

COATINGS/FILMS FOR FOOD PACKAGING: GREAT POTENTIALS OF NATURAL POLYMER NANOCOMPOSITES IN VIET NAM

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Abstract

Recently, significant efforts have been devoted to evaluate various natural polymers as alternatives to synthetic plastic packaging. Natural polymer nanocomposites consist of natural biopolymers (e.g., chitosan, starch, cellulose, alginate and many more) derived from plants, which are abundantly available in nature, biodegradable and thus eco-friendly, incorporated with nanomaterials can prevent the growth of spoilage and pathogenic microorganisms, improve food quality and safety, and extend shelf-life of food. Therefore, the objective of this work is to review the most recent developments in the fabrication of natural polymer nanocomposite films/coatings as well as their applications in various food products and the effects they had on them. These perspectives could provide information for the future research should be carried out to widen the applications of bioactive films/coatings to more foods and industries, as well as their industrial scale-up, thus helping to minimize the use of plastic materials.

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Introduction

The synthetic plastics used in food packaging are unfriendly environmental, most end up in landfills or oceans where pollution can significantly damage the local ecosystem. Therefore, the food industry is searching for the possibility of using biodegradable materials derived from natural sources such as cellulose, starch, chitosan, fibrins, alginate, hyaluronic acid etc. [1]. These biopolymers, with good film-forming properties, nontoxicity, biodegradability, antibacterial, antifungal, have been studied a lot about the applicability in preserving fruits and vegetables postharvest [2-9]. However, the low antioxidant and antibacterial potentials of natural polymer films, which are key criteria for an active food packaging film, limit their potential, thus, additional molecules such as bio-active reagents have been incorporated into them to generate novel substances with appropriate applicability [1015].

In the recent years, the potential of nanotechnology has been utilized in almost every segment of the food industry for example food processing (e.g. encapsulation), food packaging (e.g. antibacterial container, polymer films) [16, 17]. The basis of this technology is nanomaterials that are in the range of 1100 nm. Nanomaterials exhibit size-related physiochemical properties that make significantly differ from those observed in fine particles or bulk materials [18]. These structures possess large surface areas that facilitate the absorption of bioactive agents. Additionally, their tiny nanoscale dimensions enhance the effective penetration through cell membranes. The polymer nanocomposite is considered to multiphase hybrid solid material that contains one of the phases as nanoscale fillers that have at least one dimension in less than 100 nm distributed within a polymer matrix [19].

The present review will include the current literature on the fabrication methods of natural polymer nanocomposite films/coatings, on the unique properties of these materials and their applications in food packaging and preservation for shelf life extension of fruits, vegetables, meats, poultry, fish, and seafood products.

Properties of natural polymer nanocomposites

Nanomaterials or nanocomposites, owing to their enhanced surface reactivity, high surface-to-volume ratio, and physicochemical and antimicrobial properties, impede the activity of microorganisms more efficiently than their micro- or macro-scale counterparts. The efficacy and performance of active food packaging have been enhanced through nanoencapsulation or the incorporation of natural antimicrobial-loaded nanocarriers [20].

Antimicrobial activity

Different species of Gram-negative and Gram-positive bacteria, including *Salmonella* spp., *Staphylococcus aureus*, *Staphylococcus epidermis*, *Escherichia coli*, *Listeria monocytogenes*, *Pseudomonas aeruginosa*, *Enterococcus faecalis*, *Vibrio cholera*, and *Bacillus cereus*, are responsible for food spoilage. The natural polymer nanocomposites materials have potent antibacterial effects through various mechanisms of action that interact precisely with microbial cells, including the disruption of cell walls, interruption of transmembrane electron transfer, oxidation of cell components, formation of reactive oxygen species (ROS), disruption of enzyme activity, destruction of internal cell organelles, prevention of DNA synthesis, and cellular death [21].

Barrier properties

Therefore, the barrier properties of polymers are significantly correlated to their intrinsic ability to permit the exchange of low-molecular-weight substances. The structures of nanocomposites and the type and size of nanofillers can affect the degree of modification of the barrier properties of nanocomposites [22]. Well-dispersed nanofillers in the polymer matrix can influence the diffusivity and solubility of the penetrating molecules, particularly in interfacial domains, by increasing the diffusion length and the tortuous path of penetrating molecules to form an impermeable structure in the polymer matrix because of their high aspect ratio [23]. The barrier properties are also affected by the shape, polarity, and crystallinity of the diffusing molecule, the degree of crosslinking, and polymer chains [24]. Improved gas barrier properties and superior permeability have been exhibited by latex membranes and platelet-shaped fillers, respectively, compared to those of the neat membranes [25].

