

THE USE OF BIOACTIVE COMPOUNDS IN SINGLE COMPOSITE AND BLENDED EDIBLE FILMS AND COATINGS: PROGRESS AND FUTURE PERSPECTIVES FOR FOOD PRESERVATION

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Abstract

Recent years have witnessed an increase in the use of plastic alternatives for food and non-food-related purposes. In line with this, several studies have sought to examine the impact of using naturally occurring biopolymers such as lipids, proteins or carbohydrates to enhance the mechanical and functional properties of edible films and coatings. However, there is limited comprehensive evidence on the influence of bioactives (used either singularly or in combination) on the quality and functional characteristics of these edible packaging. This review therefore sought to explore the effect of bioactives on edible coats and films and the synergistic potential when multiple bioactives are combined in improving the physicochemical and functional properties of not only these edible coats and films but also in the preservation of food commodities. It was identified that the effect of bioactives on the mechanical, antimicrobial and antioxidant properties of edible films and coatings is not definite and depends to a large extent on the characteristics of both the bioactive and biopolymer used. Also, the use of multiple bioactives in edible packaging has either a synergistic or antagonistic outcome which is influenced by both the bioactive and biopolymer used in the construction of the packaging. Understanding the unique alterations that occur in the properties of a packaging material due to the nature of bioactives incorporated is beneficial in both the construction and use of these edible packaging.

Keywords

antioxidant, antimicrobial, bioactive, coatings, edible, films, packaging, synergistic

Introduction

The undesirable impact of synthetically derived packaging on the environment, the rising trend to use sustainable materials in food applications coupled with an increasing desire of consumers for healthier food has culminated in a drastic increase in the use of edible coatings (EC) and edible films (EF) in food applications to prolong the keeping quality of produce thus aiming to reduce current global post-harvest losses which range from 30 to 60% (Andriani and Handayani, 2023; Chavan et al., 2023).

Edible food packaging such as coatings or films refers to a form of food packaging that can be eaten without any adverse effects and protects a food commodity from mechanical damage and spoilage (Ganiari et al., 2017). The key difference between a film and a coating is that the former is pre-formed and then applied to the surface of the commodity in question or layers (parts) of the commodity whilst the latter is directly applied to the surface of the produce (Chavan et al., 2023; Sahraee et al., 2019). Edible coatings and films are usually developed from biopolymers obtained from either protein, carbohydrate, or lipid sources (Andriani and Handayani, 2023; Chavan et al., 2023; Dubey and Dubey, 2020; Han, 2013; Kupervaser et al., 2023). The characteristic properties of the film or coat depend to a large extent on the properties of the biopolymer used in its construction.

In recent years, there has been a surge in improving traditional packaging materials by developing active packaging composed of biopolymers with improved functionality (Feng et al., 2018). This has resulted in a steady rise in the use of blended (composite) coatings and films. These coatings and films have improved functionality due to the presence

of two or more biopolymers whereby one biopolymer makes up for the deficiencies of the other biopolymer component of the packaging. Such films are sturdier than their single-component alternatives in terms of both mechanical properties and preservation potency (Dubey and Dubey, 2020; Kupervaser et al., 2023).

Since spoilage in packaged and non-packaged food is usually premeditated by spoilage organisms, protein denaturation and/or lipid oxidation (Bayram et al., 2021; Ganiari et al., 2017; Xavier et al., 2021), factors such as weight loss, colour degradation, polyphenol oxidase content, rate of microbial growth, and modulation of gaseous exchange are used in measuring the efficacy of edible coatings or films (Andriani and Handayani, 2023). Thus, the incorporation of bioactives into a film or coat usually seeks to improve the functionality of the active packaging by regulating these factors.

