CHAPTER 6

Biomediated Synthesis of Nanomaterials for Packaging Applications

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Abstract

Change in lifestyle of humans in this present generation with huge dependence on packaging materials has encouraged several studies on development of new variety of packaging materials. Emphasis on replacement of existing non-biodegradable packaging materials with biodegradable materials paved the way for the use of biopolymers. Lack of properties, such as thermal stability and mechanical strength in biopolymers led to the development of biopolymer nanocomposites by adding metal/metal oxide nanoparticles as fillers into the biopolymers. Metal/metal oxide nanoparticles improve mechanical/tensile strength, thermal stability as well as antimicrobial properties of the binding and receiving polymer matrix. Biomediated synthesis of metal/metal oxide nanoparticles result to development of novel packaging materials at a low cost and without releasing hazardous wastes into the environments. Novel packaging materials with metal/metal oxide nanoparticles as additives are capable of increasing the shelf life of food stuffs, in certain cases they act as indicators of quality food inside the package. Summarily, this present chapter focuses on biomediated synthesis of various metal/metal oxide nanoparticles and their applications in food packaging.

Keywords: biosynthesis, filler, additive, packaging material, antimicrobial, metal nanoparticles.

Introduction

The use of non-biodegradable packaging materials has been restricted in several countries to ensure environmental safety. Therefore, there is a need for development of alternative packaging materials with enhanced properties. The development of eco-friendly green packaging materials has potentials to reduce environmental impacts caused by synthetic packaging materials [1]. The biopolymers have been used as new alternative packaging materials, they include, but are not limited to, polysaccharides (starch and cellulose derivatives, chitosan and alginates), lipids (bees and carnauba wax, and free fatty acids), proteins (casein, whey and gluten), poly hydroxyl butyrate (PHB), poly lactic acid (PLA), poly caprolactone (PCL), poly vinyl alcohol (PVA), poly butylene succinate and their biopolymer blends [2]. Considering environmental safety, the biodegradable natural or biopolymers are preferred. However, the inherent properties and mechanical strength of natural polymers limit them from being utilized as packaging materials, especially on an industrial scale. It is often essential to have their surface modification or physical cross linking or even modified as a composite material [3].

The nanometals have potential to overcome certain limitations in properties of the biodegradable materials. The nanometals have exhibited preferable properties, such as high surface area, fine particle size, high reactivity, high strength and ductility, which make them suitable to be employed frequently in a diversified range of industrial fields [4]. The available vacuum sealed food packaging polymeric materials are permeable to oxygen and moisture, which leads to spoiling of foods. To prevent this situation, polymer packaging materials are coated with nanometal, which inhibits the penetration of oxygen and moisture, thereby preserves the food materials from spoiling [5]. These nanometals also act as antimicrobial agents by preventing growth of harmful microbes, thereby prevent food spoilage and extend their shelf life [6]. Beyond these, nanometals in food packaging also act as smart indicators of change in the chemical composition, pH, gas composition, among others, inside the package that containsfood stuff. These changes are communicated to the consumers by change in colour of the tag attached to them. Therefore, the role of nanometals in the food packaging include prevention of shelf life, fighting against the microbial growth and acting as smart indicators for the exact condition of the food stuff to the consumers. Metal and metal oxide (silver, gold, zinc oxide, silica, titanium dioxide, alumina and iron oxides), carbon based nanometals and nanosized polymers are most commonly used as nanofillers in food packaging applications [7].

