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SUSTAINABLE ALTERNATIVE FOR FOOD PACKAGING: CHITOSAN BIOPOLYMER - A REVIEW

Amalia Carmen MITELUȚ, Elisabeta Elena TĂNASE, Vlad Ioan POPA, Mona Elena POPA

University of Agronomic Sciences and Veterinary Medicine of Bucharest, 59 Mărăşti Blvd., District 1, 011464, Bucharest, Romania, Phone: +4021.318.25.64, Fax: +4021.318.25.67

Corresponding author email: amaliamitelut@yahoo.com

Abstract

Food packaging technology is continuously evolving in response to growing challenges from a modern society. Active packaging is an innovative approach to enhance the shelf life of food stuffs while improving their quality, safety and integrity. Chitosan is the deacetylated derivative of chitin, which is the second most abundant polysaccharide found in nature after cellulose. Chitosan is nontoxic, biocompatible, and biodegradable and thus is considered as an environmentally friendly packaging material. Moreover, chitosan is a good inhibitor against the growth of a wide variety of yeasts, fungi and bacteria, and also displays gas and aroma barrier properties in dry conditions. Along with these characteristics, its ease for film formation, make chitosan an interesting choice for active antimicrobial food packaging applications. This study aims to present a literature review regarding chitin and chitosan biopolymers, their properties and the ability to be used in applications in food packaging industry.

Key words: *active packaging, chitosan, biodegradability, biocompatibility, antimicrobial applications.*

INTRODUCTION

Major current and future challenges to fast-moving consumer goods packaging include legislation, global markets, longer shelf life, convenience, safer and healthier food, environmental concerns, authenticity, and food waste (Realini & Marcos, 2014). Environmental concerns enhance and stimulate the use of renewable resources for producing economically convenient applications to maintain or even improve life quality (Pereda et al., 2012).
Biopolymers have been widely investigated over the last two decades because they can be a viable solution to the waste disposal of foods' plastic packaging materials (Nitschke et al., 2011; Cooper, 2013). Furthermore, biopolymer films are excellent matrix for incorporating a wide variety of functional additives, such as antioxidants, antifungal agents, antimicrobials (Zemljic et al., 2013), colours, and nutrients, and through these, active materials can also improve food quality and extend shelf life by minimizing microbial growth in the product (Abdollahi et al., 2012). Such matrix biopolymers include starches, cellulose derivatives, chitosan/chitin, gums, proteins (animal or plant-based) and lipids. These materials offer the possibility of obtaining thin films and coatings to cover fresh or processed foods to extend their shelf life (Elsabee & Abdou, 2013).
Nowadays, consumer's awareness regarding unhealthy side effects of chemicals (i.e. parabens, benzoic acids, nitrites, etc.) in food products is increasing. Food spoilage due to microbial action is also a problem (López-Carballo et al., 2012; Huang et al., 2012), resulting in the loss of more than 25% of food before consumption. A multitude of yeasts, moulds and bacteria can cause the deterioration of a specific food product, the responsible microorganism action being dependent on pH, water activity, oxygen and carbon dioxide partial pressures and temperature (López-Carballo et al., 2012). The incorporation of antioxidant and antimicrobial agents into edible films is one of the landmark advances in food technology today. For environmental reasons it is increasingly desirable for at least the packaging to be biodegradable and where possible to be obtained from low-cost, sustainable resources (Arancibia et al., 2014b).
Food packaging has many purposes. It is designed not only to contain and protect food, but also to keep food safe and secure, to retain food quality and freshness, and to increase its

shelf-life. In addition, packaging should be affordable to consumers worldwide and, more importantly; it must be naturally biodegradable upon disposal (Imam&Glenn, 2012). Packaging films incorporated with antimicrobial substances, are of great potential for food preservation due to their antiseptic properties and conveniences for food (Huang et al., 2012). This kind of materials are considered as one of the most promising active packaging systems, as they are highly effective in killing or inhibiting spoilage and pathogenic microorganisms that contaminate food, and can limit the possible undesirable flavours that are caused by the direct addition of active compounds into foods.

