

Chitosan and its Derivatives: A Step Towards Green Chemistry

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Abstract: Increasing attention is now being paid to applying green chemistry compounds, i.e., compounds that cause minimum environmental damage. Biopolymers have a leading place in this issue, and chitosan and its derivatives are leading among them. The unique structure of chitosan allows it to exhibit a set of properties such as biodegradability, antimicrobial activity, non-toxicity, biocompatibility, and others, due to which it has found wide application in various fields. Modifications of its structure by alkylation, esterification, graft copolymerization, acylation, and other types of modification and new technologies have greatly expanded the possible applications of chitosan and its derivatives. The important aspect is also that the technologies used for modification can be attributed to green chemistry, and the modification products are eco-friendly. This article provides an overview of recent trends in preparing chitosan derivatives and their use in light of environmentally friendly technologies.

Keywords: chitosan; chitosan derivatives; green chemistry; eco-friendly materials.

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1. Introduction

The current state of the environment under stress from non-degradable pollutants and the search for new and more efficient compounds based on the principles of green chemistry led to the widespread use of biopolymers [1]. The use of polysaccharide chitosan (Ch) and its derivatives (ChD) has a leading place in this issue among biopolymers due to the unique properties of Ch. The number of articles, reviews, and books on the study and application of Ch in various fields is growing rapidly. Our previous work concerned the preparation of some chitosan derivatives and the study of their properties [2,3]. In this review, we have focused on the latest publications on the preparation of ChD, their properties, and the applications of these biopolymers.

The growing interest in the study of this substance is related to the structure of Ch, which allows it to demonstrate unique physical and chemical properties [4]. Biopolymer Ch is deacetylated derivative of chitin, which is the second most abundant polysaccharide found in nature after cellulose. Being a straight-chain copolymer composed of D-glucosamine and N-acetyl-D-glucosamine, it is structurally similar to cellulose, which is composed of only one monomer of glucose. It is obtained by the partial deacetylation of chitin (Figure 1) and commercially produced from crustacean shells as well as other sources like mushrooms. The

issue of green synthesis of Ch is being discussed today since the recovering of chitosan from seashell waste leads to the formation of toxic waste [5].

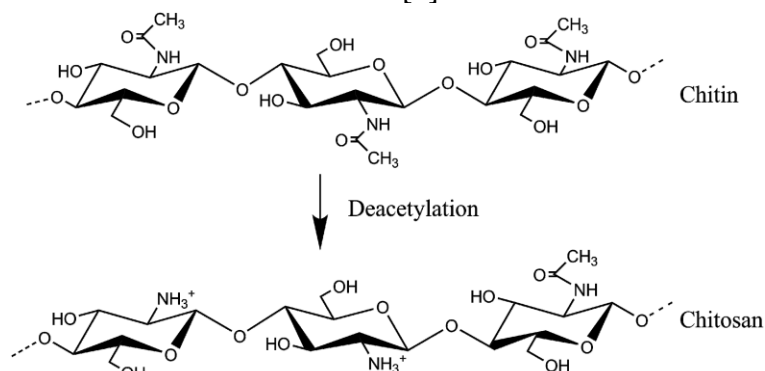


Figure 1. Structural formules of chitin and chitosan.

Being the only polycation of natural origin, it has unique properties such as high bioavailability, biodegradability, biocompatibility, non-toxicity, antibacterial activity, ability to adsorb metals, and low cost, which makes it an attractive material for future technologies [6-8]. Furthermore, the study of Ch aligns with the principles of green chemistry, which prefers natural materials over synthetic substances [9].

However, despite all the above properties, Ch has poor solubility, low surface area, and such rheological characteristics that are the main limiting factors for its application. Preparation of ChD with improved properties as well as combining them with other substances often helps overcome these limitations [10-12]. Thus, hydrophobic modified Ch and Ch succinate derivatives have been found to have high surface activity and to effectively stabilize "oil in water" type emulsions [3]. Numerous examples of the manifestation of the improved characteristics of the ChD and ChD composites for various purposes are given in this article [9].

Ch and ChD can exist in many forms, such as fibers, gels, films, nanoparticles, and granules [13,14]. Particular interest is being given to the study of Ch nanoforms and the nano compounds that are part of Ch composites.

Ch is insoluble at neutral and alkaline pH but can form water-soluble salts with inorganic and some organic acids. Upon dissolution in acidic media, the amino groups of the polymer become protonated, giving the molecule a positive charge. With the action of glycosidases such as chitosanase, chitinase, lysozyme, and cellulase Ch is found to be biodegradable. Chitosan is mainly characterized by its molecular weight (Mw) and degree of deacetylation (DD). Low Mw Ch is more soluble, but Ch samples with higher Mw are only soluble in acidic aqueous media, even at high DD. Insufficient solubility at neutral and basic pH inhibits the use of Ch in some biomedical applications (at pH 7.4). This determines a variety of work on the synthesis of ChD that increases the solubility of Ch [15]. On the other hand, for some applications (e.g., as rheological modifiers, selective sorbents, and stabilizers for various compositions), it is necessary to increase the hydrophobicity of Ch and synthesize its hydrophobic derivatives [16].