Several studies have reported on improving either the biopolymer or bioactive composition of edible coatings and films to increase the application of these edible coatings and films in food preservation (Chavan et al., 2023; Chavez-Marquez et al., 2023; Chen et al., 2021; Chettri et al., 2023; Hu et al., 2020; Krasniewska et al., 2014; Rosenbloom et al., 2020; Wardana et al., 2023; Wigati et al., 2023). However, to our knowledge, there is no existing comprehensive report on the effects of incorporated bioactives on the mechanical properties and preservation capacity of coatings and films. As such, this article seeks to review existing literature as it relates to bioactives incorporated into edible coating and films and how the incorporation of these bioactives singularly or synergistically influences the efficiency of these coatings and films in food preservation.

Table 1. Overview of some bioactives used in edible coatings and films

Bioactive Compound	Bioactive concentration (%)	Biopolymer composition of film/coat	Food Matrix	Storage Duration (Days)*	Storage Temperature (°C)	Reference
Organic Compounds						
Hexyl acetate	0.03	Alginate	Rose apple	10	4	Duong et al. (2023)
Essential Oils						
Lemongrass	7	Chitosan, Gelatin	Tomato	20	8	Erceg et al. (2023)
<i>Ocimum gratissimum</i> L. (African Basil)	0.3	Konjac glucomannan	Walnuts	12	40	Wang et al. (2023)
Plant Extracts						
Summer savory (<i>Satureja hortensis</i> L.) extract	20	Pullulan	Sweet Pepper	21	6	Krasniewska et al. (2014)
Seabuckthorn leaf extract	1.5	Chitosan	Apple Peach	21 25	2, 16 4	Rather et al. (2024)
Nanoparticles						
Silver nanoparticles (AgNPs)	20	Sodium Alginate, Gelatin	Tangerine	15	20	Li et al. (2022)
Zinc oxide nanoparticles (ZnONPs)	2, 4	Agar	Green Grapes	14, 21	–	Kumar et al. (2019)
Netamycin	0.66	Agar, Gelatin	Strawberries	6, 2	4, 20	Wang et al. (2022)

*Note: For some entries, the number of days indicates the period until significant spoilage was observed; for others, it represents the total storage period.

Incorporation of bioactives into edible coatings and films

Bioactives are organic compounds produced as secondary metabolites by animals, plants or microorganisms which possess unique properties beneficial to humans. Common ones suggested to improve the quality of ECs and EFs include phenolic compounds, plant and fruit extracts, fatty acids, minerals and vitamins (Andriani and Handayani, 2023; Kaur et al., 2023; Pai et al., 2022; Sahraee et al., 2019). The extraction yield of any bioactive from a food matrix depends to a large extent on these questions: is it a wet or dry matrix? (dry matrices give higher yields than wet); what drying treatment was the food sample subjected to before extraction? (air drying procedures give higher yield than oven drying or indirect sunlight/solar drying); and what is the particle size of the food matrix? (nano sizes give greater yield than micro sizes) (Wong et al., 2023).

The effectiveness of an edible coating or film is usually measured by its degree of transparency, opacity and/or gloss, swelling capacity, water solubility, gas and water vapour permeability, tensile strength, flexibility and thermal stability (Dubey and Dubey, 2020; Kupervaser et al., 2023). Economic viability and additional functionalities such as antioxidant and antimicrobial properties also increase the efficacy of these films and coatings (Chavan et al., 2023; Kupervaser et al., 2023). A suitable bioactive should therefore be one which when incorporated into an edible coating or film does not negatively reduce the primary desirable properties of the coating or film such as significantly altering its mechanical and physical properties (tensile strength, elongation at break etc) and sensory attributes such as taste, texture and colour of

