

USE OF CHITOSAN AS ANTIMICROBIAL, ANTIVIRAL AND ANTI-POLLUTION AGENT IN TEXTILE FINISHING

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ABSTRACT

With the industrial developments in recent times, the textile industry also needs sustainable and environmental-friendly resources. Today's world has been overburdened with the use of synthetic or hazardous materials in day-to-day life. Chitosan polymer obtained from chitin deacetylation, having a lot of properties beneficial to mankind without being hazardous to environment and humans is currently gaining popularity for research and development all over the globe. Antimicrobial and antiviral textile finishing with the help of chitosan is a new trend in the textile field. Also, chitosan having good adsorption properties finds its application in textile effluent treatments. This review reports and discusses multifunctional finishing and dyeing of textiles with chitosan and highlights its application for textile wastewater treatment.

KEYWORDS

Chitosan; Textile; Antimicrobial; Antiviral; Dye.

INTRODUCTION

Last years, in addition to the growing interest in natural eco-friendly agents, there has also been a drive toward the introduction of green processing techniques in textile manufacturing due to rigorous ecological legislation and growing environmental concerns [1]. Turning from established hazardous synthetic value chains towards more sustainable bio-based products is one of the key factors to achieve these goals. This means to substitute petrol based agents with reactants and products derived from renewable resources. Searching for materials and reagents that could contribute to functionality and sustainability of textile products, researchers identified chitosan as an agent of choice [2].

Chitosan, poly-(1,4) 2-amido-2-deoxy- β -D-glucose, is a renewable polymer derived from deacetylation of the polysaccharide chitin. Chitin, poly-(1,4)-2-acetamido-2-deoxy- β -D-glucose, is found throughout the biosphere. Its estimated production is 1012 tons per year. Chitin is the second most abundant polymer after cellulose. Sea animals, insects, and microorganisms are the sources of chitin (Table 1).

In more than 90 % of all animal species and insects, chitin-based composites are the major constituents of the exoskeletons of arthropods (Figure 1). A critical evaluation of potential sources of chitin and chitosan [3] concluded that crustaceans, fungi and insects were, respectively, the principle source of chitin and

chitosan and would remain so for the immediate future. However, it was envisaged that both krill and cultured fungi would complement these major sources of raw materials [4].

Table 1. Sources of chitin and chitosan.

Sea Animals	Insects	Microorganisms
Crustacean	Scorpion	Green algae
Mollusc	Spider	Yeast (β -type)
Coelenterate	Gnathopod	Fungus
Annelids	Ant	Penicillium
Lobster	Cockroach	Brown algae
Crab	Beetle	Chytrid
Shrimp		Ascomycetes
Krill		Spore

The generation of the highly reactive amino groups in the chitosan polymer imparts versatility in its use for food, textile, cosmetics, environmental, biomedical and pharmaceutical applications. Due to the presence of these amino groups, chitosan is easily soluble in weak acids and forms viscous solutions that can be transformed into films, sponges, beads ... etc.

We recognize the importance of chitosan in eco-friendly textile finishing processes. Therefore we present our review paper as follows. Initially we introduce a general overview of chitosan and its properties. Second, we highlight the use of ChSN as an antimicrobial and antiviral finishing agent of textile materials. The next part will be dedicated to present research investigations where chitosan was used as

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an anti-pollution dyeing agent. Finally we discuss the advantages and limitations of ChSN polymer and we suggest future research trends.

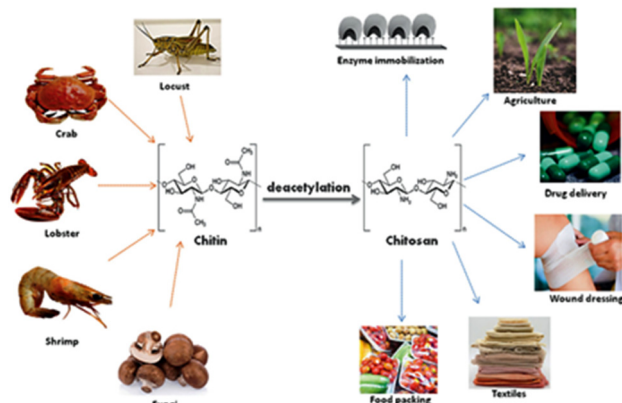


Figure 1. Main sources of chitin and chitosan and their potential applications [4].