2. Preparation of Chitosan Derivatives

Ch has three main types of reactive functional groups, an amino/acetamido group as well as both primary and secondary hydroxyl groups at the C-2, C-3, and C-6 positions. The highly reactive amino group (-NH₂) makes the DD a characteristic ChD preparation and

characterization property. Deacetylation also affects the biodegradability and immunological activity of Ch. The main types of reactions for preparing ChD are shown in Figure 2.

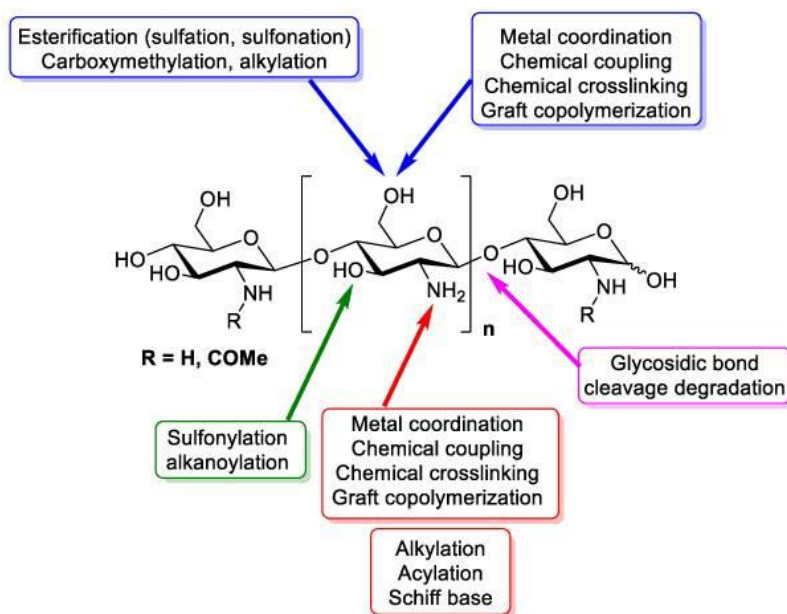


Figure 2. Types of reactions for the preparation of chitosan derivatives [15].

The Ch molecule's structure determines various modifications: via covalent conjugation and non-covalent interactions [6]. In non-covalent interactions, in addition to hydrogen bonds, there are electrostatic and hydrophobic interactions. Changes in these interactions will affect the conformational states of Ch macromolecules in solutions, the yield, and the properties of the copolymer [17]. Review and special articles are devoted to the issue of the formation of covalent ChD, e.g., the review [18] presents the broad range of functionalization strategies reported over the last five years to reflect the state-of-the-art of Ch derivatization. The preparation of ChD is intended either to improve the properties of Ch (solubility, biodegradability) or to introduce new functions or properties [19]. Thus, solubility may be improved in an aqueous media by deacetylation, depolymerization, or quaternization, among other processes [20]. The introduction of quaternary ammonium groups into Ch (so-called Ch quaternization) represents an approach to increasing both the solubility and the density of the positive charge of Ch. Amino-functionalized Ch attracts much attention due to its significant bioactivity [21]. The effect of bioactive glyoxylate bearing Schiff base, which gives antifungal and antioxidant activities to Ch quaternary ammonium salts, was shown in [22]. Novel green in situ quaternary-ammonium-functionalized magnetic Ch microspheres potentially applicable in wastewater treatment were synthesized [23]. The new approach to producing Ag nanoparticles via click chemistry using quaternized Ch is presented in [24]. A new type of cationic molecular ‘multi-subtype’ surfactant was synthesized based on Ch (via the ring-opening reaction and quaternization), which exhibits excellent surface activity and emulsifying ability [25]. Synthetic procedures and properties of quaternary ammonium salts of Ch were systematized in the review [26].

Grafted ChD can be formed by acylation, alkylation, hydroxyalkylation, carboxyalkylation, quaternization, thiolation, phosphorylation, sulfation, and graft copolymerization. The properties of the grafted copolymers obtained are controlled by the characteristics of the side chains and their quantity. The compatibility between Ch and bio-based polyesters was improved by grafting maleic anhydride and glycidyl methacrylate onto

polymer chains using peroxide [27]. The synthesis and characterization of poly(trimethylene carbonate) grafted Ch as a new water-soluble biopolymer with improved biocompatibility in solution, moderate wettability, and high biodegradability rate in the solid state is described in [28]. Optimization of grafting parameters was observed in copolymerization with styrene using ammonium persulfate as initiator, where the grafted polymer showed better antimicrobial activity [29]. Biocompatible film materials based on alkylated Ch and grafted copolymer of starch with acrylamide have been obtained by watering from a homogeneous solution of modified polysaccharides. It was found that the resulting composition could have promising applications as packaging materials and products for medical and biological purposes [30]. Tweezer-like adsorbents with enhanced surface area were synthesized by grafting aniline onto the amine sites of a Ch biopolymer scaffold [31].