Active packaging involves the interaction between the package, product and environment in order to prolong product shelf life or enhance its safety, while maintaining its nutritional quality. One class of active packages are antimicrobial packages used to reduce the growth rate and limit the maximum population of microorganisms. Antimicrobial packaging materials have to be in direct contact with the food surface if they are non-volatile and can be either immobilised on the materials surface or able to migrate into the food. Therefore, antimicrobial packaging is effective on food products were the microbial contamination occurs at the surface (Realini & Marcos, 2014).

CHITIN AND CHITOSAN POLYSACCHARIDES

Chitin was the first polysaccharide identified by man from mushrooms preceding cellulose by 30 years. After that this polymer was identified in the shells of insects and the exoskeletons of molluscs combined with minerals and proteins to harden these structures by cross-linking with polyphenols (Mati-Baouche et al., 2014).

The main sources of chitin (in % of dry matter) are crustaceans such as shrimp and crab (58-85%), insects (20-60%), molluscs (3-26%); cephalopods including squids, octopuses and annelids (20-28%), protozoans which contain a little chitin, coelenterates (3-30%), seaweed which can contain low quantities of chitin and fungi whose chitin contents are from trace to 45% (Fernandez-Saiz, 2011; Reddy et al., 2013; Mati-Baouche et al., 2014).

Chitosan is a deacetylated derivative of chitin (Arvanitoyannis, 2013), which is the second most abundant polysaccharide found in the nature after cellulose (Abdollahi et al., 2012; Kerry, 2012; Cruz-Romero et al., 2013; Leceta et al., 2013c; Nimesh, 2013; Nowzari et al., 2013; Ojijo & Ray, 2013) and commonly found in the shells and exoskeletons of crustaceans. Chitosan can also be obtained from the cell wall of some fungi (Fernandez-Saiz, 2011; Muzzarelli et al., 2012; Nitschke et al., 2011; Petrou et al., 2012; Wang et al., 2012; Leceta et al., 2013c; Sun et al., 2014), thus there is an alternative method of production that does not depend on environmentally harmful chemicals or the variable abundance of crustaceans.

Chitosan is mostly applied as a food additive or preservative, and as a component of packaging material, not only to retard microbial growth in food, but also to improve the quality and shelf-life of food (Vasilatos & Savvaidis, 2013). The solubility of chitosan depends on the degree of deacetylation, the distribution of acetyl groups along the main chain, the molecular weight and the nature of the acid used for protonation, but unlike chitin, it is soluble in dilute acid solutions below pH 6.0 due to the presence of amino groups. Apart from solubility, chitosan molecular weight can also affect the quality of the final film such as elasticity or brittleness (López-Carballo et al., 2012; Leceta et al., 2013). Chitin and chitosan offer a wide range of applications, including clarification and purification of water and beverages, applications in pharmaceuticals and cosmetics, as well as agricultural, food and biotechnological uses. Recent efforts for the use of chitin and chitosan have intensified since efficient utilization of marine biomass resources has become an environmental priority (Fernandez-Saiz, 2011).

Chitosan has been widely used in several industries and offers real potential for applications in food industry due to its natural origin and exceptional properties such as biodegradability, biocompatibility, biofunctionality, non-toxicity and chelation with metals (Abdollahi et al., 2012; Elsabee et al., 2012; Kanatt et al., 2012; Liu et al., 2012;

Cruz-Romero et al., 2013; Yu et al., 2013; Doulabi et al., 2013; Tanase & Spiridon, 2014) and antioxidant characteristics (Vargas et al., 2012; Peng et al., 2013; Schreiber et al., 2013; Cooksey, 2014). Chitosan is biodegradable and non-toxic and has some interesting biological activities, including excellent strength and elongation properties (Kaisangsri et al., 2012). Chitosan films have a selective permeability to gases (CO_2 and O_2) and good mechanical properties. However, due to its hydrophilic nature, it has poor barrier to moisture, which limits its uses (Lim et al., 2012; Pereda et al., 2012; Elsabee & Abdou, 2013).

