Emerging Trends in the Development of Chitosan Films for Active Food Packaging

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ABSTRACT
The demand for healthy and safe foods with the minimum use of synthetic materials is increasing very rapidly. Plastic polymers have negative effects on the environment, and great efforts have been made to evaluate the use of various bio-based polymers as alternatives to synthetic plastic packaging. Chitin and chitosan are mostly extracted from crustacean shells or by products from processing and handling fish and seafood. As a result, chitosan as a material for food packaging has received much attention and is widely researched because it has unique biological and functional properties. Chitosan for food packaging and food preservation applications can be applied via the method of spraying, immersing, coating, or wrapping films. However, chitosan has several disadvantages including low mechanical properties and high sensitivity to moisture, which are the main limitations for its industrial applications, including food packaging. In this study, the scientific literature of the last 5 years on chitosan-based films for their potential application in the food packaging industry has been extensively reviewed. This review also discussed various strategies that have been implemented to improve the properties of chitosan film, including using plasticizers and cross-linking agents, filling them with nanoparticles, fibers and whiskers and combining them with natural extracts and essential oils as well as with other natural and synthetic ingredients.

Keywords: chitosan, food preservation, edible film, essential oils, nanoparticle

1. INTRODUCTION
Packaging is very important in providing protection for products and ensuring food safety during distribution and during storage. Conventional food packaging in the form of plastic based on synthetic materials is the biggest source of environmental pollution because it cannot be degraded in nature. Therefore, bio-based and biodegradable materials have been widely studied to address this problem in recent years (Haghighi et al., 2020).

Along with the times, consumer demand for variety and quality of food products is increasing throughout the year. This fact has driven new developments in food packaging technology like never before to ensure the availability of safe and healthy food. The function of food packaging has shifted, where passive packaging is only limited to separating or blocking food ingredients from the surrounding environment and minimizing or preventing exposure to spoilage-causing factors, including microorganisms, oxygen, temperature and humidity (Kumar et al., 2020). Although passive packaging can delay the detrimental effects of the environment on food products, it is not effective enough in maintaining the quality and safety of fresh, additive-free, nonprocessed or minimally processed foods during prolonged storage (Mexis and Kontominas, 2014).

One of the advanced technologies in food packaging is the application of active packaging. Based on European Regulation (EC) No. 450/2009, active packaging is defined as an interacting packaging system with food by deliberately inserting active components that can release or absorbs substances into or from packaged food or the surrounding environment (European Commission, 2009). Active
packaging can work through different systems, including an active-releasing system (emitters) that adds compound to the headspace or to packaged food, and active-absorbing system (scavengers) work by removing unwanted compounds from food or its environment (Yildirim and Röcker, 2018). Biodegradable polymers derived from renewable resources have been proven in many studies to be able to function as packaging materials for future generations, including chitosan. Chitosan is commercially produced from the deacetylation process of chitin (Verlee et al., 2017), which is the second most abundant polysaccharide in nature after cellulose (Salari et al., 2018) and is contained in crustaceans, insects, and fungi (Al Hoqani et al., 2020). The use of chitosan can solve the problem of plastic waste accumulation because it is biodegradable (Priyadarshi and Rhim, 2020), plus its antimicrobial activity which can fight bacteria and fungi that cause food spoilage (Duan et al., 2019), making it an attractive candidate for continuous development as an active food packaging material. However, chitosan also has several limitations. Like other natural polymers, chitosan has low mechanical and thermal stability. Chitosan also has high sensitivity to moisture that make it difficult to apply on an industrial scale (Elsabee and Abdou, 2013). One of the strategies that can be implemented to overcome these inherent drawbacks is to mix chitosan with other polymers to obtain their superior properties while minimizing their weaknesses (Haghighi et al., 2020). Chitosan can be blended with natural or synthetic polymers to enhance the mechanical and thermal properties of films.

This review article collects all the important articles related to the latest advances in chitosan as an active food packaging material and several strategies implemented to improve the quality of chitosan-based films.