The formation of substances with surface-active properties occupies a special place in the study of ChD. This can be the synthesis of amphoteric derivatives or the production of composites based on Ch [32]. Many pharmaceutical, cosmetic, food, and chemical products exist in the form of emulsions. A common problem with emulsions is their instability. Interaction between polymer and surfactant could change the adsorption layer around the emulsion's oil droplets, affecting their stability [33-36]. As a cationic polyelectrolyte, Ch is prone to interact with anionic surfactants through electrostatic attraction as well as hydrophobic interaction. The study of the effect of sodium dodecyl sulfate (SDS) content on the structure and properties of Ch films showed that SDS binding to Ch was realized through interactions between $-\text{SO}_4^{2-}$ and $-\text{NH}_3^+$ groups, forming an ionic cross-linking film, which can be regulated by SDS adding [37]. It is known that the $-\text{OH}$ and $-\text{NH}_2$ groups on the Ch skeleton can serve as good ligands for coordination with transition metal ions. Modification of Ch via different methods may improve adsorption efficiency not only of metal ions but also of organic pollutants [38].

The dispersed state of Ch in the systems is extremely important. There is increasing interest in Ch-based nanomaterials and their composites with other substances [39]. Increasing porosity of Ch-sorbents leads to a significant increase in their catalytic properties [40]. Nanoparticles can be incorporated into nanofibers forming Ch nano-nanosystems that can be used in oncology treatment [41]. There is information on a new core-shell composite prepared by in situ growth of MgFe–Cl-layered double hydroxides plates on modified Ch wrinkled microspheres with a larger specific surface area, which was applied for the removal of nitrate from water [42]. In vitro antibacterial activity of nanocomposites (against *Staphylococcus aureus* and *Escherichia coli*) was enhanced by a modified method, resulting in greater mobility of radiation-modified Ch in the network [43]. An example of a new approach to forming multilayer thin films of fatty acids/polysaccharides based on the self-assembly of components into vesicular systems in an aqueous solution is presented in [44]. These films may be used in the fields of biocatalysis, biosensors, and drug delivery.

3. Application of Chitosan and its Derivatives

The potential of chitosan, as noted in numerous studies, is enhanced both by the growing consumer demand for natural and safe materials and additives with functional properties and by growing environmental concerns.

3.1. Application of chitosan and its derivatives in the food industry.

Due to its properties, chitosan is used as a food-packaging material, emulsifier, thickening and stabilizing agent, flocculant, agent for color stabilization, food mimetic, chemical preservative, natural flavor extender, functional food additive, as well as encapsulating material.

3.1.1. Food-packaging material.

The traditional methods of food preservation (freezing, fermentation, drying, and others) are being replaced by new ones. One of them is food packaging, which acts as a barrier against undesirable physicochemical and biological impacts [45-49].

The use of biopolymers obtained from natural sources (starch composites, collagen, gelatin, cellulose, etc.) instead of synthetic polymers (polyethylene, poly(ethylene terephthalate), etc.) is healthier and does not have a negative impact on packaging materials on the environment. Taking into account all the above properties of Ch as well as its film-forming ability, it can be proposed as a promising material for use in the food industry, for example, in the form of Ch films, especially edible Ch films [50-53].

Despite the above advantages of Ch, it has poor moisture barrier properties due to its hydrophilic nature and, therefore, poor mechanical properties in wet environments that limit its uses in the packaging industry. Ch-based composites help overcome these disadvantages [54-56]. The flexibility of the Ch film can be improved by adding plasticizers due to the disruption of the hydrogen bond between biopolymer molecules. However, the mechanical strength of Ch film, in this case, decreases. Thus, simultaneously, there is a need to improve both the strength and flexibility of Ch films [37]. One of the most effective methods for that is polymer blending. Thus, Ch-based films with incorporated thyme essential oil (to increase flexibility) and various combinations of cross-linkers (ZnO, CaCl₂, nano-clay, and PEG) were found to be an alternative to solve this issue [57]. Ch films with graphene, titanium dioxide, and zinc oxide may be considered safe, non-cytotoxic packaging materials in the future [58]. A new encapsulation method is proposed to protect unstable biologically active ingredients from unfavorable conditions. Ch capsules have proven to be suitable carriers for their delivery to target sites [59].