Chitosan has been approved as food ingredient from FDA (Food and Drug Administration). In food products, chitosan offers a wide range of applications, e.g. preservation of food from microbial deterioration, formation of edible-biodegradable films, coagulation of proteins and lipids from waste water, enhancing of gelation in surimi and fishery products and clarification/deacidification of fruit juice (Zemljic et al., 2013).

ANTIMICROBIAL ACTIVITY OF CHITOSAN

The antimicrobial activity of chitosan depends not only on the external conditions (target microorganism, nature of the medium, pH temperature, etc.), but also on different intrinsic factors such as its molecular weight, and degree of polymerization and deacetylation (López-Carballo et al., 2012; Vargas et al., 2012). The antimicrobial action of chitosan is derived from the positive charge (Li et al., 2011) that amino groups present at acidic pH. Chitosan having cationic groups along the backbone has been shown to have antimicrobial properties against bacteria, yeasts, moulds and fungi (Lim et al., 2012; Siripatrawan & Noipha, 2012; Vasile et al., 2013; Yu et al., 2013; Van den Broek et al., 2014).

Chitosan has inherent antimicrobial activity owing to the fact that long positively charged chitosan molecules interact with negatively charged bacteria membrane causing disruption on the cell walls (Xiao et al., 2011; Leceta et al., 2013c). The antimicrobial action of chitosan is hypothesized to be mediated by the electrostatic forces between the protonated amino group (NH_2) in chitosan and the negative residues at cell surfaces. The number of protonated amino groups (NH_2) present in chitosan increases with increased degrees of deacetylation (DD) which influences the antimicrobial activity (Elsabee & Abdou, 2013).

Torlak and Sert (2013) studied the antibacterial effectiveness of chitosan-coated polypropylene films alone and incorporating ethanolic extract of propolis (EEP) against six foodborne pathogens (*Bacillus cereus*, *Cronobacter sakazakii*, *Escherichia coli* O157:H7, *Listeria monocytogenes*, *Salmonella typhimurium* and *Staphylococcus aureus*). The results obtained showed that antimicrobial activity was enhanced with addition of ethanolic extract of propolis (EEP) to film formulation. Also, the antibacterial efficacy of chitosan coated film was increased remarkably with the addition of propolis against both Gram-positive and Gram-negative bacteria tested, probably due to the synergistic effect between propolis and chitosan (Torlak and Sert, 2013).

Cruz-Romero et al. (2013) investigated the antimicrobial activity of low- and medium-molecular weight chitosan. The obtained results showed that both low molecular weight (LMW) and medium molecular weight (MMW) chitosan exhibited high antimicrobial activity against all bacterial cultures tested (Cruz-Romero et al., 2013).

POLYMER BLENDING

Polymer blending is one of the most effective methods to have new material with desired properties. Films formed by blending of polymers usually results in modified physical and mechanical properties compared to films made of individual components. Blending of synthetic polymers with chitosan, such as PVA (Bano et al., 2014), PET (Torres-Huerta et al., 2014) and starch (Liu et al., 2009; Kowalczyk et al., 2015), have been reported to improve mechanical properties of chitosan films. Blending of chitosan with other natural polymers gives films and coatings with good properties, chitosan addition in edible films leads to good film forming and mechanical properties, no toxicity, biodegradability, relative more hydrophobic nature that could provide higher moisture barrier and water resistance. Many researchers studied different properties of chitosan and chitosan blends (preparation, physical, mechanical, rheological, water vapour permeability and antimicrobial properties) their results show that chitosan and chitosan blends are extremely promising materials for bio-based active films preparation and that chitosan based edible films and coatings can be useful for preserving and extending the shelf life of foods (Elsabee & Abdou, 2013).

Chitosan nanoparticles have been successfully used as fillers to improve mechanical and barrier properties as well as the thermo-stability of films, decrease solubility and produce more compact and dense materials (Antoniou et al., 2014). In Table 1 some chitosan based materials and their reported characteristics are presented, stated in many studies.