Furthermore, the nanometals improves functionalities, such as durability, flexibility, temperature and flame resistance, barrier properties, optical and recycling properties of the recipient packaging materials [8]. Nanometals are added to polymeric packaging materials, such as polyamides (PA), nylons, polyolefin, ethylene-vinyl acetate copolymer, polystyrene (PS), epoxy resins, polyurethane, polyvinyl chloride (PVC) and polyethylene terephthalate (PET) [9]. Development of packaging materials using nanoparticles can be broadly classified into two categories/types: (I) biocomposite materials with inorganic metal nanoparticles as fillers [10, 11], (II) biocomposite materials with plant extracts/agro wastes as fillers [12]. Type I materials possess appreciable strength and shelf life and they are best suitable for packaging applications, whereas type II lacks shelf life and strength. The responses of the type II materials towards the environmental factors, such as humidity, temperature and pressure are higher, when compared with type I. The conventional methods of synthesis and distribution of nanometals into the matrix of the packaging materials results to generation and deposition of hazardous chemicals into the environments. In recent years, studies have been focusing on bioreduction process for the synthesis of nanometals. This is very simple, efficient and cost effective, but it consumes time [13]. In this method, the choice of bioreductant includes plant extracts, which contains flavonoids capable of reducing the metal to its nano state [14]. In this chapter, specific discussion on nanometals synthesized via bioreduction process and utilized as nanofiller in packaging materials is presented.

In spite of the advantages of utilization of nanometals in food packaging materials, most of studies on these materials are still on the stage of demonstration, and their real time applications are yet to receive approval concerning their safety issues, which could be caused by the migrations of nanomaterials from packaging to food stuffs [15] . Additionally, the absorption, distribution, metabolism, excretion and toxicological assessment of nanoparticles in food in humans are to be assessed [16]. Therefore, the use of nanometals in food industry provokes various assessment methodologies to ensure usage as well as the real-time analysis of nanometals in the environment and their impact on various levels of organisms [16]. This is an emerging and evolutionary area involving multidisciplinary studies and provides scope for interdisciplinary research.

Metal nanoparticles as fillers in packaging materials

The unique behaviors of metal nanoparticles have attracted researchers to employ them in many different types of matrices [17]. Though, these nanometal particles are added as nano fillers in a minimum quantity, they do not lose their properties, such as antimicrobial nature, thermal resistivity and/or tensile strength [18]. They usually enrich the properties the matrices to which they are added. The antimicrobial activity of metal nanoparticles varies accordingly with their methods of synthesis; either physical, chemical or biological [19]. Interest on biosynthesized metal nanoparticles increases incredibly, owing to their reliability and cost effectiveness [20]. The most preferred and commonly used metal nanoparticles are zinc, iron, copper, gold, aluminium, nickel and silver. In addition, certain metal oxides include titanium, zirconium, iron and zinc are also used as nanofillers in packaging applications. Among various aforementioned metal nanoparticles (MNPs), silver nanoparticles (AgNPs) is observed to be most appropriately employed in packaging materials, biomedical appliances, cosmetics, pharmaceuticals and textile sectors [21]. The toxicity of AgNPs is found to be very minimum in animal cells. Certain inorganic metal nanoparticles are also recognized as safe materials by US Food and Drug Administration (FDA).

Biosynthesized AgNPs

Biological synthesis of AgNPs using bacterial strains

Biosynthesis of AgNPs includes bioreduction of silver salts with various plant extracts and biological synthesis of AgNPs with bacteria, fungi and biomolecules [22]. Biological synthesis of AgNPs can be via either intracellular or extracellular mechanisms [23]. Comparatively, the extracellular mechanism is observed to be simpler and more economical. Though, the mechanism is not yet completely understood, it is predicted to take place by the presence of nitrate reductase enzymes released by microorganisms, which reduces metallic ions to metallic nanoparticles. Among the biological syntheses of AgNPs, the bacterial synthesis has been identified to be most convenient [24], owing to its availability and vulnerability for genetic modification. The shape, size and nature of AgNPs determine method of synthesis. Hence, many variety of bacterial strains are used for the biological synthesis of AgNPs. These include *Bacillus spp.* [25], *Streptomyces spp*. [26], *Acinetobacter spp*. [27], *Pseudomonas spp*. [28], to mention but a few. Minimum quantity of AgNPs synthesized by biological method using bacterial strains shows a good activity towards major strains of microbes from attacking food stuffs. Amongst them are *S. aureus, P. aeruginosa, B. subtilis* and *E. coli.*

