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Combined use of physical treatments and edible coatings in fresh produce: moving beyond

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Purpose of the review: In this review we describe the general principles and recent trends in edible coating of fresh produce. We also discuss some features that may link coating applications with common postharvest physical methods (modified atmospheres, heat treatments and irradiation).
Main findings: Coatings of intact and fresh-cut produce must be considered a supplement to proper temperature control. Works aimed at determining the outcome of edible coatings must then compare their benefits under proper postharvest storage conditions (optimal temperatures and high relative humidity with minimal delays). Extensive research has been conducted in the last decade to generate material combinations that would meet the specific needs for coating fruits and vegetables. Substantial improvements in the materials properties (eg, through the evaluation of novel matrix constituents and functional ingredients, as well as by developing multilayer, blended or nanocomposite materials) are still possible. In spite of their relative long history as treatments to reduce deterioration and their joint categorization as environmentally-friendly approaches, coating technologies and postharvest physical methods have evolved quite independently. Combined developments may be useful to increase their positive effects. Rather than simply adding the improvements of different treatments applied sequentially, we argue that active collaboration between researchers working on postharvest physical treatments and edible coatings offers opportunities to develop better and innovative approaches that maximize the benefits on quality maintenance.
Directions for future research: Innovative approaches superseding the additive effects of combining coatings and physical treatments are needed. One area to be explored more deeply is the application of physical treatments during or after coating formation, which may improve the material′s properties, while inducing hormetic responses in the commodities. Coating-induced modified atmospheres need to be revisited and improved. The feasibility of using mild post-coating physical treatments to induce custom changes in the materials and to facilitate the release of active principles also requires further work. Finally, research aimed at solving technical difficulties arising during scaling-up processes may help to facilitate the transfer of coating applications from academia to industry.

Keywords: coatings; biopolymers; physical treatments; postharvest; quality

Postharvest technology in the 21st century: revisiting and refining traditional tools

Postharvest science has greatly improved in recent years. Sequencing of some important fruit and vegetable species, development of powerful high-throughput analytical ('omic′) approaches, and progress in deciphering the epigenetic regulation of plant development are contributing to increase our understanding of the physiology of harvested fruits and vegetables [1-4]. Some relatively new technologies such as the use of the ethylene action inhibitor 1-methylcyclopropene and the refinement of dynamic controlled atmospheres have provided alternative ways to control ripening and senescence [5-8]. In spite of this progress the main challenge on a global scale is still to develop sustainable tools to maintain the safety, nutritional and organoleptic quality of freshly harvested products and to reduce the unacceptably high losses occurring during distribution, storage and retail [9].

Recently there has been increased interest in revisiting the use of physical treatments (heat, irradiation) and natural edible coatings to prevent fruit and vegetable deterioration [10]. The fact that these methods have been envisioned as environmentally-friendly approaches to complement traditional postharvest handling methods [11] has raised the interest of industry and academia.

Postharvest physical treatments and edible coating applications have evolved relatively independently. However, they may have high potential as complementary approaches for postharvest management. Herein, we briefly describe the general features and trends in fresh produce edible coating applications. We also discuss the potential of combining coating technologies with appropriate postharvest physical methods. Active collaboration between researchers working on postharvest physical treatments and edible coatings offer opportunities to maximize their benefits on quality maintenance.

Edible coatings for fruits and vegetables: uses and formulations

Edible coatings consist of a thin layer of protective material added by dipping or spraying that can be consumed as a part of the product [12, 13]. The main interest in edible coatings is generally based on their potential to prevent quality loss and extend the shelf life of the commodities by modifying their surface properties. Coating applications have been used in fresh fruits and vegetables for a long time. Some records indicate that citrus fruits were waxed already in China in the 12th and 13th centuries [14]. Suspensions of oils or waxes in water were among the first fruit-coating formulations likely used to reduce dehydration. Since these early practices the aims of coating applications have expanded. Fruits and vegetables could be coated:

- to improve surface gloss,
- to reduce abrasion,
- to prevent the absorption of undesirable odors,
- to minimize solute leakage,
- to establish a barrier to moisture loss,
- to change CO_2 and O_2 levels inside the commodity, thereby modulating ripening and senescence,
- to retard decay, or

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- to enhance produce stability and safety by including functional ingredients such as antioxidants and antimicrobials.

A variety of materials, including lipids, polysaccharides, and proteins, alone or in combination, have been used in edible coating formulations [12, 15]. Coating matrices generally comprise a major constituent, to which other components such as plasticizers, emulsifiers, reinforcements, additives and solvents (eg, water and alcohols) are added. The large battery of materials used and their possible combinations provide coating systems with a wide range of physical and chemical properties that could fit different applications.

