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EDIBLE NANOFILLER FOR DEVELOPMENT OF EDIBLE BIO-NANOCOMPOSITE FILM: A REVIEW

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ABSTRACT

For recent years, application of biopolymer as an edible film has received growing interest because of their advantages including environmental friendly and non-toxic food barrier property. However, the mechanical and barrier properties of biopolymer film are poor compared to conventional synthetic film. Incorporation of nanofiller into biopolymer matrix may become a novel innovation in food packaging industry. Nano-sized fillers provide large contact surface area that favors strong matrix-filler interaction compared to micron-sized fillers. They also provide other desired functions of active food packaging film including antimicrobial agent, biosensor, and oxygen scavengers. The objective of this paper is to provide a general review on the potential edible nanofiller, preparation, properties and performances as well as application of nanofiller into edible film.

Keywords: Edible film, edible nanofiller, biopolymer, bio-nanocomposite

INTRODUCTION

Biopolymer has the potential to replace conventional plastic from fossil based. Recently, application of biopolymer as edible film has led to a new interest in food packaging technology. Edible film is a thin layer of edible material applied on food products for preservation, distribution, and marketing [1]. Edible film is more suitable to be used as food packaging because it is a promising non-toxic and non-pollutant product which is safe for human use [2], [3]. Edible film made up from natural and biodegradable agriculture source is easy to decompose and environmental friendly.

Ideal edible film should have excellent mechanical and barrier properties. As food packaging film, their main functions are to maintain food quality, hinder gain or loss of moisture, prevent bacteria growth, and act as barrier to oxygen, water vapor, carbon dioxide, and volatile compounds [4], [5]. However, edible film produced from biopolymer has low mechanical and barrier properties compared to fossil based plastic. Implementation of nanotechnology into biopolymer material has the potential to improve the lack properties of biopolymer [5], [6]. It consists of biopolymer matrix such as starch, cellulose, agar, whey protein and chitosan reinforced with nanofiller having dimensions smaller than 100 nm [2].

There are lots of recent works on bio-nanocomposite films as edible films such as development of alginates from seaweed with nanocellulose [7], starch with cellulose nanofibrils [8], and hydroxypropyl methylcellulose (HPMC) with chitosan nanoparticles [9][10]. Abdollahi et al. [7] have compared the properties of bio-nanocomposite films filled with organic nanofiller (cellulose nanoparticle) and non-organic nanofiller (montmorillonite). Tensile strength (TS) and Young's modulus (E) of the nanocomposite films has improved with the increase in cellulose nanoparticle content compared to nanocomposite with montmorillonite (MMT). This is because similar polysaccharide structure of cellulose nnoparticle and alginate has given good interfacial interactions. He concluded that organic nanofiller was better to improve biopolymer film properties compared to non-organic nanofiller. This finding shows that it is very important to compile and review the work that has been done by previous researchers on the edible bio-nanocomposite in order to develop efficient edible food packaging material. The objective of this paper is to provide a general review on the potential edible nanofiller, preparation, properties and performances as well as application of nanofiller in edible film.

NANOFILLER

Nanofiller may improve desired properties of food packaging materials such as tensile strength, thermal stability, and barrier properties [11]. Incorporation of nanofiller into biopolymer may produce bio-nanocomposite. However, a critical issue with nanocomposite is the possibility of migration of nanofiller from food package into food products because of its very small dimension (<100nm), thus easier to enter food tissue. Nanofillers have the potential to harm humans and environment as it is possible to increase toxicity [12]. For example, clay and zinc oxide nanofillers which are possibly toxic may give adverse effect on human health [13]. Therefore, organic nanoparticles such as cellulose, chitin and chitosan [14] may be used as alternative nanofiller to produce bio-nanocomposite film which is edible.

Cellulose nanoparticle

Cellulose is the most abundant bio-material as it can be obtained from plant sources such as ramie, grass fiber; bacterial cellulose, and algae tunicates [15]. Application of cellulose nanoparticle into food packaging materials has gained increasing attention due to their high strength, stiff, light weight, biodegradability, and renewability [16]. The main steps of cellulose nanoparticle preparation are milling of raw materials, treatment with alkali, bleaching treatment with sodium chlorite ($NaClO_2$) to eliminate lignin and cellulose from lignocellulosic component [17]. Then, bleached fibers must undergo further treatment of acid hydrolysis to produce cellulose nanoparticle. However, separation of plant fibers into smaller elementary constituent is a very challenging and costly process. This limitation has encouraged researcher to find alternative sources of nanoparticles such as chitosan and starch.