2. POTENTIAL OF CHITOSAN

2.1 Source and Production

Chitin consists of three types of reactive functional groups, which are amino groups at C-2, and primary and secondary hydroxyl groups at the C-3 and C-6 positions, respectively (Kumar et al., 2020). Chitin is mainly present as the main constituent of the exoskeleton in marine invertebrates and insects, as well as a component of the cell wall in certain fungi (Al Hoqani et al., 2020).

The process of producing chitosan from animal wastes (skins and shells) is generally carried out in two main steps, namely the extraction of chitin from the waste and followed by the conversion of chitin to chitosan (Priyadarshi and Rhim, 2020). Chitin extraction can be done in two ways, namely by using chemical method and biological method. Chemical method is widely used on a commercial scale because of its shorter processing time compared to biological method, although there are some disadvantages such as uneconomical and not environmentally friendly (Arbia et al., 2013).

Extraction of chitin from marine shell waste in the industrial level usually uses chemical methods. The most common industrial process applied for chitin extraction consists of three main steps: deproteinization of the raw material by addition of an alkaline solution, demineralization with the use of an acid solution, and finally discoloration of product by the addition of an alkaline solution (Muxica et al., 2017; Philibert et al., 2017). Biological method is safe and has become a very popular topic these days, but its use still limited to laboratory research (Kaur and Dhillon, 2015).

Conversion of chitin to chitosan can be done through enzymatic or chemical processes. The enzymatic process employs chitin deacetylase enzyme which has complicated fermentation requirement in the preparation step (Priyadarshi and Rhim, 2020). The chemical conversion is preferred because of its lower cost and suitable for large quantity production (Younes and Rinaudo, 2015). The chemical deacetylation of chitin usually involves treatment using hydroxides at high temperatures. Under these conditions, the deacetylation occurs really fast (Muxica et al., 2017).

2.2 Deacetylation Degree and Antimicrobial Activity

The degree of deacetylation (removal of acetyl groups) (DD) is one of the most significant chemical characteristics of chitosan. The DD of chitosan is defined as the ratio of \( \text{d-glucosamine} \) units to the number of \( \text{d-glucosamine} \) and \( \text{N-acetyl d-glucosamine} \) units present in the chain (Kumar et al. 2020).

Chitosan can be claimed as the only natural polysaccharide that has antimicrobial activity against bacteria, fungi, and yeast. The nature of this antimicrobial activity depends on its cationic nature, deacetylation degree (DD), concentration, period of exposure, and the type of organism being tested (Priyadarshi and Rhim, 2020). In many studies, chitosan has been shown to have good antimicrobial property against various microorganisms such as Gram-positive and Gram-negative bacteria, filamentous fungi and yeast (Hosseinnejad and Jafari, 2016; Pan et al., 2015).

There are several proposed mechanisms for the antimicrobial activities of chitosan, but the exact mechanism is not yet clear (Kumar et al., 2020). The following are the mechanisms proposed by several researchers:

(i) Interaction of negatively charged microbial cell membranes with positively charged amine groups in chitosan changes the barrier properties of the membrane, leading to leakage of intracellular contents, and ultimately to cell death (Hosseinnejad and Jafari, 2016).

(ii) The chitosan layer on the food surface limits the gas exchange between the food and the outside environment. This causes a barrier to oxygen
transport, thus making the gas state confined to the aerobic microbes in the food, causing them to be eliminated (Dotto et al., 2015).

(iii) Formation of a protective layer by chitosan on food surface, so that physically prevent it from attack by food spoilage microbes (Kumari et al., 2015)

(iv) Chitosan forms an impermeable layer around bacterial cells and blocks the exchange of important solutes between the interior and the exterior resulting in failure of the metabolic system and ultimately cell death (Diez-Pascual and Diez-Vicente, 2015).

3. CHITOSAN FILMS

3.1 Chitosan Pure Films

Films derived from pure chitosan have been prepared and studied, without the use of other polymers. This chitosan-based film has also been applied to extend the shelf life of several food products such as vegetables, fruits, fish, and meat with different application methods such as spraying, immersing, coating, or wrapping (Suseno et al., 2014).