Mechanical properties

Certain nanofillers are dispersed or reinforced in polymer matrices to improve mechanical properties such as strain at break, tenacity, and maximum stress via a reinforcement mechanism for use in food packaging systems [26]. This reinforcement primarily depends on the size, shape, concentration, orientation, surface area, dispersion state, and polydispersity of the nanofillers, possibly resulting in their grafting to the matrix polymers [27]. Particle/polymer matrix interface plays the key role in determining the performance of advanced composite materials such as mechanical properties and dimensional stability. Interfacial adhesion occurs when two different materials such as particle and polymer matrix are blended or combined to create a better dispersion of materials into the matrices [28].

Methods of natural polymer nanocomposite preparation

Basically, nanocomposite systems are made of polymer matrix and incorporated nanosized components. Among nanofillers are those in the form of particles, crystals, rods, whiskers, fibers, tubes or nanogels, and nanoemulsions, which are encapsulated into or deposited on the surface of the polymeric matrix [29].

Polymer nanocomposites can be formulated by various approaches, including solution casting, layer-by-layer assembly, coating/spraying methods, in-situ polymerization.

Solution casting

The solution casting method is one of the most widely used methods for preparing polymer films and coatings because of its convenience. In this technique, the polymer is dissolved in a specific solvent by continuous stirring, and the nanofillers are dispersed into the polymer solution to form a homogeneous mixture [21, 30]. It is a relatively economical and simple method, during which intermolecular electrostatic and hydrogen bond formation results in the polymer structure [31]. However, such intermolecular entanglement also makes the film brittle. Several plasticizers such as polyols like glycerol, sorbitol, and polyethylene glycol, sugars (e.g. glucose and sucrose), and lipids are added to improve mechanical properties of the films [32, 33]. Although, solution casting is a simple and relatively low-cost method, scaling-up of the film-forming process by this technique may pose challenge(s), and needs further development.

Layer-by-layer assembly (LBL)

It is a method used for fabrication of multi-component films that does not need any sophisticated instrument. In LBL assembly, the surface modification mainly depends on the deposition and mutual attraction of alternating polyelectrolytes with opposite net charges onto solid support [34-36]. The polyelectrolytes can be biopolymers such as proteins, polysaccharides, etc., that hold net charge. Deposition can be achieved either by submersion of the substrate into alternate polyelectrolyte solutions or by spraying of solutions onto the substrate. LBL deposition can be used to prepare active packaging films and coatings by the incorporation of active agents either between layers or within the structure of an individual polyelectrolyte [37-39].

Coating/spraying

Coating is often applied on the surface of fresh foods such as fruits and vegetables, fish, meat, etc. for enhancing their shelf-life [69]. Coatings can be performed by spreading, spraying, dipping, or immersing of food materials into polymer based composite solutions [40].

Dipping techniques involve submerging the food in an already-prepared film forming solution, which may be incorporated with natural preservative (s) and plasticizer(s) to improve effectiveness of the applied coating/film [41].

Spray-coating involves similar steps like spread coating except spraying is achieved using compressed air-assisted sprayer [42]. Spraying natural polymer-based film forming solution involves application of the treatment solution on food surface using aerosol sprayer [43].

In situ polymerization

Typically, nanomaterials and monomers or multiple monomers are mixed in a suitable solvent, followed by polymerization with an appropriate reagent to yield polymer nanocomposites. This method enables the fabrication of well-defined multidimensional structures with distinct properties from the initial precursors. Homogenous dispersion in a polymer matrix can be achieved by this technique, which also assists in controlling the size, shape, and morphology of the nanomaterials [44].

Natural polymer-based nanocomposite films/coatings utilization as food packaging

Natural polymer-based packaging materials are more environmentally friendly compared to traditional plastic packaging. Besides, bio-materials provide protection between food and the surrounded environment, thus avoiding the deterioration of food quality, such as decontamination with microorganisms, changes in gas conditions, and the relative humidity of the environment and especial they can be degradable using living microorganisms [45].

Fruits and vegetables

Agricultural produce is perishable in nature and in particular most of the horticultural commodities have a very short postharvest life. Fruits and vegetables exhibit continuous respiration and transpiration even after harvesting, resulting in short shelf life and loss of quality. Therefore, various researchers have searched for natural non-toxic substances that can slow down the process of maturation or reduce the decay of fruits and vegetables after harvest [46, 47]. In a study, Snchez-Gonzlez et al. developed hydroxypropylmethylcellulose (HPMC) and chitosan-based edible coating activated with bergamot essential oil (EO) for cold-stored grapes [48]. They suggested chitosan and HPMC coating containing bergamot EO as the most effective formulation in comparison to chitosan alone with bergamot EO, in displaying the most potent inhibitory action and maximum control over respiration rates with minimum weight losses during postharvest cold-storage. Similar results were obtained in studies based on apples and melons by Salvia-Trujillo, Rojas-Gra, Soliva-Fortuny and Martn-Belloso [49] and Raybaudi-Massilia, Mosqueda-Melgar and Martn-Belloso [50], respectively. Furthermore, a candelilla wax-based edible film containing ellagic acid (phenolic compound) as the active agent was prepared by Saucedo-Pompa et al. (2009) to improve the microbiological quality and shelf life of avocados. The 6-week long storage study of the developed active films provided satisfactory results in terms of the product quality, indicating the possible application