Table 1. Chitosan based materials and their reported characteristics

Composition of chitosan based materials	**Reported effects**	**References**
Chitosan /PEO	increasing chitosan content in the blend solutions led to a significant reduction in nanofiber diameters - fact that is related to viscosity reduction and increased conductivity	Pakravan et al., 2011
PET/PP/chitosan	higher antimicrobial activity against *E. coli* and *B. subtilis* when chitosan was added	Lei et al., 2014
Chitosan/PVA/pectin	the formed films demonstrated antimicrobial activity against *E. coli*, *S. aureus*, *B. subtilis*, *Pseudomonas* and *C. albicans*	Tripathi et al., 2010
PLA/chitosan/keratin	improved Young's modulus; decreased tensile strength; significant increase in hardness compared to PLA	Tanase & Spiridon, 2014
PLA/starch/chitosan	improved hydrophilicity of the blends; the release procedure for chitosan could be divided into two stages: an initial fast stage and a following slow stage. The two stages exhibited the effectiveness and long residual action of antimicrobial property of the blends, and showed that the blend material was very suitable for foods with high water activity.	Bie et al., 2013
PEG /chitosan	higher antibacterial activity compared to native chitosan film	Davidovich-Pinhas et al., 2014
Gelatine/chitosan films	higher mechanical properties and thermal stabilities and lower water vapour permeability with the addition of chitosan	Liu et al., 2012
Cassava starch/chitosan/glycerol	the addition of chitosan increased mechanical resistance and decreased WVP of starch films	Pelissari et al., 2012
Chitosan/starch (modified with monomer 2-hydroxyethyl methacrylate)	modified films possess improved water stability, thermal stability than chitosan–starch film	Tuhin et al., 2012
TPS/chitosan/chitin	WVP was reduced with chitosan/chitin incorporation; TPS films with chitosan reduced *S. aureus* and *E. coli* growth in the contact zone	Lopez et al., 2014
Wheat gluten/chitosan	films presented lower moisture uptake and lower diffusivity than chitosan; once with the addition of chitosan higher toughness/extensibility and better microbial resistance was exhibited compared to wheat gluten	Chen et al., 2014
Chitosan-based /glycerol	functional properties of chitosan-based films were improved with the addition of glycerol; being adequate for packaging purposes	Leceta et al., 2013a
Chitosan/PVA with aqueous mint extract (ME)/pomegranate peel extract (PE)	antibacterial activity against Gram-positive bacteria (*S. aureus* and *B. cereus*) and ineffective against Gram-negative bacteria (*E. coli* and *P. fluorescens*)	Kanatt et al., 2012
PVA/ chitosan/nano-ZnO	increased antimicrobial activity when nano-ZnO was added	Wang et al., 2012
Chitosan-N-arginine (CS-N-Arg)	antibacterial activity against *S. aureus* and *E. coli* at the concentrations higher than 150 ppm; at the concentrations lower than 50 ppm, it might be digested by microbes and absorbed as nutrients to promote the growth of microorganisms	Xiao et al., 2011
Chitosan/cellulose/caffeic acid	superior antioxidant and antibacterial activity compared to the caffeic acid-free films	Yu et al., 2013
Chitosan–nanocellulose	excellent inhibitory effects against Gram-positive and Gram-negative bacteria	Dehnad et al., 2014
Tara gum/chitosan	better antimicrobial activity	Antoniou et al., 2014
PET/chitosan	successfuly inhibited *E. coli*, *L. monocytogenes*, and *C. albicans*	Zemljic et al., 2013