the product after being coated (Falguera et al., 2011; Ganiari et al., 2017). Table 1 summarizes studies on the use of various bioactive compounds in edible packaging for preserving agricultural produce. The incorporation of these bioactives has attracted significant attention because they enhance shelf life, safety, and quality while meeting consumer demand for natural preservatives. One notable aspect concerning the use of bioactives as can be identified in the table is the wide range of bioactive compounds which can be used. This includes essential oils, plant extracts, and nanoparticles. These compounds serve multiple functions, such as antimicrobial, antioxidant, and antifungal activity, thereby reducing microbial spoilage and oxidative degradation. Another key thing to note is that the biopolymer composition plays a crucial role in the functional properties of an edible packaging. The table highlights various polymer matrices, including polysaccharides (e.g., chitosan, alginate, pectin) and proteins (e.g., gelatin). The choice of polymer directly influences film permeability, mechanical strength, and the potency of an incorporated bioactive. The various food matrices covered in the table demonstrate the applicability of bioactive enriched edible packaging across a broad spectrum of perishable products. This is not limited to only fruits and vegetables as has been presented in the table but can include other perishable products such as dairy, meat and meat products and fish and fish products.

It should be noted however that the ability to preserve the functionality of a film and the potency of an incorporated bioactive is by both the type of bioactive used and also the concentration at which it is incorporated. This is because too low or too high concentrations may yield undesirable results, as excessively high concentrations can interfere with cross-

links in the polymer chain reducing its efficiency while too low concentrations may not be potent enough to achieve the desired antioxidant and antimicrobial effects (Nguyen et al., 2020). Other factors which may influence the functionality of a film and the potency of an incorporated bioactive are the nature of extraction of the bioactive and the nature of the food product to which the film or coating is applied (Ganiari et al., 2017; Han, 2013).

Effect on mechanical properties

The influence of bioactives on some of the key mechanical and rheological properties of edible films and coatings such as tensile strength, elongation at break, and thickness varies and depends to a large extent on both the chemical composition of the bioactive incorporated and the properties of the biopolymer or biopolymer blend used. In some cases, an increase in the concentration of incorporated bioactives can result in thicker (sometimes more ductile) films with reduced tensile strength and reduced elongation at break properties. This was identified by Khedri et al. (2021) when casein polypeptides were incorporated in increasing proportions in gelatin films. Cavdarolu et al. (2020) also noticed an increase in thickness and decrease in tensile strength as the concentration of eugenol incorporated in a pectin biopolymer increased from 1% to 3%. This decrease in tensile strength could be attributed to bonds and cross-links interfering with the structured matrix of the film. However, in the case of Cavdarolu et al. (2020), the decrease in mechanical properties seems to be influenced by not only the concentration of bioactive used but also the source of biopolymer used because whereas films formed from pectin extracted from sun-dried figs exhibited a decrease in mechanical properties, films formed from pectin which had been extracted from citrus did not show a decrease in mechanical properties.

In other instances, an increase in bioactive concentration did not only result in thicker films but was accompanied by an increase in elongation at break and a decrease in tensile strength thus giving thicker more pliable (softer) films (Kumar et al., 2019; Wang et al., 2022, 2023), or an increase in tensile strength and decrease in elongation at break as was identified by Guo et al. (2024) at 1% incorporation of antagonistic yeast in a trehalose-gelatin composite blend. In other cases, the incorporated bioactive can result in an increase in both tensile strength and elongation at break values (Erceg et al., 2023). It is also not unusual for there to be neither increase in tensile strength or elongation at break values when the bioactive is added at concentrations which are outside the optimal range of the bioactive (Li et al., 2022).

Effect on antimicrobial capacity

Although biopolymers such as chitosan possess antimicrobial properties (Salgado et al., 2015), effective inhibition of microbial growth cannot usually be solely achieved by the structural characteristics of the biopolymer. Thus, the incorporation of a bioactive in the form of a biocide agent is vital to the antimicrobial efficiency of a film/coating (Erceg et al., 2023).

Bacteriocins produced by lactic acid bacteria especially those of the lactobacillus and bacillus genus, lime essential oil, proteins such as chickpea albumin and phytochemicals such as cinnamaldehyde and carvacrol when incorporated into edible films and coatings have been shown to increase the antimicrobial capacity of both single component and blended films and coatings which in turn extends the shelf life of coated agricultural produce (Dubey and Dubey, 2020; Erceg et al., 2023; Sahraee et al., 2019; Wajahat, 2023).