However, these films still need the addition of other materials to help film formation such as dilute acids, plasticizers, and emulsifiers (Priyadarshi dan Rhim, 2020). In the film fabrication, chitosan is usually dissolved in an acidic solutions, such as acetic acid or hydrochloric acid. Plasticizers such as glycerol are often used to increase the film flexibility and strength (Irawan et al., 2019), while emulsifiers have also been used for property enhancement such as Tween 20, Tween 80, Span 20, and Brij 56 (Rui et al., 2017; Zahid et al., 2012).

3.2 Chitosan and Its Blends Films with Natural and Synthetic Polymers

Chitosan has to be blended with other natural or synthetic polymers to improve the physical, thermal, mechanical, barrier, and optical features of films. Chitosan-based films were blended with biopolymers such as polysaccharides, proteins, lipids, and their derivatives to enhance its functionality as a packaging material for food. It has been found that chitosan can be blended with starch, cellulose, alginate, gelatin, protein, and some other biopolymers (Hasan et al., 2020; Deng et al., 2017; Arroyo et al., 2020; Hu et al., 2020; Samsalee and Sothornvit, 2019).

Chitosan has been blended with synthetic polymers such as polyvinyl alcohol, polyvinyl pyrrolidone, and polyactic acid to maintain the physical, mechanical, and biological properties of composite films (Yu et al., 2018; Lo’ay and Taher 2018; Fathima et al., 2018).

3.3 Chitosan Films with Plant Extracts and Natural Oils

Chitin, chitosan, and their derivatives have been reported to have natural antioxidant activity (Anraku et al., 2018). Several studies have also resulted in increased antioxidant activity with the addition of active substances derived from plant extracts containing phenolic compounds (Kumar et al., 2020). Chitosan-based films which were embedded with ginger essential oils showed a great potential use as active packaging for shelf life extension of food products (Irawan et al., 2019; Bonilla et al., 2018). An increase of film elasticity was also obtained after the incorporation of the oil (Bonilla et al., 2018).

Another study used tea extract containing polyphenols and showed more excellent antioxidant and antimicrobial activity compared to chitosan film alone (Zhang and Jiang, 2020). A study also reported the preparation of active film from chitosan and kombucha tea. It revealed that the addition of kombucha tea into chitosan films increase the antioxidant activity and water vapour permeability. Moreover, chitosan/kombucha tea film extended the shelf life of minced beef and retarded microbial growth significantly (Ashrafi et al., 2018).

Some plant extracts that have been used as additives in the manufacture of chitosan-based films include grapefruit seed extract (Wang et al., 2019), lemon essential oils (Jiang et al., 2020), cinnamon oil (Mohammadi et al., 2020), and turmeric essential oil (Li et al., 2019).

3.4 Chitosan Films with Nanoparticles

Metal and metal oxide nanoparticles have long been found to be applied as fillers in food packaging materials because of their extraordinary properties on antimicrobial and antioxidant activity which as a result can increase the shelf life of the product (Priyadarshi and Rhim, 2020). For those purposes, zinc oxide (ZnO), titanium dioxide (TiO2), and silver (Ag) nanoparticles have been commercially used as antimicrobial agents in the preparation of chitosan-based active film (Amjadi et al., 2020; Wang et al., 2020; Cao et al., 2020; Qin et al., 2019; Lin et al., 2020; Kustiningsih et al., 2019).

In addition to nanoparticles in the form of metal and metal oxides, nanoclays or biopolymers in the form of nanoparticles can also be used such as nano-sized chitin, cellulose, and lignin (Priyadarshi dan Rhim, 2020).

4. CONCLUSION

Chitosan is a biopolymer whose availability is very abundant in nature and has enormous potential in the field of food packaging technology. Chitosan has superior properties, among others, it is easily degraded naturally and has excellent antimicrobial activity to fight bacterial and fungal proliferation which can cause food spoilage. To improve the properties of chitosan film as an active packaging for food products, it can be combined with other natural such as starch, cellulose, alginate, gelatin, and protein.
and/or synthetic polymers such as polyvinyl alcohol, polyvinyl pyrrolidone, and polyactic acid. 

In order to improve antimicrobial and antioxidant activity, chitosan films can be enriched with plant extracts or essential oils. Nanomaterials like ZnO, TiO$_2$, and Ag are also frequently used to improve antimicrobial properties of packaging materials including chitosan film

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6. REFERENCES