Biological synthesis of AgNPs using fungi species

The biological synthesis of AgNPs using fungal species is observed to be efficient with extracellular mechanism. The fungal species extracted from plants are more efficient than other species [29]. The following species of fungi are most prominently tested for the biological synthesis of AgNPs, *Fusarium oxysporum* [30], *Guignardia spp*. [31], *Penicillium aculeatum* [32], *Alternaria spp*. [33], *Phenerochaete chrysosporium* [34]. AgNPs synthesized via biological method using fungi have antimicrobial activity against a variety of fungal species as well as bacterial strains; both Gram-positive and negative. Therefore, the AgNPs synthesis using fungal species is an effective process to be employed in packaging material, since it effectively attacks microbes attacking the food stuffs.

Bioreduction of AgNPs using plant extracts

The plant extracts with flavonoid contents are mainly reported bioreductants for the synthesis of AgNPs [35]. Almost all the parts of a plant with flavonoids are been used. The experimental condition and active flavonoid contents play major roles in geometry and activity of the AgNPs. In most cases, capping agents are not used during bioreduction of AgNPs with plant extracts, which results in aggregation of the reduced AgNPs. The agglomeration of AgNPs may lead to reduced behavior of AgNPs. In this case, certain researchers generated the AgNPs *in-situ*; inside the polymer films directly, using plant extract as a reducing agent [36- 38].

Anti-microbial activity of AgNPs

AgNPs have a wide spectrum of antimicrobial activity, including Gram-positive and negative bacteria, fungi and viruses. The mechanism of antibacterial activity of AgNPs have been studied based on its ability to release $Ag⁺ ions$ [39] and their potential to inhibit the growth metabolism of bacterial cells [40]. The mechanism of action of AgNPs against bacteria is proposed in three ways [41]: (1) penetration of AgNPs in the range of 1 to 10 nm into the cell membrane and interfere with its respiration, (2) interaction of AgNPs with the compounds of

sulphur and phosphorus, such as DNA and (3) release of active silver $(Ag⁺)$ ions, which reacts with negatively charged cell membrane and damages them.

Moreover, the release of Ag⁺ ions are influenced by availability of oxygen atmosphere. In anaerobic condition, the antibacterial effect is almost nil even with higher concentration [40]. The antimicrobial activity of AgNPs is also influenced by the shape and size of the synthesized AgNPs, the symmetry of the particles which offers greater contact surface have greater antimicrobial activity.

Packaging materials with AgNPs

The addition of nanoparticles into the already existing packaging materials provides improved physical-chemical properties, reduces hydrophilic behavior and induces biodegradability and antimicrobial activity. AgNPs are added as fillers in both biodegradable and non-biodegradable polymers. Few examples of food packaging material with AgNPs are presented in Table 1.

Packaging material		Type of food material Preferred storage period	References
PVC/AgNPs	Beef	Up to 14 days; 4 ± 2 °C	$[42]$
	Dried fruits and nuts	Up to 21 days; 4 ± 2 °C	[43]
LDPE/AgNPs	Meat, pork and chicken	Up to 21 days; 4 ± 2 °C	[44]
	Cheese	Up to 28 days; 4 ± 2 °C	[45]
PLA/AgNPs	Cheese	Up to 25 days; 4 ± 2 °C	[46]
	Mangoes	Up to 15 days; room temperature	$[47]$

Table 1: Food packaging materials with AgNPs as additives.

Biosynthesized ZnNPs/ ZnONPs

Among various inorganic metal oxide nanoparticles available, Zinc oxide nanoparticles (ZnONPs) are more preferred, because they are inexpensive and easy to prepare. The US FDA has enlisted ZnONPs as part of safe materials [48]. As mentioned in the previous sub-sections, similar to AgNPs, the ZnNPs/ZnONPs can also be synthesized via bioreduction process, using plant extracts as well as biological synthesis with microorganisms. The following strains of bacteria have been effectively utilized for the biological synthesis of Zn/ZnONPs: *Aspergillus strain, Aspergillus fumigatus, Aspergillus terreus* [49], *Candida albicans* [50]. In addition, few used algal strains include *Chlamydomonas reinhardtii* [51], *Sargassum muticum* [52] and *S. myriocystum*.