The main matrix components used in coating applications are hydrocolloids, lipids and their mixtures (composites) [16*]. Polysaccharides (cellulose derivatives, alginate, pectin, starch, chitosan, carragenan and gums) and proteins (from vegetal origin like soy protein, wheat gluten, corn zein, and from animal sources such as gelatin, whey, casein and keratin) have been tested [17, 18]. Special interest has been devoted to the re-utilization of industrial byproducts (ie, gelatin, soy protein, whey and chitosan from the meat, oilseed, cheese and seafood industries, respectively) [19**].

Both protein and polysaccharide films and coatings generally exhibit excellent barrier properties against oxygen, and aroma, but have high water vapor permeability [19**]. A number of different lipid sources including waxes (beeswax, candelilla wax, carnauba wax), free fatty acids, fatty alcohols, fatty acid and sucrose esters, edible terpene resins, such as shellac and paraffins, have been used for fruit and vegetable coating [18, 20-22]. They usually increase surface gloss and limit water loss. Finally, composite coatings are produced by combining two or more constituents. They could be emulsions, multilayer coatings or blends [23, 24]. In emulsions and lipid-hydrocolloids composite coatings, the type of lipid, location, volume fraction, polymorphic phase, and drying conditions have significant impact on the final moisture-barrier properties [25]. The size of the dispersed component in an emulsion also has great effect on the coating characteristics. It can range from 0.2-50 µm in macro-emulsions to 10-100 nm in micro-emulsions [26-28]. Multilayer coatings are in general more effective water vapor barriers than emulsion films. However, the fact that they should be generated in various steps by sequential deposition of materials of interest in the commodity surface represents a technological drawback. An alternative promising strategy to improve the properties of edible coatings is biopolymer blending [29, 30].

As in the field of synthetic polymers the new generation of materials in the area of biopolymers is focusing on obtaining bionanocomposites. These materials consist of a biopolymer matrix reinforced with nanoparticles (particles having at least one dimension in the nanometer range 1-100 nm). They are expected to exhibit better mechanical and barrier properties, thermal stability, chemical resistance and surface appearance, even at low loading levels (5-10%), than traditional materials due to the high surface area of the nanoparticles, which allows them to interact strongly with the remaining components [31, 32]. The addition of nanoclays, like montmorillonite, to protein and polysaccharide formulations has enhanced the mechanical and barrier properties of biopolymeric films [33, 34]. Cellulose nanofibers, chitin and starch nanowhiskers have improved the mechanical properties of casein, starch and chitosan films [35-37]. Nanocomposites may also modulate the release of active ingredients incorporated in the polymeric matrix [28-41].

Different ingredients can be incorporated into edible coating matrices to improve or modify their functionality. Plasticizers are commonly added in polysaccharide and protein-based coatings to decrease brittleness [42-44]. Common plasticizers used in edible coatings include water, glycerol, propylene glycol, sorbitol, sucrose, polyethylene glycol, fatty acids, and monoglycerides [42]. Added in different proportions (15-40% relative to the main matrix constituent) plasticizers may enhance film flexibility and susceptibility to humidity and decrease its strength and barrier properties against moisture and oxygen [45]. Hydrophobic plasticizers, such as citrate esters are also being studied [46].

The functionality of these main matrix constituents can be also modified by chemical, physical or enzymatic treatments [47-49]. Protein cross-linking may reduce the material solubility, generate stronger matrices and in some cases increase surface hydrophobicity [49-50]. Traditional chemical cross-linkers may not be suitable for edible materials due to their toxicity. In this case protein modifying-enzymes such transglutaminase, lipoxidase, lysyl-oxidase, polyphenol oxidase and peroxidase may be useful. Phosphorylation of soy proteins by a protein kinase can also be used to modulate the material solubility and emulsifying capacity [51]. Polysaccharide properties can be also modified by changing their degree of polymerization, side-chains, degree of acetylation or methylation [52].

Emulsifiers are surface-active agents of an amphiphilic nature and are able to reduce the surface tension of water–lipid or water– air interface. They can be also added to improve the formulation wetting, spreading and adhesion ability [53**]. Protein-lipid or polysaccharide-lipid composite coatings often require the addition of emulsifiers to facilitate the dispersion of the lipid component in aqueous media. Common emulsifiers include the polyethylene sorbitol esters, some fatty acids and salts, and phospholipids such as lecithin [46, 54, 55].