The amount and type of bioactive present also greatly influences microbial activity. Studies such as Cavdarolu et al. (2020) recorded the highest zones of inhibition (effective antimicrobial activity) against *Listeria innocua* at bioactive concentrations of 2% while Duong et al. (2023) identified promising results when hexyl acetate was used as a bioactive at a concentration of 0.03% and not higher in alginate cross-linked with calcium chloride film. Sensitivity of gram-positive and gram-negative bacteria to bioactives incorporated into these edible films and coatings vary and are not always clear cut. Some studies have shown that films incorporated with bioactives produce larger inhibition zones against gram-positive bacteria than gram-negative bacteria. This difference is attributed to the presence of a double cell membrane in gram-negative bacteria, as opposed to the single cell membrane in gram-positive bacteria (Feng et al., 2018; Khedri et al., 2021; Krasniewska et al., 2014; Seydim et al., 2020). However, other studies such as Li et al. (2022), have reported an opposite effect, where films incorporated with silver nanoparticles inhibited the growth of *E. coli* (a gram-negative bacterium) more than *S. aureus* (a gram-positive bacterium). The increased sensitivity of gram-negative bacteria to the nanoparticles was attributed to the thicker cell wall of gram-positive bacteria which inhibited the migration of these nanoparticles into their internal matrix.

Effect on antioxidant capacity

Over the years, synthetic antioxidants such as butylated hydroxyanisole and butylated hydroxytoluene which inhibit oxidation in food commodities caused by UV light have been in high demand for food preservation because of their low cost and high stability. Although edible film biopolymers such as chitosan have shown great antioxidant potential (Sahraee et al., 2019), the incorporation of a bioactive or blend of bioactives can heighten these properties.

Production of active packaging that has been infused with bioactive compounds from plant sources such as vegetables, herbs, and spices is on the rise as consumers have become increasingly aware of the detrimental health effects of synthetic preservatives. These plant sources possess high concentrations of phenolic compounds which in turn enables them to function as antioxidants (Cui et al., 2020; Ganiari et al., 2017; Sahraee et al., 2019).

Naturally sourced bioactive compounds such as hydroxytyrosol (HT) and 3,4-dihydroxyphenylglycol (DHPG) have successfully been used to improve the preservation of refrigerated

raw beef (Dubey and Dubey, 2020) with some of these bioactives such as polyphenols even conferring a greater antioxidant effect than synthetically derived ones (Sahraee et al., 2019). An interesting discovery in the use of bioactives in films and coatings is that blended films/coatings have been shown to facilitate faster release of antioxidant bioactives than single component ones (Ganiari et al., 2017).

Also, unlike the non-linear relationship between bioactives and the desired mechanical properties of a film/coating, a linear relationship between bioactive concentration and antioxidant capacity can exist whereby an increase in bioactive concentration is accompanied by a directly proportional (equal) increase in the desired antioxidant properties of the film or coating. Feng et al. (2018) identified a directly proportional relationship between tea polyphenols in a starch-based film and its antioxidant capacity. Likewise for casein polypeptide in gelatin-based films (Khedri et al., 2021) and for *Ocimum gratissimum* L. essential oil encapsulated in zein-gallic acid nanoparticles in a konjac glucomannan film (Wang et al., 2023) a proportionate relationship has been identified. Another interesting discovery is that encapsulation of bioactives does not negatively affect their antioxidant capacity when incorporated in a film or coating. This was exhibited by Xavier et al. (2021) when encapsulation of *Cinnamodendron dinisii* essential oil in zein and then incorporated into a chitosan matrix still exhibited a high amount of antioxidant activity when measured by both 2,2-Diphenyl-1-picrylhydrazyl (DPPH) and ferric reducing antioxidant power (FRAP) assays.