Ch nanoparticles and their combinations with other nanomaterials have been successfully used as fillers to improve mechanical and barrier properties and the thermostability of films, decrease solubility and produce more compact and dense materials with enhanced antimicrobial properties [60-67]. For instance, a novel nanocomposite antibacterial agent based on Ch nanoparticles and antimicrobial peptides was applied as an effective antimicrobial agent with lower cytotoxicity in the food industry to prevent bacterial contamination [68]. In addition, the release of Ch nanoparticles during the prolonged storage period of food positively affects the quality of products [69].

3.1.2. Emulsifier and emulsion stabilizer.

Ch and its amphoteric derivatives may act as emulsifiers and emulsion stabilizers through the adsorption of the protective layer at oil-water interfaces [70,71]. That depends on the nature and concentration of Ch, pH, and ion strength of the solution [72]. The use of hydrophobically modified Ch (ionic complex of chitosan with hexadecyl-oligo-oxyethylene hemisuccinate) [3] or Ch-SDS complexes [2], as well as the combined use of Ch and other food

components: e.g., corn fiber gum [73], is an efficient method to enhance the Ch emulsifying properties. It is shown that a mixture of thyme oil with a surfactant introduced into the Ch matrix leads to a significant reduction in particle size in film-forming solutions and an increase in the barrier properties of Ch films [74]. The effect of the sequence and concentration of Ch addition on the emulsifying properties of egg yolk hydrolysates is considered in [75]. The combination of Ch with whey protein gives the Ch complex that may work to stabilize squalene in water emulsions and can find potential application in the formation of stable emulsion systems for encapsulating highly unsaturated oils [76]. The effectiveness of Ch and ChD as an alternative to synthetic surfactants for polylactide microparticle stabilization was studied in [77]. Some studies associated with Ch and ChD used in emulsification are given in [78].

3.1.3. Thickening agent.

Using Ch as a thickener by including it in food products (beverages, dairy desserts, chocolate) allows for functional nutrition with bactericidal and antifungal properties and follows medical recommendations [79]. Furthermore, an alternative replacer of undesirable trans- and saturated fats can be the production of food oleogels. These porous materials may contain a large amount of edible liquid oil entrapped in their gel structure [80].

3.1.4. Flocculant.

Being a good flocculant, Ch at low concentration can be used to clarify fruit juices, beverages, whey, etc. Flocculation methods facilitate the removal of fine particles suspended in liquid by forming aggregates with Ch, which can quickly settle from suspensions [52,81].

3.1.5. Functional food additive.

Ch has been shown to be useful for treating lifestyle-related diseases, e.g., Ch affects lipid metabolism, which is an important factor against the development of cardiovascular disease [82,83]. It reduces the total cholesterol and triglycerides in the blood and liver by binding cholesterol from food due to the positive charge of Ch and combining with bile acid (through which cholesterol is absorbed) and removing them from the body.

3.1.6. Food preservation.

The most prominent features of Ch determining its use in the food industry are its antimicrobial and antioxidant properties [84]. It should be noted that Ch has antimicrobial activities only if it is in the polycationic form at film pH values below the pKa. However, there are other beneficial effects on food quality that Ch exhibits, e.g., retardation of lipid oxidation, retention of color and nutrients, and maintaining freshness. The impact on food quality depends on the food matrix and the way of Ch application (incorporation or surface application) [85,86].

3.2. *Environmental applications.*

3.2.1. Water treatment.

Coagulation, flocculation, and adsorption are the main methods in water and wastewater treatment technologies. Commonly used coagulants are metal salts, whereas flocculants are synthetic organic polymers. Nevertheless, their use affects the state of the environment, in particular water pollution by metals and the formation of a large amount of

sediment. The use of bioflocculant Ch is an attractive, environmentally friendly alternative to commercial flocculants [87,88]. This is due to its high efficiency in removing organic substances, metal ions, and other pollutants, such as fungi, radionuclides, and fluorides, as well as its ability to bacterial decomposition of sediment after flocculation [23,38,42,89-95]. The mechanism of heavy metals removal as a result of adsorption on Ch and ChD is discussed in detail in [96]. Surfactant-modified Ch beads were proposed as an adsorbent for Cd²⁺ ion removal from an aqueous medium [32]. To increase the efficiency of flocculation, ChD is used, as well as their mixtures with other polysaccharides (e.g., dialdehyde starch) [97-100].

3.2.2. Air purification.

The Ch nanofibers with high surface area, good mechanical properties, high antibacterial capacity, and the ability to incorporate multifunctional nanoparticles make Ch fibers an effective substrate for air filtration [101].

3.2.3. Soil remediation.

Various modified forms of Ch for soil remediation are reviewed in [102]. In particular, citric acid and water-soluble Ch, as natural and degradable washing agents, were used to remove Zn in the soil by a two-step washing method [103]. Another use of Ch is the improvement of soil mechanical properties, especially for sandy soil stabilization due to increasing interparticle soil cohesion in the presence of Ch [104].

