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Review paper

CHITOSAN APPLICATION IN FOOD TECHNOLOGY: A REVIEW OF RECENT ADVANCES

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ABSTRACT

The review focused on global trends in the development of scientific research and the practical applications of chitosan in food technology in recent years. Chitin and its derivative chitosan obtaining from the crustacean shells and the cell wall of fungi are among the most common biopolymers in the world. Chitosan is a polysaccharide discerned by a large number of unsubstituted amino groups. Featured properties of chitosan providing its high chemical and biological activities. Chitosan has various abilities as polycationite, film former, antimicrobial and antioxidant agent. Multifunctional properties open up broad prospects for the chitosan applications in various fields of technology, medicine and industry. The most attention in the review is paid to the works on extending food products shelf life with chitosan based primary edible film coatings and biodegradable packaging. At the same time chitosan applications as an emulsifier, a flocculant, as well as functional food additive, nutrient encapsulating material and dietary supplement are highlighted.

1. Introduction

Chitosan — poly-β-(1,4)-d-glucosamine is a deacetylated product of chitin — poly-β-(1,4)-N-acetyl-d-glucosamine, one of the most abundant natural polysaccharide, found in composition of crustacean shells and fungal cell walls [1]. There is great scientific and practical interest in chitin and chitosan caused to the unique properties as biocompatibility, safety, biodegradability, sorption performance of heavy metals and radionuclides [2]. Scientific publications amount related to chitin and chitosan is increasing annually and several scientific papers related to its production and application are published weekly according to the Web of Science and Google Scholar. Chitin global production in 2015 was 28 thousand tons, and its still increasing [3]. Due to its properties, chitosan has been widely used in various fields of technology, medicine and food industry. Chitosan properties are significantly influenced by its molecular weight (MW), which occurs in a range from 2 to more than 200 kDa. Deacetylation degree (DD) is other important chitosan property affecting an acid solubility which differs it from chitin. Chitosan with DD more than 55% becomes soluble in a 1% acetic or hydrochloric acids [4]. Solubility associated with the protonation of the amino group at the C-2 position of the d-glucosamine unit converts the polysaccharide to a polyelectrolyte under acidic conditions [5].

The purpose of present work is the development progress problem study on food technology chitosan applications and analysis of these trends.

2. Main part

A biological activity of chitosan along with safety for the human brings its wide application for food industry. Antioxidant and antimicrobial activity of chitosan allows to use it for increasing the foods shelf life, excellent emulsifying properties gives an opportunity to replace synthetic surfactants in food technologies, also chitosan can be used as a functional ingredient against hypercholesterolemia, hypertension and inflammations, and for the encapsulation of nutrients for a functional food development [6]. Polyelectrolyte and flocculant behavior of chitosan allow to use it as an effective clarifying agent for beverages, as well as for preliminary purification of drinking water [7].

Chitosan applications for food shelf life prolongation

Most significant factors limiting the shelf life of food products are an oxidative and microbial spoilage. Antioxidant activity of chitosan due to scavenging effect on free radicals. This ability depends on DD and increases with unsubstituted amino groups amount increased. High MW chitosan exhibits a less pronounced antioxidant effect, because its structure is more compact due to the formation of strong intramolecular hydrogen bonds [6]. The antioxidant activities of chitosan in lard and rapeseed oil is comparable to ascorbic acid [8]. Also an antioxidant effect of chitosan in semi-finished products from meat, fish and seafood is noted [9].

The antimicrobial and antifungal activity of chitosan caused by cationite properties appeared in an acidic media. At pH values below pK_a of chitosan protonated amino groups binding negatively charged carboxyl groups of lipopolysaccharides and peptidoglycanes at the bacterial cell wall surface, leading to permeabilization and destruction of external membranes [10]. The mechanism of antifungal action of chitosan is similar to antibacterial and appears to fungi containing a significant amount of polyunsaturated fatty acids [11]. Processing with chitosan solutions of fresh or chopped fruits, berries and vegetables extends their shelf life, inhibits the development of microorganisms and molds. Chitosan addition inhibits bacterial growth in noodle dough, hummus, and cheese [9,12].