Synergistic effect of bioactives

Although a synergistic effect is desired when two or more bioactives are combined in an edible packaging, this is not always achieved either when the coat or film is formed or when it is applied in the preservation of a food commodity. This is because the unique properties of some bioactives make them work better single-handedly than when combined with others. The synergistic or antagonistic effect of combining bioactives can influence beneficial parameters of edible packaging such as its antimicrobial, antioxidant, tensile strength, pH and moisture regulation capacities when used in food preservation.

The pH of a film or coat greatly influences the extent to which that film or coat can promote or inhibit microbial growth thus influencing the preservative qualities of the film or coat. Given this, a bioactive when incorporated into a coat or film should either lower the pH of the coat and thereby inhibit microbial growth or not cause a significant increase in pH when the coat is used in produce preservation. Combining certain bioactives in a coating can create a synergistic effect, with coatings containing multiple bioactives exhibiting lower pH values than those with only one bioactive (Sharifimehr et al., 2019; Zolfaghari et al., 2023).

Depending on the type of bioactive used, however, the synergistic effect may not be as pronounced (Das et al., 2024) or may be evident in the coat or film when formed but not existent when the coat/film is used in the preservation of a

food commodity. Sharifimehr et al. (2019) identified a synergistic effect when bioactives (aloe vera and eugenol) were combined in an edible coat and this effect increased as the aloe vera concentration increased. However, the effect was antagonistic (detrimental) in nature when the coat was used in the preservation of shrimps; with coats containing multiple bioactives exhibiting a higher increase in pH over the storage period than coats containing only one bioactive. Another scenario is when the combined bioactives may not exhibit a synergistic effect in the coat or film but when it is applied in the preservation of a food commodity; a synergistic effect manifests. Nguyen et al. (2024) identified that although coats containing either *Piper betle* L. leaf extract or *Sonneratia ovata* Backer leaf extract had similar pH values to coats containing both bioactives; when the coats were applied in the preservation of banana fruits, fruits whose coat contained both leaf extracts had a lower pH by day 10 of storage as compared to fruit whose coat contained only one leaf extract.

One of the major causes of food spoilage that results in food loss or waste is microorganisms (Saini et al., 2021). Hence, increasing the antimicrobial capacity of films and coatings by combining bioactives would be an advantage. Combinations of bioactives such as dill leaf extract and dill essential oil (Zolfaghari et al., 2023) or aloe vera and eugenol (Das et al., 2024) have exhibited synergistic antimicrobial potential when used in food preservation. This synergistic effect can limit the proliferation of psychrotrophic bacteria, lactic acid bacteria, and Enterobacteriaceae, especially *Escherichia coli*, *Staphylococcus aureus*, and *Bacillus subtilis*. Combining bioactives in edible packaging can also increase the antioxidant capacities of that packaging. An increase in antioxidant capacity implies that such edible films or coats will have a greater preservation power than coats or films with only one bioactive. The overall sensorial acceptability of preserved food products also tends to increase when the preservative coat contains more than one bioactive (Das et al., 2024; Majumder and Ganguly, 2020; Sharifimehr et al., 2019; Zolfaghari et al., 2023).

Senescence in agricultural produce is manifested primarily by loss of weight (seen either as wrinkling, shrivelling or shrinking), increase in Total Soluble Solids content (TSS), or decrease in Titratable Acidity (TA) content. The combination of bioactives in edible coats and films can modulate weight loss even in succulent fruits thereby ensuring minimal weight loss at the end of a set storage period (Das et al., 2024; Majumder and Ganguly, 2020; Nguyen et al., 2024). However, the synergistic potential of multiple bioactives when combined to prevent an increase in TSS associated with a corresponding decrease in TA cannot be conclusively drawn. This is because some studies have reported no difference in TSS and TA values when bioactives are used singularly or in combination (Majumder and Ganguly, 2020) whilst other studies reported greater signs of senescence as evidenced by an increase in TSS and decrease in TA when bioactives are used in combination than when they are used singularly (Das et al., 2024) and some yet still showing minimal increase in

