrose agar; MGI: mycelial growth inhibition.

Table 5. Recent studies on natural antifungal compounds incorporated into coating-forming formulations (in vitro tests) with potential use in fruits and vegetables.

Application to Coatings (In Vitro)						
Compound	Fungus	Antifungal Test	Coating Matrix	Main Findings	Reference	
Cinnamon and ginger oil	Aspergillus niger	Spore suspension (0.1 mL of 10^6 spores/mL) on PDA plates. Film discs (4 mm) were placed on the inoculated surfaces.	СН-СМС	Cinnamon oil was more effective than ginger oil at reducing fungal growth.	[68]	
Basil and thyme essential oil	Aspergillus niger Botrytis cinerea Rhizopus stolonifer	Spore suspension (0.1 mL of 10 ⁴ spores/mL) on PDA plates. Contact and head space methods. Film circles (2.5 cm diameter) were placed on PDA (contact method) or fixed to the plate cover (head space method).	СН	CH films with thyme or basil essential oils did not inhibit the GR of the tested fungi.	[44]	
Chitosan Cinnamon leaf essential oil	Aspergillus niger Botrytis cinerea Rhisopus stolonifer	Spore suspension (0.1 mL of 10^4 spores/mL) on PDA plates.	СН	Chitosan-cinnamon leaf essential oil films exhibited antifungal activity against the tested fungi.	[69]	
Lemon essential oil	Botrytis cinerea	Spore suspension (0.1 mL of 10^5 spores/mL) on PDA plates. Film discs with the same diameter as the Petri dishes were placed on the inoculated surfaces.	СН	CH film led to reduction in the GR of <i>B. cinerea</i> . The antifungal activity of CH films was enhanced by the addition of the essential oil.	[60]	
Cinnamon, clove and oregano essential oil	Colletotrichum gloesporoides Fusarium oxysporum	Spore suspension (5 \times 10 ⁶ spores/mL spores) on PDA plates. Film discs (2.4 cm diameter) were placed on the inoculated surfaces.	S-G	Films containing cinnamon essential oil were more effective against the <i>F. oxysporum</i> fungus, while films with clove and cinnamon essential oils were more active against the <i>C. gloeosporiodes</i> fungus. The films with oregano essential oil were always less effective than the other two.	[70]	
Mexican oregano, cinnamon and lemongrass essential oils	Aspergillus niger Penicillium digitatum	Spore suspension (10 μ L of 10 ⁶ spores/mL) on PDA plates. Vapor contact assay. Film discs with the same diameter as the Petri dishes were placed on the inoculated surfaces.	A, CH, S	CH films incorporated with Mexican oregano or cinnamon essential oils inhibited <i>A. niger</i> and <i>P. digitatum</i> by vapor contact at lower essential concentrations than those required for amaranth and starch edible films.	[71]	

GR: growth rate; PDA: potato dextrose agar; CH: chitosan; CMC: carboxymethyl cellulose; S: starch; G: gelatin; A: amaranth.

Table 6. Natural antifungal compounds applied to coatings (in vivo tests) to preserve fruits and vegetables.

Application to Coatings (In Vivo)								
Compound	Fungus	Antifungal Test	Matrix	Fruit/Vegetable	Result	Reference		
Moringa leaf (LE) and seed (SE) extracts	Colletotrichum gloeosporiodes Alternaria alternata	Inoculation in wounded fruit	CMC	Avocado	Compared to the untreated fruit, the LE and SE + 1% CMC coated avocados had lower disease incidence and severity	[58]		
Aloe vera gel	Fusarium oxysporum	Inoculation of 5 μ L of a 10 ⁶ espores/mL, before coating application.	S	Cherry tomato	Coatings improved the preservation of cherry tomatoes by reducing fungal decay.	[48]		
Cinnamon essential oil	Colletotrichum gloeosporiodes	Non-inoculated	S	Guava	Cassava starch, associated with cinnamon EO, was effective at controlling anthracnose and preserving fruit quality. On the eighth day of storage, the fruits treated with EO were free of anthracnose and similar to the day of harvest.	[49]		
Aloe vera extract (liquid fraction)	Botrytis cinerea	Inoculation of 10 μ L of 10 ⁴ spores/mL, before coating application	СН	Blueberry	A fungistatic effect was observed during the storage period.	[41]		
Lemongrass essential oil	Colletotrichum capsici	Inoculation of 10 μL of $10^5 spores/mL$, before coating application	СН	Green bell pepper	EO was found to be less effective in vivo than in vitro, but the combination of EO with CH did enhance the antimicrobial activity of the coating.	[62]		
SMP, SEP, SB	Alternaria alternata	Inoculation of 10 μL of 10^6 spores/mL, before coating application	HPMC and BW	Cherry tomato	Antifungal coatings reduced the incidence and severity of alternaria black spot on inoculated cherry tomatoes, with the SB-based coating being the most effective.	[5]		
Food preservatives: Ammonium carbonate, ammonium bicarbonate, potassium silicate, potassium sorbate, sodium carbonate, sodium diacetate, sodium benzoate, sodium propionate, sodium methylparaben, sodium ethylparaben	Monilinia fructicola	Immersion in a spore suspension (10^3 spores/mL) before coating application	HPMC and BW	Plum	Coatings containing bicarbonates and parabens reduced incidence of brown rot in plums, and potassium sorbate at 1.0% was the most effective agent. All the tested coatings reduced severity of disease to some extent.	[4]		
Chitosan Cinnamon leaf essential oil	Rhizopus stolonifer	Immersion in a spore suspension (10 ⁵ spores/mL) before coating application.	СН	Strawberry	All the coatings were effective at extending the shelf-life of cold-stored strawberries.	[69]		
Lemon essential oil	Botrytis cinerea	Immersion in a spore suspension (10 ⁵ spores/mL) before coating application	СН	Strawberry	CH coatings reduced the percentage of infected strawberries as compared to non-coated ones after three storage days, especially when lemon essential oil was incorporated.	[60]		
Bergamot, thyme and tree oils	Penicillium italicum	Immersion in spore suspension (10^5 spores/mL) . Preventive and curative assays	СН	Orange	The greatest antifungal effectiveness against <i>P. italicum</i> was obtained for preventive treatments with coatings containing tea tree oil. Curative treatments were less effective and, in this case, the coatings with thyme oil showed the greatest antifungal activity.	[39]		
PS, SB, SP, and their mixtures	Penicillium digitatum Penicillium italicum	Inoculation of spore suspension (10 ⁵ spores/mL) before coating application	HPMC, BW and shellac	Mandarins	All the coatings reduced the incidence of green and blue moulds after 2 weeks of cold storage and the severity of the disease after 6 weeks of storage.	[61]		