Chitin Nanofibrils

Chitin is a white, hard and inelastic mucopolysaccharide which is easily obtained from crab, shrimp and fungal *mycelia* [18]. Chitin can be extracted by acid treatment to dissolve calcium carbonate followed by alkaline treatment to dissolve proteins. Further deacetylation under alkaline condition will produce chitosan [19]. Chitin nanoparticles can enhance strength of starch-based materials, exhibit antifungal, and improve barrier properties [20]–[23]. Nonetheless, application of chitin nanofiller as food packaging application is still lacking and quite untouched because solid state chitin has a compact structure and insoluble in most solvents [19]–[21], [24], [25].

Chitosan Nanoparticles

Chitosan can be produced from deacetylation of chitin and it is the most abundant polysaccharide after cellulose [18], [26]. Unlike chitin, chitosan is easily soluble in aqueous acidic solution and thus, it is widely used in different applications as solution, gel, film or fibre [19]. It is also non-toxic, biodegradable, biofunctional, biocompatible, and exhibit antimicrobial properties [27]–[30]. Studies on producing chitosan nanoparticle and its applications have been done since several years ago. There are several methods to produce chitosan nanoparticles freshly in laboratory. The most popular method to produce chitosan nanoparticle is through ionic gelation [31], [32]. This method has been applied in most study because it possesses many advantages over other method such as avoiding the use of toxic reagents, simple, mild process, improve biocompatibility and reduced undesirable effects [32], [33]. During the process of ionic gelation, positive charged amino group on chitosan react with negative tripolyphosphate (TPP) ions at room temperature and form molecular linkages [34]. Then, chitosan nanoparticle is formed spontaneously with overall positive surface charge [35]. Formation of chitosan nanoparticle depends on the concentration of TPP added into the chitosan. Optimum mass ratio of chitosan/TPP to form chitosan nanoparticle was found to be 5:2 [36].

EFFECT OF NANOFILLER ON MECHANICAL PROPERTIES OF EDIBLE FILM

Nanosized filler may increase the contact surface area within biopolymer matrix which favors the strong filler-matrix interaction compared to microcomposite [4], [13]. Good affinity between biopolymer matrix and nano-sized filler can enhance mechanical properties due to the high rigidity of the nanofiller [2]. Antoniou *et al.* [37] has done a study to compare the mechanical properties between biocomposite of bulk chitosan/tara gum film and bio-nanocomposite of chitosan nanoparticle/tara gum film. Tensile strenght of bio-nanocomposite was found to be higher than biocomposite although the same amount of chitosan nanoparticle (CNP) was used. However, elongation of nanocomposite slightly decreased when amount of CNP used was increased. This findings show that addition of nano-sized filler has the potential to improve film strenght but not effective for film elasticity.

Size of nanofiller also play an important role to improve mechanical strength of bio-nanocomposite. Moura *et al.* [9] reported that tensile strength of hydoxypropyl methylcellulose (HPMC) films with bigger CNP diameter (221 nm) was below 40 MPa had increased to a maximum value of 62.6 MPa when the particle size was reduced to 85 nm [38]. CNP tend to occupy the empty pores of HPMC. This study reveals that small surface area of nanofiller has the ability to improve fillers-matrix interaction and increase mechanical strength of composite.

Apart from particle size, amount of nanofiller added can also affect the mechanical properties of nanocomposite. Chang et al. [39] studied the effect of CNP concentration on the mechanical properties of potato starch film. He found that increasing CNP amount improved the tensile strength of composite but decreased the elongation of composite. When the CNP amount was varied from 0-6% weight of starch, the tensile strength increased from 2.84 to 10.80 MPA. However, adding more CNP into the matrix (> 6% weight of starch) will lead to the agglomeration of the nanofillers which reduced the affinity and molecular interaction between fillers and matrix. Therefore, rigidity properties of the film may decrease and reduce the strength of the produced films [40].

EFFECT OF NANOFILLER ON BARRIER PROPERTIES OF EDIBLE FILM

Good dispersion of nanofiller with large aspect ratio in biopolymer matrix layer will lead to diffusive path become tortuous [39], [41]. Tortuous path (Fig 1) forcing water vapor, oxygen, carbon dioxide, and others volatile materials to travel through the polymer matrix in longer path length for diffusion and reduce the rate of water vapor diffusion [2], [42], [43].