Edible film coatings are the most promising chitosan application for food shelf life extending. Edible films are defined as the primary packaging designed to protect foods against external impacts, also it can be used as carrier of active ingredients such as preservatives, flavors and colorants. [13]. Chitosan film coatings are prepared as a solution in dilute acids. Depending on DD, solution concentration, additional components and the drying conditions, there is a wide variety to change physicochemical and biological properties of the resulting compound [14]. Edible coatings reducing weight loss during storage, prevent oxygen permeability, shows antimicrobial and antioxidant activities to, increase the shelf life of processed foods significantly.

The tensile strength of chitosan films produced with acetic acid is higher in comparison with citric, lactic, and malic acids and increases during long-term storage [15]. Elasticity of the coatings could be increased by addition of glycerol or sorbitol as plasticizer. Chitosan-based film coatings effectively extend the shelf life of carrots [16, 17], tomatoes [18], cucumbers [19], bell peppers [20], bananas [21], apples [22], pomegranates [23], pears [24], plums [25], strawberries [26], papaya [27], mango [28], fish [29], meat [30] and minced meat [31, 32].

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A number of studies concerning coatings composed of chitosan blends with other film-forming biopolymers allowed to keep biodegradability, non-toxicity, compatibility with food products and improve functional properties. Chitosan-starch blend films shows good water vapor permeability and a high antioxidant activity [33,34]. The addition of cellulose and its derivatives to chitosan films increases elasticity, tensile strength, and optical transparency while maintaining high antimicrobial activity [35,36]. Chitosan-alginate multilayer coatings applied to fresh cut fruits allows to save succulence during a longer while [37,38]. Pectin and chitosan combination occurring a strong intermolecular electrostatic interaction, and the films obtained are characterized by high transparency, strength, and elasticity [39]. Compositions of chitosan with proteins, such as casein and gelatin [40,41,42], are characterized by good mechanical properties, low vapor permeability, and UV-protection due to absorption by peptide bonds [43]. Soy protein or beeswax included to coating composition increasing film hydrophobicity [44,45].

Because of active functional groups chitosan can be used as part of composite materials containing not natural only but synthetic polymer compounds [46]. Opuntia mucilage — a complex mixture of functional polysaccharide residues used as film-forming agent with polyvinyl alcohol and chitosan obtaining edible coatings with different properties when the composition is varied. These coatings are hydrophilic due to the opuntia mucilage. Inclusion of chitosan has technological approach because it is able to stabilize the film-forming solution increasing zeta potential and also a functional since films with higher content of chitosan became tougher. When increasing polyvinyl alcohol amount film flexibility is increased. That make those mixtures compatible with different food applications required [47]. Films formed by chitosan and polyvinyl alcohol with lignin nanoparticles are characterized by increased strength compared to films formed by individual components, antibacterial action against gram-negative microorganisms and the synergistic antioxidant effect of chitosan and lignin [48].

Natural plant extracts included with the chitosan coating composition has a significant positive effect on the mechanical and biological properties of the protective edible films formed. For example, polyphenols of thyme extract increase tensile strength of chitosan-based films as crosslinkers, and also increase the antioxidant activity [49], turmeric extract addition reduces the UV-permeability, significantly increases the tensile strength and antimicrobial activity [50], milk thistle extract reduces films solubility and water vapor permeability, as well as improves antioxidant properties [51]. Barberry seed extract increases the hydrophobicity of coatings, while whole fruit extract reduces it compared to a control sample. The antimicrobial activity of chitosan films with barberry extracts based quorum sence inhibitory property [52]. Ethanolic extracts of rosemary, cinnamon, guarana and peumus added to chitosan and gelatin based films allows to achieve very high antioxidant and antibacterial activities [53], grapefruit seed extract increases the antifungal activity, elasticity and elongation at break of chitosan films [54].

A significant synergistic antimicrobial effect is observed for composite coatings based on the chitosan matrix including nanoparticles of inorganic antimicrobial agents such as silver [55,56,57] and zinc oxide [58,59,60].