CMC: carboxymethylcellulose; S: starch; CH: chitosan, HPMC: hydroxypropyl methyllcellulose; BW: beeswax; SMP: sodium methyl paraben; SEP: sodium ethyl paraben; SB: sodium benzoate; PS: potassium sorbate; SP: sodium propionate.

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Table 7. Representative antagonistic fungi and yeasts used as biocontrol agents for the preservation of fruits

Biocontrol Agent	Source	Pathogen	Application	Reference
Cryptococcus podzolicus	Soil	Penicillium expansum	Apple	[72]
Hanseniaspora opuntiae Metschnikowia pulcherrima	Breva crops	Penicillium expansum	Cherry	[73]
Metschnikowia pulcherrima	Fig	Botrytis cinerea Cladosporium cladosporioides Monilia laxa Penicillium expansum	Apple Nectarine	[65]
Pichia membranaefaciens	-	Colletrotichum gloeosporioides	Citrus	[74]
Trichoderma spp.	Soil	Fusarium oxysporum	Melon	[75]
Cryptococcus laurentii	Pear	Penicillium expansum	Pear	[76]
Saccharomices cerevisiae Wickerhamomyces anomalus Metschnikowia pulcherrima Aureobasidium pullulans	Naturally fermented olive brine Pomegranate	Botrytis cinerea	Grape	[77]

5. Final Remarks

Consumer demand for minimally processed, additive-free foods and products has led to the development of new packaging/coating materials with active properties. Starch-based edible films and coatings are an environmentally friendly alternative to synthetic polymers due to their low cost, availability, biodegradability and food contact properties. Starch can be used in combination with other polymers or compounds to improve the functional properties of the polymeric matrix, which can also carry active compounds to better control the product shelf-life. Edible coatings can be applied to fruits and vegetables to extend the product shelf-life, decrease water loss, slow down the colour change, pH and titratable acidity during storage and modify the internal atmosphere. Despite the significant benefits gained from using edible coatings for the purposes of extending the product shelf-life and enhancing the quality and microbial safety of fresh or minimally-processed fruits and vegetables, commercial applications are still very limited.

Knowledge of the surface interactions between coating-forming dispersions and the product skin is essential if film adhesion is to be understood and the coating performance optimized in terms of barrier or mechanical properties. Good surface wettability and the proper adhesion of the coating are required to ensure its functionality. Coating adhesion and durability are important for the preservation of food quality during storage. In order to truly receive the benefit of edible coatings on fruits and vegetables in commercial applications, the coating must adhere to the food surface during processing, storage, and transportation.

Given the variability in both the surface characteristics of the different fruits and vegetables, as well as in their physiological behaviour, further studies should be carried out optimizing the coating formulation for specific applications. Starch-based formulations have the advantage of their food contact properties since starch is edible and low cost and has good carrying properties for different actives which can protect fruits and vegetable from microbial decay or physiological disorders. Most of the studies into food applications have been conducted on a laboratory scale; thus, research into cost reduction and large-scale production is required.

Author Contributions: Both authors organised, wrote and revised the review.

Acknowledgments: The authors acknowledge the financial support from the Ministerio de Economía y Competitividad (MINECO) of Spain, through the projects and AGL2016-76699-R and RTA2015-00037-C02. Mayra Sapper thanks the Conselleria de Educación, Investigación, Cultura y Deporte de la Comunitat Valenciana for the Santiago Grisolía grant GRISOLIA/2015/001.

Conflicts of Interest: The authors declare no conflict of interest.

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