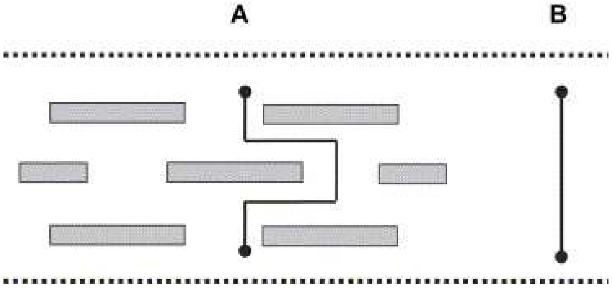

Fig. 1 Schematic illustration of the tortuosity of (A) filled polymer (B) unfilled polymer [44]

Azerado et al. [41] reported that addition of at least 0.1% weight of cellulose nanoparticle (CN) into mango purees films can improve the water vapor barrier of the films. This result is similar to results reported by Chang et al. [39]. When 1–4% of chitosan nanoparticle was added to the starch, the water vapor permeability (WVP) barrier obviously increased. However, when more than 6 wt% CN was added to the starch, WVP barrier were slightly decreased [45]. This was because of nanoparticles agglomeration in the film which produces poor structure of nanocomposite film.

POTENTIAL APPLICATION OF NANOFILLER

Organic nanofiller can be incorporated into biopolymer matrix such as hydroxypropylmetylcellulose (HPMC), starch, fish gelatin, and fruit fibre to produce edible film. Application of nanofiller into edible film can enhance desired properties of food packaging such as improve mechanical strength, barrier ability, thermal stability, antioxidant carrier, and inhibit microbial growth [46]. For example, edible film filled with CNP can be applied to fresh cut product, meat, chicken, and other fresh food as it has the ability to control microbial growth and prolong the product shelf life [47]. Addition of CNP into fish gelatin films may be applied directly on food contact to protect food from excessive dehydration, light and oxygen [27], [48]. Another desired properties of food packaging material is able to withstand heat treatments during preparation, sealing, and storing of food [49], [50]. Incorporation of nanofillers into biocomposite may also enhance thermal stability of edible film [9], [15], [28], [51]. Table 1 shows examples of application of nanofiller as reinforcing agent in edible film.

Table 1 List of nanofiller and its application

Nanoparticle	Sources	Mean diameter (nm)	Application
Cellulose nanoparticle	Microcrystalline cellulose	392±36nm	Potato starch film [52]
	Mulberry pulp fiber	40-50nm	Agar film [53]
	Coconut fiber[a] Cotton fiber[b]	5.3-5.5[a] 12[b]	Alginate-acerola puree film [54]
	Bacterial cellulose pellicles	20±5	Gelatin film [55]
	Commercial cellulose nanofiber	7.2	Mango puree film [41]
Chitin Nanofibril	Commercial chitin from crab shell	25-300	Carrageenan film [56]
	Yellow squat lobster waste	90	Thermoplastic starch film [21]
Chitosan Nanoparticle	Chitosan (MW 71.3kDa, degree of deacetylation 94%)	59-110	Hydroxypropyl methylcellulose films [10]
	Chitosan (MW 71.3kDa, degree of deacetylation 94%)	85-221	Hydroxypropyl methylcellulose films [9]
	Chitosan originating from shrimp shell	50-100	Potato starch films [45]
	Chitosan (medium molecular weight, deacetylation 75-85%)	40-80	Fish gelatin films [27]

CONCLUSION

Organic nanofillers can be produced from their sources such as crab shell, ramie, grass fiber and others through several processes such as deacetylation and bleaching treatment. Reduction of micron size filler into nano size can open up a new potential to meet up food packaging demand as it can enhance mechanical, thermal, and barrier properties. They also exhibit other desired properties such as antimicrobial, protect food nutrition, and easily to degrade as well as environmental friendly. Nanofiller from natural source is non-toxic for food packaging application and thus reduce the potential of adverse effect on human health. Although studies on edible nanofiller are still new, it promises many benefits to food packaging development as it has the potential to replace conventional plastic from nonrenewable sources thus safe the environment. Further studies are needed to find more potential edible nanofiller and maximize the desired quality of the edible film in order to replace conventional plastic.

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