3.3 *Agricultural industry.*

3.3.1. Plant growth regulation.

The natural origin of Ch permits its utilization in agricultural appliances as a promising antibacterial agent. It acts as a catalyst to inhibit the growth of plant pathogens, enhance antagonist microorganisms, and can mitigate the broad use of chemical pesticides [105-107]. Ch-based nanoparticles act as botanical pesticides showing photoprotective and slow sustained release effects that may control plant diseases [108]. The use of Ch as seed and leaf covering, fertilizer, and time-delivered drug (e.g., target delivery of nutrients to plants) has been confirmed by results on improved germination, rooting, leaf development, seed yield, and soil moisture preservation while reducing the incidence of fungal diseases and infections [109-111]. Ch induces plant growth by influencing plant physiological processes like nutrient uptake, cell division, cell elongation, enzymatic activation, and synthesis of protein that can eventually lead to increased yield [105]. Also, applying Ch can reduce stress damage in plant cells by decreasing water content and accelerating several biological macromolecule activities. Ch induces excellent resistance to drought, salt, and low-temperature stress and reduces their negative impact on plants [112-115]. The application of Ch in metal-contaminated areas leads to the remediation of polluted soils [116,117]. The combined action of Ch with other substances can enhance Ch's influence on plants [118,119].

3.3.2. Post-harvest preservation of crop.

To preserve the grown crop, "green" technologies using Ch and irradiated chitosan food coatings were used [57,120,121]. Irradiation is a good choice for obtaining low-resistance Ch with a high degree of deacetylation and a good sterilization effect [84]. The use of Ch and ChD

in plant production to improve yield quality, as well as the benefits and limitations of their application, are presented in the review [122].

3.3.3. Animal husbandry and poultry farming.

Currently, the use of enterosorbents on base on Ch in animal husbandry and poultry farming is a very relevant direction since much attention is paid to the quality of feed [123]. The ability of Ch to bind heavy metals and radionuclides made it possible to reduce the level of contamination of milk and meat of experimental animals fed with contaminated feed when Ch was added to the diet. Ch preparations make it possible to reduce the cost of treatment of animals and eliminate or significantly reduce the use of antibiotics and sulfonamides with cumulative effect [124]. Ch and ChD are known to be used as adjuvants in vaccines against listeriosis, pseudomoniasis, brucellosis, tuberculosis, foot-and-mouth disease, influenza, and other infections [125].

3.4. Pharmacy, medicine.

Due to the unique physicochemical properties such as biodegradability, cytocompatibility, and others, Ch has been investigated for use in a wide range of biomedical applications, including wound healing, tissue engineering, vaccine adjuvants, drug delivery vehicles, enzyme immobilization for biosensing, and others.

3.4.1. Antiviral and antimicrobial agent.

Ch has attracted wide interest due to its antimicrobial activity on bacteria and fungi [9, 126-127]. A literature survey shows that it depends upon several factors such as the pH, temperature, Mw, ability to chelate metals, DD, source of Ch, methods of ChD synthesis, and the type of microorganism involved [43,128-132]. One of the possible reasons for the high antimicrobial activity of Ch is the change in cell permeability due to interactions between the positively charged Ch molecules and the negatively charged microbial cell membranes [69]. It is noted that low Mw Ch (less than 10 kDa) has a higher antimicrobial potential compared to high Mw Ch due to the better solubility of the former, which makes it suitable for binding to microorganisms and exhibits an inhibitory effect [133].

Ch and ChD have been shown to exhibit direct antiviral activity, to be useful vaccine adjuvants, and to have potential anti-SARS-CoV-2 activity [134-136]. The review of proposed mechanisms of the antiviral activity of Ch is presented in [135]. Ch nanoparticles have attracted wide interest due to their antimicrobial activity on bacteria and fungi [126]. The use of ChD obtained by attaching bioactive substances to Ch via covalent bonds or mixed with other materials is very effective for obtaining ChD with enhanced solubility and antimicrobial properties [137,138].

3.4.2. Wound and burn treatment.

Ch-based functional materials, due to their antibacterial and hemostatic activity as well as the ability to promote skin regeneration, were found to use in skin wound repair [139]. One of the reasons for the effectiveness of Ch in wound healing is its stimulating effect on the immune system: it can be considered an analog of lipopolysaccharides of the cell walls of microorganisms that act as macrophage activators. The possible mechanisms of how Ch enhances coagulation and wound healing are discussed in [130]. Ch's hemostatic properties

additionally enable it to lessen torment by blocking nerve endings. Mechanisms with which Ch-based functional materials promote wound repair and the latest information about their applications are presented in [138]. A perspective trend in this direction is to enhance gels' healing effect and mechanical properties by incorporating other chemical components (e.g., bioactive molecules) into the Ch wound dressings. That accelerates wound healing by inducing intracellular signaling and stimulating the synthesis of skin repair-related proteins [140].