Chitosan/eugenol	improved thermal stability of eugenol; showed antioxidant activity and decreased water vapor permeability	Woranuch et al., 2013a; Woranuch & Yoksan, 2013b
Chitosan/clay-rosemary	exhibit antimicrobial properties and more phenol content	Abdollahi et al., 2012
Chitosan/ ferulic acid	improved thermal stability of ferulic acid; greater antioxidant activities compared to naked ferulic acid; improved water solubility	Woranuch et al., 2014; Woranuch & Yoksan, 2013c
Chitosan/ grape pomace extracts (aqueous extract)	antioxidant properties without modification of their water solubility and mechanical properties	Ferreira et al., 2014
Chitosan/ ferulic acid (FA) and ethyl ferulate (EF)	improved antioxidant properties	Aljawish et al., 2014
Chitosan/caffeic acid	modulated antioxidant and antimicrobial properties	Bozic et al., 2012
Chitosan/gallic acid	improved antimicrobial properties; increased TS; improved barrier properties	Sun et al., 2014
Chitosan/glycerol/olive oil	physical, mechanical and barrier proper-ties were strongly related with the chitosan concentration	Pereda et al., 2014
Chitosan/ carvacrol, grape seed extract	improved antimicrobial and antioxidant activity against *P. aeruginosa*, *S. aureus*, *L. innocua*, *E. faecalis*, *S. cerevisiae*	Rubilar et al., 2013
Chitosan /carvacrol	improved antimicrobial activity	Kurek et al., 2013
Gliadin-chitosonium acetate	excellent antimicrobial properties	Fernandez-Saiz et al., 2008
Gelatin-chitosan/oregano essential oil (OEO)	improved antimicrobial activity	Wu et al., 2014
Chitosan-based carbon dioxide (CO2) indicator	great potential to be used as sensors for monitoring the fermentation process and spoilage of foods	Jung et al., 2012

CHITOSAN BASED EDIBLE COATINGS

Edible coating is a thin layer of material formed as a coating on a food product, while an edible film is a preformed thin layer, made of edible material, which once formed can be placed on or between food components (Nowzari et al., 2013). Films prepared with these polymers are usually based on polysaccharides (e.g. chitosan), proteins (e.g. zein) and lipids (e.g. waxes), which are generally biodegradable, nontoxic, and some of them are effective barriers to oxygen and carbon dioxide, so they can be used as protective coating to maintain food quality and, at the same time, reduce the environmental impact of packaging wastes (Leceta et al., 2013a; Fabra et al., 2014). The main drawbacks of these types of materials are their inherently high rigidity and the difficulty of processing them in conventional equipment. A further disadvantage of proteins and polysaccharides is their very strong water sensitivity caused by their hydrophilic character (Fabra et al., 2014). Interest in edible coatings and films on highly perishable unmodified and/or fresh foods has intensified in recent years.

The use of edible films and coatings has several aims, most importantly: the restriction of moisture loss, control of gas permeability, control of microbial activity (e.g., chitosan, which acts against microbes), preservation of the structural integrity of the product and the gradual release of enrobed flavours or antioxidants into the food (Fabra et al., 2014).

Table 2 presents a series of researches performed using chitosan based coatings and solutions in food products applications.

Table 2. Chitosan based films and solutions and their reported effects on food products

Composition of chitosan based materials	**Food product**	**Reported effects**	**References**
Chitosan/ PVA	minimally processed tomato	a viable alternative in shelf-life extension of minimally processed tomato	Tripathi et al., 2009
chitosan/antibrowning agents	fresh-cut lotus root	lower respiration created by chitosan coating	Xing et al., 2010
Carvacrol/ chitosan	green bean	strong antimicrobial activity against *E. coli* O157:H7 and *S. Typhimurium*	Severino et al., 2015
Chitosan/GA	peanut powder	reduce lipid oxidation	Schreiber et al., 2013
Chitosan/whey protein	Ricotta cheese	reduced growth of microbial contaminants and extended the shelf-life of the product; delayed	Di Pierro et al., 2011