The functionality of edible coatings can be expanded by adding antioxidants, antimicrobials, colorants, flavors, nutrients, and spices to the formulation. These compounds could be retained on the food surface enhancing food quality, stability, and safety [56-60]. Common antimicrobial agents used in food systems, such as benzoic, propionic and sorbic acid salts, may be incorporated. Starch-based coatings containing potassium sorbate were applied on fresh strawberries to reduce decay [61]. Hydroxypropyl-methyl cellulose coatings containing ethanol inactivated *Salmonella montevideo* on fresh tomatoes [62]. Antimicrobial enzymes such as lysozyme have been incorporated into chitosan coatings to control *Escherichia coli* and *Streptococcus faecalis* [63]. Essential oils have been also widely investigated as natural antimicrobials for coated fruits and vegetables [64]. Interestingly, chitosan shows antimicrobial effects *per se* [65-67].

Early work by Swenson *et al.* [68] showed that coatings containing butylated hydroxyanisole (BHA), butylated hydroxy toluene (BHT) and citric acid reduced rancidity in nuts. Antioxidants such as ascorbic acid have been used to reduce browning in coated mushrooms [69]. Xanthan gum coatings mixed with α-tocopherol enhanced the nutritional quality and improved the surface color of peeled baby carrots [70]. Texture enhancers have been also incorporated into edible coatings. Hernández-Muñoz *et al.* [71] observed that the addition of calcium gluconate to the chitosan (1%) coating formulation increased the firmness of refrigerated strawberries. Flavor and coloring agents may also be added to edible coatings. However, very little has been reported regarding these applications. Finally, some researchers have endeavored to incorporate micronutrients and bioactive compounds [63-72].

Challenges in edible coating technologies

Several works show that the benefits obtained by coating applications may be substantial [73-77]. However, compared to the large number of studies reported in literature relatively few applications have been adopted by the industry. Although various raw materials employed are industrial byproducts, the procurement, transport, distribution and storage costs of these materials as well as the infrastructure for large-scale coating production may be in some cases still too high. Modification of established procedures, changes in process operations and processing line layouts, and the need to implement specific training programs may have in some cases delayed technology transfer. Future studies aimed at helping to recategorize coatings as a "widespread technology" should focus on:

i. selecting the formulation, and coating-formation conditions that optimize the properties required for each commodity;
ii. anticipating potential consumer concerns especially when complex activated formulations are tested. For example nanotechnology offers opportunities to develop novel packaging strategies [78]; but the debate about the potential benefits and risks of human consumption of some nanoparticles is still open and legislation on this issue is in many cases diffuse;
iii. predicting potential difficulties during scaling-up processes may be highly useful. Simple procedures in lab settings such as generating continuous and even coatings, while avoiding physical damage and temperature abuses may be challenging in commercial settings.

The evaluation coatings at unacceptably high temperatures and low relative humilities for fruit and vegetable handling could overestimate their practical benefits. Technologically-oriented studies must evaluate coating applications under optimal storage conditions (temperature and relative humidity) since they should be considered a complement to proper refrigeration.

Intersecting fruit coating technology and postharvest physical methods

Physical treatments before or during coating formation

The application of physical methods to raw materials may modify the properties of the resulting coatings [79, 80]. Heat, high pressure and irradiation treatments have been shown to induce changes in the functionality of the resulting films. The effects are generally more marked for solubility and mechanical properties than for water vapor permeability [81-88].

Mild heat treatments are used in some cases to accelerate coating formation [89, 90]. The nature of the treatment applied may affect the physical-chemical properties of the coating itself [91]. The drying process applied to protein films and coatings may affect the type and proportion of covalent (S–S bonds) or non-covalent (hydrophobic interactions, ionic and hydrogen bonds) interactions and, consequently, material properties [92]. In addition, for polysaccharides the drying temperature may affect film performance [93]. Similarly, the relative humidity and pressure during the drying period has been also shown to affect the properties of soy and amaranth biopolymers [92, 94, 95].

While these physical treatments have been in many cases optimized based only on the material requirements, they may be potentially exploited to induce hormetic responses in the commodities, maximizing the final outcome of the combined treatment on quality maintenance. The presence of a protein, polysaccharide, lipid or composite coating on the commodity surface would affect the heat transfer properties and consequently the optimal treatment conditions will likely differ compared to the scheduled in uncoated products.

Physical treatments after coating formation

This approach includes: i) the modified atmosphere that would be established in coated commodities due to the change in the fruit or vegetable surface permeability to gases (CO_2, O_2, ethylene); or ii) any change in the environment aimed in eliciting a desirable response in the commodity or in the coating material.

Coating-induced modified atmospheres

Once the coating is established the gas exchange between the fruit and the environment is affected, providing the opportunity to generate single fruit modified atmospheres. The equilibrium gas composition to be reached is determined by a number of product factors, coating properties and environmental conditions.