3.4.3. Hypolipidemic activity.

It was demonstrated that dietary fibers, e.g., pectin and particularly Ch, may play crucial roles in decreasing plasma total cholesterol concentrations, low-density lipoprotein cholesterol, and triacylglycerols, decreasing the risk of cardiovascular disease [141]. As mentioned above (in 3.1.3), using food additives with Ch effectively reduces blood lipid [79].

3.4.4. Drug and gene-delivery systems, cell carriers, microcarriers.

Ch-based nanocarriers (ChNCs) are considered suitable drug carriers because of their ability to encapsulate various drugs and overcome biological barriers to deliver cargo to its destination [142]. Another application of fluorescent ChNCs is cell labeling to detect disorders *in vivo* and live control cells *in situ* [143]. A lipid-modified Ch containing mucoadhesive self-emulsifying drug delivery systems has been designed to enhance cefixime oral delivery [44]. Ch aerogels as drug delivery vehicles can offer improved drug bioavailability and drug loading capacity due to their highly porous network, providing a large specific surface area [144]. There is information about using Ch in gene therapy to introduce genetic material into cells for various purposes, e.g., for developing non-viral DNA vectors [145]. Gold nanoparticles conjugated with Ch and acylated Ch were used to evaluate the transfection efficiency of plasmid DNA into cell culture (HEK-293) [146]. Special attention was put on applying Ch nanoparticles in developing a new system for anticancer drug delivery [3,147]. Ch has a mucoadhesive character and can disrupt tight epithelial junctions, thus acting as a permeability enhancer, so it is well-suited for the challenging field of ocular drug delivery [148]. An interesting application of Ch for delivering nucleic acids into cells was used in Ch-mediated gene delivery for fish biotechnology applications [149].

3.4.5. Surgery and dental therapy, regenerative medicine.

Ch is a promising biomaterial in surgery, tissue engineering, and bone regeneration due to its ability to stimulate the recruitment and adhesion of osteogenic progenitor cells [150]. Combining Ch-based scaffolds with natural or synthetic polymers induces their mechanical properties, accelerates bone regeneration, and promotes osteogenesis [150, 151]. The grafting of short and medium chains of oligo-lactides on Ch gives hybrid systems that show promise for producing biodegradable and biocompatible materials for use in regenerative medicine [77]. Ch's good film-forming ability allows it to be used in dental implant coating, demonstrating good compatibility of coated surfaces with osteoblastic and fibroblastic cells [153]. Surgical treatment of inflammatory periodontal diseases is another application of Ch [154,155]. It was reported that chewing gum or mouthwash containing Ch can decrease the number of cavity-causing bacteria in the mouth and significantly reduce gingival inflammation. The combination of Ch with chlorhexidine gave more effective results [156]. Based on the remineralizing property of Ch, which hardens tissues of the tooth and therefore behaves as a

desensitizer, Ch can be used in toothpaste. According to the systematic review, the use of chitosan has shown better surgical healing of post-extraction oral wounds [157]. Surgical sutures coated with Ch were found to prevent microbial community biofilm adhesion [158].

3.4.6. Blood anticoagulant.

The interaction of the Ch amino groups with the acid groups of blood cells and plasma proteins leads to the formation of blood clots and can cause thrombogenic and hemostatic action. Many authors have reported that Ch has thrombogenic action because it has the ability to activate both complement and blood coagulation systems. Ch itself demonstrates weak anticoagulant properties but may manifest them when modified [159,160].

3.4.7. Blood dialysis membranes, artificial blood vessels, contact lenses.

Natural polymers possess high bioactivity and biocompatibility, so they have been successfully applied in implanted biomedical devices [161]. The electrospun scaffolds obtained using Ch or Ch functionalized with aliphatic chains demonstrated the absence of cytotoxicity, inflammation response, and eryptosis. They can be used for blood purification devices, hemodialysis membranes, and vascular grafts [162].

3.4.8. Immunostimulatory, immunoregulator, antitumor, anti-inflammatory, and antioxidant agent.

Ch exhibits considerable immunostimulatory activity by inducing innate immune cells to release a wide range of pro- and anti-inflammatory cytokines, chemokines, growth factors, and bioactive lipids [163]. Ch-based nanoparticles are important in nano-medicine, particularly in developing new delivery systems for anticancer drugs [164]. Nanoparticles prepared from negatively charged Ch sulfate and positively charged hydroxypropyl trimethyl ammonium chloride Ch showed immunostimulatory effects on dendritic cells [165]. It was found that nanoparticles prepared by using positively charged quaternized Ch and negatively charged mucopolysaccharides such as chondroitin sulfate, heparin, and hyaluronic acid can significantly promote the secretion of immunoglobulins in mice [166]. Ch was shown to be an excellent candidate for a cancer cure or its diagnosis [167-169]. The action of some antitumor agents increases when Ch has been applied. So, a colchicine-Ch conjugate demonstrated significantly better tumor growth inhibition than a new all colchicine derivative – furanoalcolcolchicinoid, possibly as a result of better accumulation in the tumor [170]. The possible pathways of how Ch reacts with cancer and the body's immune system to demonstrate immune and antitumor effects are discussed in [130]. Antiviral, antitumor, and other medical applications of Ch fibers are summarized in [171]. Ch is known to exhibit anti-inflammatory activity. However, combinations of Ch with anti-inflammatory agents (e.g., 5-amino salicylic acid) have shown the possibility of optimizing its anti-inflammatory potential [172]. The antioxidant properties of Ch are discussed in [9,173].