of these edible films for preserving the quality and extending the shelf life of avocados. Application of chitosan and alginate-based coatings incorporated with pomegranate peel extract in capsicum exhibited significant antifungal activity against different strains of *Colletotrichum gloeosporioides* with a shelf life extension of 25 days while these coatings in guava extended fruit life up to 20 days [51]. Coating fruits such as raspberries and strawberries with alginate films containing green tea extracts and oleic acid can significantly reduce the food-borne illnesses being considerably effective in antiviral action against human enteric viruses such as hepatitis A virus (HAV) and human noroviruses (NOVs) [52].

Meat and poultry products

The degradation process of meat and meat products can occur basically for three reasons: (i) lipid oxidation and (ii) spoilage caused by microorganisms and (iii) enzymatic action. Amongst various choices standing in the queue, the use of ecological biopolymer packaging materials that ensure good quality and safety maintenance in meats products are widely employed [53]. Antimicrobial activity of whey protein edible films with oregano EO as the active agent has been established by Zinoviadou, Koutsoumanis and Biliaderis [54] in refrigerated beef samples, whereas Emirolu, Yemi, Cokun and Candoan [55] reported reduced microbial counts in fresh ground beef patties at refrigeration temperature when coated with soy-based edible films embedded with oregano and thyme EOs. In a study by Juck, Neetoo and Chen (2010), different active packaging coatings developed using various biopolymers such as starch, alginate, xanthan gum, pectin, and -carrageenan, and activated with various bioactive compounds as nisin, sodium lactate, Novagard CB1, potassium sorbate, sodium diacetate, etc., were applied on turkey products to prevent the contamination caused by *Listeria monocytogenes* [56]. Zhang et al. [57] developed coatings based on chitosan and gelatin incorporated with free or nanoencapsulated tarragon essential oil to extend the shelf life of pork slices during 16 days of refrigerated storage. The authors were able to extend the pork slice shelf life by 8 and 12 days with the coatings formulated with free and nanoencapsulated tarragon essential oil, respectively.

Fish and seafood

Fish and seafood products are highly perishable foods due to their high water content, which provides good conditions for microbial and biochemical spoilage reducing their shelf life. Therefore, employing active antimicrobial packaging techniques such as biopolymer-based antimicrobial films for extending the shelf-life of fish products through preservation and protection against surface microbial contamination is a viable option [58]. Hake and sole fillets wrapped in edible gelatin films supplemented with chitosan, pepper or clove EOs displayed reduced microbial growth owing to the strong inhibitory action of the films against *L. monocytogenes*, *Aeromonas hydrophila*, and *Staphylococcus aureus* [59]; whereas cellulose films containing potassium sorbate, sodium

benzoate, and nisin reported a significant decline in the count of *L. monocytogenes* when applied to cold-smoked salmon [60]. Kakaei and Shahbazi (2016) also reported improved shelf-life of minced trout fillets when wrapped with gelatin and chitosan-based films containing *Ziziphora clinopodioides* EO and grape seed extract as active agents than unwrapped fillets [61]. Active films not only reduced the microbial population (total viable counts, Enterobacteriaceae counts, psychrotrophic bacteria counts) but also lowered the total volatile base nitrogen and peroxide values during 11 day-storage period.

Future trends and perspectives

As a responsible member of the United Nations, Vietnam is committed to taking action to reduce plastic waste to protect the environment. Since 1990, there has been a spectacular increase in plastic use in Vietnam, rising from 3.8 kg/capita in 1990 to 81 kg/capita in 2019, only about 15 percent of the countrys plastic waste is recycled, and more than half, the equivalent of 3.6 MT/year is mismanaged [62]. To protect the environment, under the Master Plan on Plastic Waste Management in Vietnam adopted by Deputy Prime Minister Le Van Thanh in 2021, by 2025 Vietnam will use only environmentally friendly packaging at shopping centers and supermarkets and will collect, reuse, recycle and treat 85% of all plastic waste. In addition, food safety is of great and increasing importance to Vietnamese consumers, there were 373 outbreaks of foodborne diseases reported in 2014 and 2015 involving over 10,000 cases and resulting in 66 deaths [63].

Incorporation of nanomaterials into biopolymer improves properties of the fabricated films and coatings that lead to improvement in food quality and shelf-life extension, and simultaneously reduce use of synthetic plastics promoting healthy food and healthy environment. Further research is still needed to be undertaken with the aim of finding new sources of bioactive compounds from natural and new polymer blend combinations, determining the mechanical, barrier and functional properties of other types of natural polymerbased nanocomposite films/coatings, and allowing these products to compete with commercially available packaging materials.

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