Essential oils addition to film-forming composition gives for the edible films supplemental functional properties. Essential oils increasing antioxidant ability and activity against molds and microorganisms and also changing physical properties of the chitosan films, such as strength, moisture resistance and transparency. That coatings can be used for berries, fruits, fish or meat. Combined action of essential oils and chitosan may lead to synergistic effects. Rosemary oil containing chitosan coating on rainbow trout fillets exhibits more pronounced antibacterial and antioxidant effect than these components separately [61]. Chitosan coating containing eucalyptus essential oil and α-tocopherol prolongs the shelf life and reduces fat oxidation of fresh silver carp, which is more related to the combined presence of chitosan and eucalyptus oil than α-tocopherol used as an regular antioxidant food additive [62]. Cinnamon and clove essential oils enriched chitosan-arabic gum films enhances moisture resistance as well as antimicrobial activity against Escherichia coli and Staphylococcus aureus more in comparison with the inclusion of these oils by itself [63]. In chitosan-carboxymethylcellulose edible coating cinnamon oil exhibits better antifungal activity add reduces the water vapor permeability by increasing the hydrophobicity than ginger oil but in conjunction emerging a significant synergistic antifungal and moisture resistance effect [64]. Addition of cinnamon and ginger essential oils slightly reduces the transparency of chitosan films, but allows to obtain a thinner coatings. That films effectively prevent caused by fat oxidation and the action of microorganisms fresh pork spoilage [65]. In some cases, essential oils addition to the coating may not have any effect or even negatively affecting on the resulting products. The synergistic antifungal effect on *Penicillium digitatum*, the main cause of citrus green mold when cinnamon essential oil incorporated chitosan coating, showed in vitro was not confirmed when applied to the mandarins [66]. Oregano and basil essential oils reduce tensile strength, while increasing transparency and improving the antimicrobial and antioxidant properties of chitosan-based films [67]. Thyme oil with chitosan coating reduces moisture loss, retains color and inhibits yeast growth, but does not affect on enterobacteria, mesophilic and lactic acid bacteria for ham [68]. Cumin and eucalyptus essential oils as part of the chitosan coating of chicken breasts reducing spoilage caused by various types of microorganisms, extending shelf life and enhancing, the taste and aroma of fried chicken [69]. Lemon oil compared to oregano oil in the chitosan coating composition shows a more pronounced bactericidal effect, both have not adversely affect chilled chicken breast organoleptic [70]. However, lemon oil added with a strawberry coating despite an antifungal resistance increasing negatively changing a typical aroma due to terpenes contained in lemon oil [71].

A promising direction of food quality monitoring is visual edible film colorimetric indicators that change color when the pH accompanying spoilage changes. That coatings were developed for fresh fish on the chitosan-starch base containing red cabbage extract [72], and as polyvinyl alcohol films including chitosan nanoparticles and mulberry extract [73].

Development of biodegradable polymer packaging materials is one of modern trends to extend food shelf life. A microcrystalline cellulose-polyvinyl alcohol-chitosan composition showed two orders of magnitude higher water vapor permeability with comparable strength to polyethylene films and relative elongation exceeds cellophane because of plasticizing effect of polyvinyl alcohol [74]. Polylactic acid with chitosan nanoparticles using polyvinyl alcohol as a plasticizer and polyethylene glycol as crosslinking agent is characterized by a significant increase in tensile strength and antimicrobial activity against aerobic microorganisms [75]. Amphiphilic nanoparticles formed by a chitosan grafted polylactic acid oligomers used as a filler for polylactic acid makes resulting material more flexible and significantly increase the oxygen barrier properties [76].

Chitosan emulsifying applications

Chitosan is an excellent emulsifier to stabilize heterogeneous oil-in-water systems. Chitosan increases viscosity of the continuous phase, complicating the diffusion of disperced particles and reducing the droplet aggregation rate. In addition, positively charged amino groups, making chitosan an amphiphilic surfactant polymer.

Chitosan can be used as a single emulsifier, however the emulsions obtained are reversible due to the sensitivity to the pH values. In an acidic media with pH < pK_a protonated chitosan exhibits surface-active properties, interacting with triglycerides carbonyl groups. With pH increasing, the solubility of chitosan decreases, while the emulsion remains stable because of oil phase droplets adsorbed on chitosan particles. If acidity increases back chitosan passes into a soluble form desorbing a disperced oil droplets and the emulsion is reversibly separating [77]. The emulsifying ability of chitosan largely depends on the DD and MW and increased for low MW chitosan with DD less than 60% or more than 86%, whereas when the values of DD from 65% to 77%, that properties are significantly dependent on concentration [78]. Most of the practical studies on chitosan applications as an emulsifier consider it as part of complex compositions containing other surfactants. Presence both chitosan and protein emulsifiers makes a heterogeneous system more stable. Incorporation of chitosan with soybean isolate protein can increase the digestibility and stability of emulsified carotenoids [79]. The complex containing chitosan with modified β-lactoglobulin fibers stabilizes the fish oil emulsion in water [80]. Also, stable Pickering emulsions with corn oil are formed as a result of electrostatic interaction of chitosan with gelatin [81]. The ability of chitosan to form micelles in aqueous solutions may be used for unstable compounds as carotenoids [82] and anthocyanins [83] sustainability increasing.