3.4.9. Antigastritis, antacid, antiulcer.

Ch is an alkaline polysaccharide containing free amino groups that can neutralize gastric acid. Creating a protective layer in the stomach can be an effective remedy for peptic ulcer disease. It has been shown that nanoparticles of Ch-bilirubin conjugate (CS-BR) have an anti-ulcer effect in gastric epithelial cells, effectively improving the antioxidant capacity of BR

and inhibiting the secretion of inflammatory cytokines in the cells and tissues [174]. The allantoin-loaded Ch/sodium tripolyphosphate nanoparticles showed good results as an auspicious oral delivery system for gastric ulceration management [175]. Ch-curcumin solution has been found to be a safe and potential alternative agent in treating oral ulcers [176].

3.4.10. Anti-hypertensive activity.

Ch is useful for preventing and treating hypertension because of inhibiting the absorption of Cl^- in living bodies, where the Cl^- has an action to activate angiotensin I converting enzyme having a hypertensive action [177]. The beneficial effects of Ch on hypertension as well as other metabolic syndromes (obesity, dyslipidemia, diabetes mellitus, and hyperglycemia) are summarized in the review [178] with a focus on possible mechanisms of the prevention and treatment of them by Ch.

3.4.11. Veterinary medicine.

Ch preparations have recently become increasingly in demand in veterinary medicine and animal husbandry. Ch is used in veterinary medicine in two directions: externally as a wound-healing agent and internally as an enterosorbent, anti-inflammatory and bacteriostatic agent, as well as part of vaccines [179]. Having a positive effect on the healing of skin lesions in animals, Ch is a safe biomaterial for skin treatment in veterinary practice [180].

3.5. *Technological processes.*

3.5.1. Production of substances.

The presence of electron-donating amino and hydroxyl groups provides ample opportunities for introducing various ionogenic groups of an acidic and basic nature, making ChD very promising for chromatography in the separation and purification of biologically active compounds (nucleic acids and products of their hydrolysis) [181]. Recent development trends for novel chiral stationary phases in high-performance liquid chromatography based on ChD are presented in the review [182]. Quite promising results for the large-scale production of enzymes were proposed using Ch as a commercially available carrier for enzyme immobilization [183]. An effective method for recovering protein from starch wastewater of sweet potatoes using modified Ch for subsequent use as ingredients for livestock feed is proposed in [184].

3.5.2. Catalytic applications.

The idea of the use of environmentally benign and sustainable catalysts requires searching for natural catalysts such as biopolymers. Ch and ChD are excellent alternatives for this purpose [6]. A main challenge in the field of catalyst development at present is how to design porous Ch-based catalysts to increase the amount of available active sites and thus improve the efficiency of catalytic processes [40, 185]. The use of Multi-Wall Carbon nanotubes (MWCNTs) was applied for facile fabrication of ternary MWCNTs/ZnO/Ch nanocomposite for enhanced photocatalytic degradation of methylene blue [186]. Fe_3O_4 nanoparticles, obtained by the green method, included in the Ch-matrix are a good example of the functional nanostructures, which can be used not only as a catalyst but also as a magneto-

optical storage device, protection against electromagnetic interference in biomedical sensing and so [187].

3.5.3. Paper industry.

Ch may be used as a substitute for fillers in paper processing and as an adhesive to improve paper properties [188]. The greater strength and smoothness of the paper during Ch processing is due to the formation of ionic bonds compared to hydrogen bonds that occur in traditional paper production. Valuable recommendations for improving the printing capability and physical properties of high-quality paper products for many promising applications are provided in [189]. The Ch-coated papers demonstrate significant enhancements in water repellency and mechanical properties and also show significant bacterial inhibition effects due to the natural anti-microbial activity of Ch [190]. Ch, a chelating agent, is also used to treat pulp and paper wastewater to remove lignin, color, and undesired contaminants [191].