Moving forward, biosynthesized Zn/ZnONPs possess appreciable antimicrobial activity against broad spectrum of bacteria and fungi. ZnNPs/ZnONPs also possess antitumor activity, it is cytotoxic and genotoxic towards certain types of human cells [53]. Further large scale production of ZnONPs by bioreduction process is possible [54], by coating the synthesized ZnONPs over cotton fabric, which possesses antimicrobial activity and washing durability. The biosynthesized ZnNPs/ZnONPs show a better catalytic activity than those ZnNPs/ZnONPs obtained from chemical methods. ZnONPs synthesized from the fungal strain, *Aspergillus fumigatus* is stable up to 90 days, while ZnONPs obtained from seaweed, *Sargassum myriocystum* is stable up to 6 months [55]. In case of bioreduction of Zn/ZnONPs using plant extracts, the carboxylic and phenolic groups present in the extract act as bioreductants as well as capping agents [56].

Packaging materials with ZnNPs/ ZnONPs

Zn/ZnONPs are well known for their stability under extreme conditions. They are effective against a wide spectrum of microbial strains, more specifically the food born microbes at a lower concentration [53, 57]. ZnONPs possess a unique property of filtering ultraviolet (UV) rays [58]. Addition of Zn/ZnONPs to both biodegradable or non-biodegradable polymer matrices increases the mechanical properties of the polymer, such as tensile strength, Young's modulus and thermal resistance [59]. ZnONPs coated packaging materials are suitable for storing fish samples [60]. Zn/ZnONPs coated packaging materials also act as a good scavenger of oxygen. All the aforementioned characteristics qualify Zn/ZnONPs to be used as potential additives for packaging applications [61].

Packaging Material	Type of food material	Preferred storage period	References
PVC/ZnNPs	Cheese	$30-40$ days	[62]
	Sliced apples	Decay rate decreased by 60%	[63]
PLA/ZnONPs	Sliced apples	Decay rate decreased by 65%	[64]
OBG/ZnONPs	Spinach	Up to 7 days	[65]
PU/ZnONPs	Sliced carrot	Up to 9 days	[66]
PVA/ZnONPs	Aqueous food stuffs	pH indicator	[67]

Table 2: Food packaging materials with ZnNPs as additives.

Biosynthesized CuNPs/ CuONPs

Copper metal is well known for its antimicrobial activity from the ancient days. This is the reason behind the use of copper vessels for storing water. Even now, copper nanoparticles (CuNPs) are used for water treatment. CuNPs/CuONPs have commendable antimicrobial activity against a variety of bacteria, fungi and viruses [68]. The mechanism of interaction of CuNPs/CuONPs with microbes is through their cell membranes, thereby making them inactive. The bioreduction of CuNPs is successful with extracts of various plant species, such as, *Citrus medica Linn*. [69], *Ziziphus spina-christi* [70], *Asparagus adscendens Roxb*. Used root and leaf [71] include *Eclipta prostrata* leaves [72], *Ginkgo biloba Linn.* [73], *Plantago asiatica* leaf [74], *Thymus vulgaris L.* [75], black tea leaves [76], to mention but a few. Biological synthesis of CuNPs can be carried out with the following bacterial strains: *Escherichia coli, M. psychrotolerans* and *M. morganii RP42*. Biomediated synthesis of CuNPs/CuONPs is an economical as well as simple method, which can be executed with minimum infrastructures. In certain cases, CuNPs synthesized are observed to be more efficient than those synthesized via commercial chemical methods.