Product factors: The respiratory rate of the commodity is the most important factor affecting the modified atmosphere reached. It will depend on the species, cultivar and on its developmental stage [96]. Ethylene production and accumulation may be important especially in climacteric coated commodities in which it may increase respiration.

Coating properties: All factors affecting the mass transfer through the film and the diffusion process such as permeability to O_2, CO_2 and thickness will determine the atmosphere established. Plasticizers, generally increase film permeability. The polymerization and/or cross-linking degree of the protein or polysaccharide as well as the functional groups/substituents added or removed would also contribute to the coating permeability. In general protein and polysaccharide coatings have high permeability to polar substances, such as water vapor, and low permeability to non-polar substances, such as oxygen. Protein coatings appear to have lower oxygen permeability than cellulose-based coatings. Besides the absolute permeability of the different coatings their selectivity is also a main determinant of the internal atmosphere of fruits [97, 98].

Environmental conditions: Coated fresh fruits and vegetables would be stored mostly at relative humidities between 85 and 95%. Under these conditions some coatings may hydrate, substantially changing their physical and chemical properties [28, 99]. In general, at low and intermediate relative humidity, the permeability to gases of protein and polysaccharide materials is much lower than that of polyethylene, which makes them interesting materials for certain applications. However, the permeability of polysaccharide and protein coatings are highly dependent on the relative humidity [100]. For instance, the permeability to oxygen and carbon dioxide may increase up to a thousand times when these materials are held at high relative humidity. In protein materials, this effect is much greater for the "hydrophilic" gases (CO_2) than for the "hydrophobic" gases (O_2) [101, 102]. It is important to highlight that many of the standard methods of material characterization use relative humilities which are far away (58%) from those in which they will be used in when coating fresh fruits and vegetables. The storage temperature also has a significant impact on the atmosphere reached, as it exponentially affects fruit and vegetable respiration rate. Variations in the cold chain may lead to fermentation in coated commodities [103]. The CO_2 and O_2 partial pressures in the storage atmosphere may also influence the levels of gases at steady state through alterations in the commodity respiration rate.

Post-coating physical treatments

As previously indicated some physical treatments may be used to induce desirable changes in the vegetable physiology [103]. The evaluation of post-coating heat treatments has received little attention so far. Post-coating UV-irradiation may be expected to affect mostly the coating formulation given its low penetration. Some works have shown that the functional properties of biopolymers also can be improved if subjected to some physical treatments after their formation. The tensile strength of soy protein films was improved when subjected to UV or γ radiation. This is probably due to the high content of tyrosine and phenylalanine, since these amino acids can cross-link when irradiated [104, 105]. Changes in the postharvest environment may also be potentially exploited to modulate the functional properties of activated coatings [106]. When incorporated into tomato fruit packages, soy protein pads absorbed water, facilitating the release of cyclodextrin -1-methylcyclopropene formulations which delayed ripening [107]. Commodity compatible heat treatments during storage may also be considered as a strategy to modify the diffusion rate of additives present in the coating matrix. In some cases they may also be envisioned as a strategy to increase the rate of temperature dependent reactions between coating ingredients (eg, enzymatic reactions). The release of active compounds added to the coating by photochemical means (eg, UV-vis radiation) may be another strategy to explore.

Concluding remarks

Edible coatings and postharvest physical methods have a relatively long history as treatments to reduce deterioration of stored fruits and vegetables and have been envisioned as environmentally-friendly approaches to supplement refrigeration. Although both strategies have evolved quite independently, combined developments may maximize the beneficial effects on quality maintenance of fresh produce. Coatings should be considered as a supplementary treatment to proper temperature control in intact and fresh-cut produce. Consequently, studies must compare their benefits against controls subjected to proper postharvest management and storage (refrigeration and high relative humidity with minimal delays). This will unequivocally determine their benefits relative to the recommended handling practices. Continuing the currently active work oriented to maximize coating functionality will be useful. Active collaboration between postharvest technologists and material scientists may reduce the boundaries between these two fields. Innovative approaches superseding the additive effects of combined coating-physical treatments application will be necessary. Some areas that may be explored include the application of physical treatments before or during coating formation which may improve the material properties while inducing hormetic responses in the commodities. Optimization of the single-fruit modified atmospheres generated after coating establishment is still needed. The feasibility of using mild post-coating physical treatments to make custom changes in the material properties and/or to facilitate the release active principles still needs to be determined. Finally, research aimed at solving technical difficulties arising during scaling-up processes may help to boost the transfer rate of coating applications from academia to industry.

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