3.5.4. Textile industry.

Environmental sustainability is one of the main motivations for development and innovation in the textile industry. Ch is well suited to improve the aesthetic and functional properties of textiles as well as to avoid the action of microorganisms [192,193]. Treatment of cotton with Ch prior to dyeing can improve dyeability in reactive and solophenyl dyes. Incorporating metal nanoparticles in textiles allows for obtaining multifunctional properties, such as ultraviolet protection, self-cleaning, electrical conductivity, antimicrobial, antistatic, antiwrinkle, and flame retardant properties, without compromising the inherent characteristics of the textile [194]. Due to the high content of amino and hydroxyl groups, Ch can provide active sites for forming complexes with silver ions which can be further reduced to a silver layer on the surface of Ch fibers. Such fibers can have promising applications in the field of wearable electronics materials [195].

3.5.5. Leather industry.

Ch and its composite materials are used in the leather industry. Having cationic properties, Ch can interact with the anionic part of collagen molecules (the main protein of animal skin), enhancing adhesion between collagen fibers and forming a more stable network of collagen fibers in the composite. An overview of methods for obtaining composites based on Ch for use in the leather industry and the mechanism of their antibacterial action is presented in [196]. In addition, bacterial resistance of leather surfaces using nano/microparticles of Ch and nano/microstructures of Ch with silver nanoparticles was discussed in [197].

3.5.6. Nuclear industry.

Localization of radioactive waste and separation and concentration of radionuclides from the environment and industrial solutions are other possible uses of Ch. Thus, the Ch hydrogel polymer could be promisingly applied in removing both Ar-III and ARS dyes from radioactive waste solutions [198]. The graphene oxide/Ch sponge could be used as an alternative adsorbent for removing radioiodine from wastewater [199].

3.5.7. Mining industry.

The possibility of ecofriendly metal removal from the acid mine drainage was investigated through a combination of neutralization and adsorption methods using Ch [200]. For the use of Ch as an adsorbent for Al^{3+} and Cd^{2+} ions in acidic conditions of mine drainage waters, Ch has been modified with hydrophobic hexanoyl chloride to reduce the solubility at a lower Ph [201].

3.5.8. Protection against corrosion.

Ch and ChD are widely used as effective replacements for toxic inorganic and organic substances against the corrosion of metals [202,203].

3.5.9. Cosmetics.

The different functional roles of Ch as a skin care, hair care ingredient, oral hygiene agent, and carrier for active compounds are known. The good surface activity of Ch complexes with surfactants can be used in the formulations of face wash, body wash, liquid detergents, and so on [33]. Further study of Ch-based products' physicochemical and functional properties will promote their use in personal care and the cosmetic industry [204,205].

3.5.10. Energy applications.

Ch and ChD solutions are among the most promising electrolytes studied for use in electrochemical devices (biosensors, fuel cells, lithium-ion batteries, electromagnetic interference protection). A pair of electrons in amino groups demonstrates an affinity for metal ions, which makes Ch a suitable material for obtaining sensors. In this context, much literature has been reported on using Ch and ChD for that purpose [206-214]. The ability of chitosan to form transparent film attracted great attention among scientists for developing solar energy-based cells. It may work as an efficient cathode interlayer for organic solar cells [215]. Polymer membranes based on Ch and variable content of carbon nanotube-TiO₂ composites were prepared by solvent casting at room temperature. It is again an example of an environmentally friendly material with attractive properties for energy conversion device applications.

The transformation of Ch from shrimp to solid-state photovoltaics is really impressive.

3.6. Modeling of natural processes.

Another interesting application of Ch is its use in models that allow the study of processes occurring in nature. For instance, the variety of factors affecting the processes in the river-sea mixing zone required the study of the transfer process in laboratory conditions, where Ch is chosen as a model of fresh dissolved organic matter (flocculant) released in situ in the river-sea area [216]. When studying the formation of the soil structure, chitosan can be used as a model of the in situ forming organic matter capable of binding mineral particles modified by humic substances [217].

4. Conclusions

Ch is a versatile nontoxic compound with a diverse spectrum of action that positively impacts human health, environment protection, remediation, and some technological processes. Much interest in studying Ch is related to its unique physicochemical and biological

properties, which open up the possibility of their application in various fields. The current state of the environment requires using alternative energy sources and biodegradable biopolymers to reduce their impact on nature [218]. The ability of such biopolymers to remediate the environment, the search for new non-toxic, biocompatible medical forms, and numerous other tasks are drawing attention to Ch's study. Nanocomposites of Ch and its derivatives and their combination with other substances are greatly interested in this issue.

An indication of the constantly growing importance of this biopolymer is the total of over 58,625 scientific articles published between 2000 and 2017. The top ten most cited reviews published over the last two decades on the topics “Chitosan” and “Review” of a total of 2702 reviews are presented in [219].

In this work, we would like to provide an overview of recent research related to the preparation of chitosan derivatives as well as the use of these substances as agents of green chemistry. We have not covered many valuable studies on this subject carried out in previous years. Many of these works will provide ideas for future research as interest in studying Ch, and its derivatives will continue to grow.

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Conflicts of Interest

The authors declare no conflict of interest. The funders had no role in the study's design; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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