Packaging materials with CuNPs/ CuONPs

CuNPs are effective materials against both Gram-positive and negative organisms [77], which make them more suitable for packaging applications. The antimicrobial effect of CuNPs/ CuONPs more specifically towards *E.coli* in food stuffs and ability of CuNPs/CuONPs to be blended into polymer suggest them to be good additives in food packaging materials [78]. Impregnation of CuNPs/CuONPs into polymer films or biopolymer films [79] enhances their tensile strengths, transparency, thermal stability, mechanical strengths [80]. More also, it adds to the antimicrobial activity, UV barrier property and prolonged shell life, which make the material suitable for food packaging applications [81]. CuNPs coated packaging materials act as freshness indicator in case of meat, by turning dark by reacting with the volatile sulfide released during the spoilage of meat [82]. Currently, researchers are interested in examining the CuNPs/CuONPs impregnated films for electronic as well as catalytic applications.

Table 3: Food packaging materials with CuNPs/CuONPs as additives.

Titanium dioxide nanoparticles used as additives in food packaging

Titanium dioxide nanoparticles (TiO₂NPs) are effective additives in food packaging, due to their thermal stability, economical, non-toxic and stability properties towards UV light. TiO2NPs are extensively employed as promising photo catalysts in various types of reactions [88]. They are also used in water treatment and self-cleaning applications. Due to the antimicrobial property of $TiO₂NPs$, the interest of several researchers have increased towards using them in food packaging. Unlike previously discussed metal nanoparticles, TiO₂NPs are more prone to agglomeration; therefore, ionic surfactants are used as capping agents to prevent agglomeration. TiO2NPs are also synthesized from various plant extracts [89] and microbes [90]. TiO2NPs can be fabricated with biodegradable fish skin gelatin [91], potato starch [92], pectin [93], super hydrophobic paper [94], poly vinyl chloride [95], polyethylene [96] and PLA [97]. Besides, TiO2NPs as additives in packaging materials improve the mechanical strength, tensile strength, hydrophobicity, thermal stability, transparency, water vapour permeability, UV transmittance and antimicrobial properties. More specifically, application of $TiO₂NPs$ has been effectively analyzed for foodstuffs, such as bread [98] and strawberry [99] for their nutritional values and decay periods. Also, TiO₂NPs are used for controlling hematophagous fly and sheep-biting louse $[100]$. Biosynthesized TiO₂NPs are employed in solar cells $[101]$.

Other metal and metal oxide nanoparticles used as additives in packaging

The magnesium oxide nanoparticles (MgONPs) are recognized as safe materials by the US FDA. MgONPs are added as additives to PLA, poly ethylene (PE) and biodegradable polymers. MgONPs improve oxygen barrier, tensile strength and antibacterial property of polymer packaging materials [102], but they have poor water barrier property. Similarly, iron oxide nanoparticles (FeONPs) additives to polypropylene (PP), PVA and biopolymers show good gas barrier and improved thermal stability [103]. Also, silicon dioxide nanoparticles (SiO2NPs) provide good insulation, low toxicity and stability to polymer, when they are used in packaging materials [104].

Conclusions

The use of biosynthesized metal/metal oxide nanoparticles as additives in packaging materials has been elucidated. It is evident that they increase shelf life of foodstuffs by inducing water vapour permeability, UV barrier, gas barrier and antimicrobial properties. Metal/metal oxide nanoparticles also increase thermal stability and mechanical/tensile strength of the receiving polymers used in packaging materials.

Besides, metal/metal oxide nanoparticles are found to be compatible with both synthetic as well as biopolymers. Biomediated synthesis of metal/metal oxide nanoparticles result to manufacturing of intelligent packaging materials in an economical and eco-friendly manners. Most metal/metal oxide nanoparticles are recognized as safe materials by US FDA. Therefore, biomediated synthesis of metal/metal oxide nanoparticles and their application in food packaging are important research areas to be explored and promising field to satisfy our dayto-day needs.

List of abbreviations

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