		development of undesirable acidity; better maintained the texture and did not seem to modify sensory characteristics	
gelatin-chitosan/oregano essential oil (OEO)	grass carp muscle	Both TPC and TVB-N were much lower than control samples; incorporating OEO can extend the shelf-life of fish muscle	Wu et al., 2014
chitosan films (CS)	fresh fillets of hake (Merluccius merluccius) and sole (Solea solea)	decreased final bacterial population; the CS-AP(in air) extended the shelf-life of hake and sole fillets by 7 to 9 days	Fernández-Saiz et al., 2013
Chitosan–gelatin	rainbow trout (Oncorhynchus mykiss) fillets	chitosan–gelatin coating and film retained their good quality characteristics and extend the shelf life of fish samples during refrigerated storage; showed antioxidant effect	Nowzari et al., 2013
Chitosan film	fresh swordfish steaks	the combined use of chitosan and VP could inhibit bacterial growth; retard evolution of amines TMA-N and TVB-N; maintain or improve the sensory quality of fresh swordfish stored under refrigeration (4 °C)	Tsiligianni et al., 2012
Chitosan films	fish soup	proved to have biocidal effect without affecting the sensorial properties of the commercial product	Loâpez-Rubio et al., 2011
Chitosan/cyclodextrin incorporating carvacrol	packaging of fresh chicken	antimicrobial effect depending on the size of the film and storage time; increased concentration of carvacrol affected sensorial attributes of chicken meat	Higueras et al., 2014
Chitosan solution	aerobically packaged chicken meat	shelf life extension of 6 to 7 days; shelf life extension using the combination chitosan dip plus MAP was 9 days	Latou et al., 2014
Chitosan, oregano and their combination	modified atmosphere packaged chicken breast meat	this combination could inhibit growth of microbial spoilage flora; retard lipid oxidation; maintain lightness; improve the sensory quality of fresh chicken meat	Petrou et al., 2012
Chitosan solution/ pure thymol oil	Ready-to-cook chicken and pepper kabobs	extended shelf life to 14 days; significantly reduced microbial counts throughout the storage period	Cooksey, 2014
Chitosan/rosemary EO	turkey meat	under VP conditions could inhibit growth of microbial spoilage flora; improve the sensory quality of fresh turkey meat stored under refrigeration (2 °C)	Vasilatos & Savvaidis, 2013
(LY-CS-OREC/ALG) 10.5 film-coated CA	fresh pork meat	best antibacterial activity; could extend the shelf life of fresh pork for about 3 days;	Huang et al., 2012
Chitosan/green tea (CGT-film)	pork sausages	maintain qualities and extend shelf life of the refrigerated pork sausages	Siripatrawan & Noipha, 2012
Chitosan film	ground beef	reduced lipid oxidation; improved surface red color of beef patties	Cooksey, 2014; Suman et al., 2010

Chitosan has proven its antimicrobial activity when used as a coating on several food products, cheese, fish patties, strawberries or bologna. In other cases, the role of chitosan is that of a matrix for the delivery of other antimicrobial compounds such as acids, salts, cinnamaldehyde, lysozyme, nisin or plant extracts (López-Carballo et al., 2012). Also, chitosan coatings have been successfully probed at an experimental level on food such as eggs, fruits, vegetables, dairy products and meats. However, for certain food products, the limited antimicrobial activity of pure chitosan films does not reach the antiseptic level desired by packers (Sun et al., 2014).

CONCLUSIONS

The potential of chitosan, as shown by research and development efforts, is supported and enhanced by both the increasing consumer demand for natural and safer additives with functional properties, and increasing environmental concerns. It is likely that some of the future applications of chitosan in the food area will come from the medical and pharmaceutical sectors, where it has been extensively used because of its antioxidant and antimicrobial properties. Potentially, chitosan could be incorporated into recycled materials such as multilayer plastic packaging materials in which each layer of polymer would have a specific function: i.e., chitosan could act as antimicrobial/antioxidant agent and another material could act as a water vapour barrier. Moreover, since chitosan properties are much affected by the pH and the ionic strength of the surrounding medium, chitosan-based systems could also be used for the controlled release of active ingredients (Vargas et al., 2012).

The information presented in this review demonstrated the favourable effects of chitosan alone or in combination with different polymers, essential oils and other agents. Many researchers demonstrated that chitosan treatment can offer protection against contamination and microbial spoilage (Van den Broek et al., 2014).

Chitosan coatings were also proved to be beneficial in maintaining a higher product quality during the storage period, and an increased shelf-life of food products was observed.

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