TSS when bioactives were used in combination as compared to when they were used alone (Nguyen et al., 2024). It should not be ignored that variations in the inherent physiological characteristics of agricultural produce may influence the accuracy of the results obtained to some degree. This is because the intrusive/invasive nature of most quality assessment methods makes it impossible to keep a product (especially fruits and vegetables) in storage after testing thus necessitating the need to pick a new sample whenever a quality parameter is to be assessed. This can bring variations and inaccurate measurement of content primarily because initial content was not accurately identified.

Future trends

Edible coatings and films are projected to be one of the key drivers of fruit quality in the future (Chavan et al., 2023) thus ensuring the optimal functioning of these edible packaging is an inevitable step. Over the years, tremendous improvements have been made in the incorporation of bioactives in coats and films and one recent advancement is the incorporation of Lactic Acid Bacteria (LAB) possessing probiotic and/or bacteriocin (antimicrobial) properties into edible films and coats. As with other bioactives, interactions that can occur between the probiotic and the film or coat can change either or both the chemical and physical properties of the edible packaging and thus render these packaging as either very efficient or inefficient in the preservation of a food commodity. The sensitive nature of these probiotics also requires that additional attention be paid to the composition of the coat/film, how it is produced, and the temperature and other storage conditions at which the coated food product is kept to prevent the probiotics from being killed or damaged. The use of blended films or coats or the inclusion of prebiotics such as oligofructose and polydextrose also tends to increase the viability of these probiotics in the formed edible packaging (Silva et al., 2023). The use of nanoengineering for the incorporation of bioactives into edible films and coats is also on the rise. Nanostructures such as nanofibers, nanoliposomes, or nanoparticles are used in the construction of the film or coating and to encapsulate bioactives such as antimicrobial or antifungal agents. Encapsulation of these bioactives in these nanostructures not only improves the stability of the film or coating but can also modulate the release of the bioactives out of the film/coating matrix onto the food product or the headspace region of the packaging which does not only reduce microbial contamination but also prolongs the freshness of the packaged food product (Juric et al., 2024; Koirala et al., 2023).

One interesting development in the incorporation of bioactives into these edible packaging is the nature of bioactives being added. Phytochemicals such as phenolic compounds when added to films and coatings do not only influence the strength of the packaging but also its functionality thus ensuring that the quality characteristics of the packaged food are preserved. The use of phenolic compounds such as lignans, stilbenes, phenolic acids, and tannins which possess anti-

microbial, antioxidant, antifungal, and UV-blocking properties or others such as lignin which can positively alter the structural properties of the film or coat is on the rise (Singh et al., 2022).

Conclusion

The incorporation of multiple bioactive compounds into edible coatings and films holds great promise for food preservation but also presents challenges. While combining bioactives is often expected to enhance mechanical strength, antimicrobial efficacy, antioxidant capacity, gas modulation, and physicochemical properties, the results are not always consistent. Certain attributes, such as sensory quality, antioxidant activity, and water vapor regulation, tend to improve with bioactive combinations. However, parameters like total titratable acidity and total soluble solids content may not necessarily improve when bioactives are combined. This variability in the effectiveness of bioactive compounds stems from the distinct characteristics of each bioactive compound, the mode of extraction used, the concentration at which it is incorporated, the type of biopolymer used in the creation of the edible packaging and the nature of the food product which is to be preserved. The intricate nature of these interactions underscores the importance of a targeted approach in both the extraction and formulation of bioactive-enriched edible packaging. Addressing these challenges is crucial to developing more reliable and effective edible coatings and films that enhance food preservation, extend shelf life, and support the growing demand for natural preservation methods.

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