Applications of chitosan as a flocculant

Polycationite chitosan properties can be used for drinks clarification from solid suspended particles comprising polyphenols, proteins, polysaccharides and mineral compounds [5]. Chitosan can be effectively used to clarify fruit juices [84–87], fruit [88] and grape [89] wines, beer [90] and also at the preliminary drinking water purification since this increases suspended particles sedimentation rate and increases the degree of separation of bacteria and viruses [91].

Chitosan application in the functional foods production

Chitosan may be used as a carrier for encapsulating and controlled delivery of probiotic ingredients in functional food products beacuse of its high biocompatibility, emulsifying ability, polycationicity and hypoallergenic properties [92]. Components encapsulation used to prevent chemical changes during the preparation process or stomach acidic media, for lipophilic components as vitamins or flavonoids with polymer matrix delivery, to control nutraceuticals release and to mask smell and taste of some substances. The most common method of nutraceutics encapsulation is ion gelation [6] using polyanionites — tripolyphosphate or proteins as gelling agents. Particle size depends on chitosan properties and increases with a low DD and MW associated with a decrease in the zeta potential, determined by unsaturated amino groups of the macromolecule interacting with polyanionite [93]. Can be obtained nanoparticles containing vitamins C [94,95] and B2 [96], dyes as curcumin [97] and lutein [98], flavonoids, including catechins [99,100,101] and quercetin [102], bioactive minerals selenium [103,104] and zinc [105].

A promising direction of nanoencapsulation of biologically active substances is chitosan-based multilayer composite coatings development. In this way is possible to obtain orally digestible insulin [106], and keep stable vitamin D3 in food products for up to 60 days [107].

Whell known that chitosan itself exhibits biological activity. It binds fats, cholesterol and bile salt due to both hydrophobic interactions and the formation of hydrogen bonds, and as a result of electrostatic attraction of positively charged amino groups and negatively charged carboxyl groups of fatty acids.

The binding of cholesterol to chitosan was confirmed by a number of clinical studies, so chitosan may be used in the hypercholesterolemia treatment [108, 109] and included in the formulations of special foods with an anticholesterol effect. The fat binding ability of chitosan can be used in the production of dietary foods reducing the rate of lipids absorption and digestion [110]. Moreover, a composite film coating containing chitosan, gelatin and gallic acid can be used to simulate fat in food products, acting as a fat substitute by imparting organoleptic surface properties [111].

Chitooligosaccharides have antitumor activity. Despite the large number of clinical studies, antitumor mechanism still not well understood, however, the most important factors in the action of chitosan are cancer cells viability reducing associated with the electrostatic interaction between chitosan and cell wall glycoproteins, inhibition of angiogenesis, which is the main reason for the rapid growth of cancer tumors, also a complex immunostimulating effect is possible [112].

3. Conclusion

A global trends analysis in the research of chitosan in food technology applications shows that the most attention to this problem is paid in Asian countries that have an abundant raw material base for its production. A large number of developments are currently proceeded in China, unlike Japan which lost its past leadership in this direction. Widespread research is also carries out in India, Spain and Iran. It should be noted that in Russia, unlike the 90s, the amount of studies on the issue has significantly decreased.

In recent years most intensive researches focused on the use of chitosan applications to extend the shelf life of foods, and also as an emulsifier, flocculant and dietary supplement. Today the explosively developing area of chitosan applying is the edible films producing. Edible coatings perform not only the function of food primary packaging, but also prevent moisture loss and oxygen access, have antimicrobial and antioxidant activity due to the unique properties of chitosan. At the same time is possible to enrich the products with additional functional ingredients as natural plant extracts, essential oils, vitamins and antimicrobial components. Thoset components enhancing the properties of the films obtained up to reach a synergistic effect over prolonging the foods shelf life. The latest trends of chitosan application are associated with freshness indicating coatings development, with vitamins, nutrients and probiotics encapsulation and delivery, and also with the biodegradable packaging engineering.

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