CHITOSAN PROPERTIES

Chitosan is obtained by deacetylation of chitin biopolymer. Crustacean shells are the most important source of chitin for commercial use due to their high chitin content and availability. Chitin is isolated from crustacean shells in three steps: demineralization (DM), deproteinization (DP), and elimination of lipids and pigments. The chemical methods for the preparation of chitin from crustacean shell waste consist of mechanical grinding, DM with strong inorganic acids, and DP with alkali at elevated temperature. The chemical DM and DP processes have several drawbacks such as poorly controlled depolymerization, resulting in the reduction of molecular weight and viscosity and hydrolytic deacetylation. In addition, chemical treatments engender hazardous environmental problems such as disposal of wastewater, making this process ecologically aggressive and a source of pollution to the environment because of the high concentration of mineral acids and caustic chemicals employed [5]. Along with increased demands for an environmentally friendly society, eco-friendlier processes using enzymatic and microbiological methods for producing chitin have attracted great interest in the field of green biotechnologies. As an alternative to chemical processes for extracting chitin, fermentation processes using proteolytic microorganisms or proteolytic enzymes have been studied for decades for various crustacean shells. Procedures for chitin and chitosan production by chemical and biotechnological treatments are briefly compared in (Figure 2).

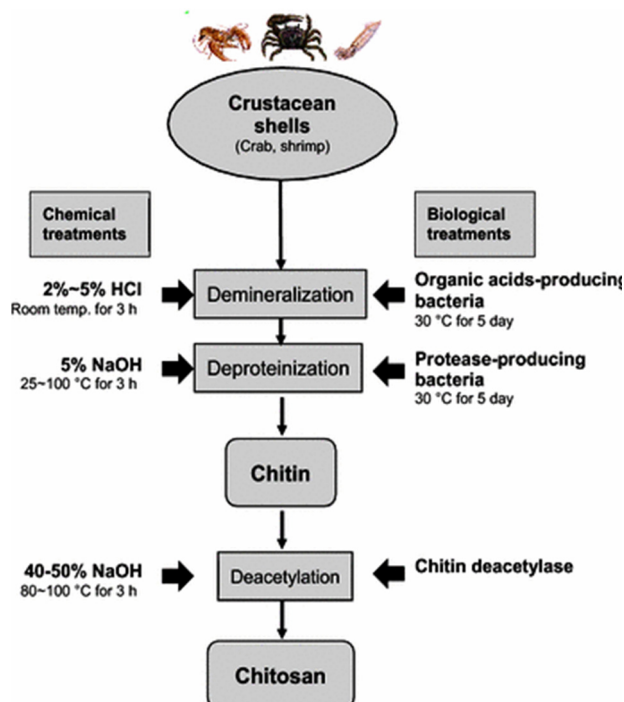


Figure 2. Chemical and biological methods for the extraction of chitin and its conversion to chitosan [6].

Obtained chitosan is a linear polyamine with too much nitrogen content and reactive amino and hydroxyl groups (Figure 3). The amino group of chitosan is not protonated in neutral medium and therefore it is insoluble in water. Chitosan dissolves readily when electric repulsions (corresponding to cationic charges) are more important than the attracting interactions (such as hydrogen bonding and Van der Waals interactions).

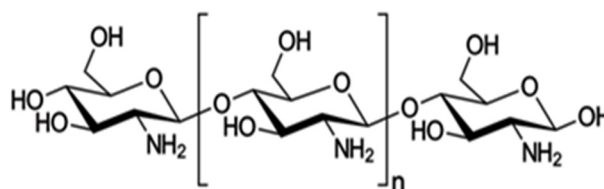


Figure 3. Chemical structure of chitosan.

The physicochemical characteristics (Figure 4) determine the relationship that exists between the structure of chitosan compounds and their potential uses. Deacetylation degree (DD) and molecular weight (MW) are the most important parameters affecting ChSN properties [7]. Often analyzed are other characteristics such as solubility, crystallinity, viscosity, nitrogen and ash contents and water retention value [8].



Figure 4. Physiochemical characteristics of chitosan [9].

Deacetylation degree

The degree of deacetylation of chitosan is defined as the ratio of the number of glucosamine GlcN groups to the overall number of N-acetylglucosamine GlcNAc and glucosamine groups [10]. The DD value of a ChSN sample is one of the most important factor in assessing its applications in the medical, nutritional, environmental and biotechnological fields [11]. It can also be related back to the specific biological and structural properties and functions of chitosan. It should be clear that chitosan is the deacetylated form of chitin and it must be characterized by a degree of acetylation when a degree of deacetylation is valid for chitin, the initial form of the polymer nearly fully acetylated. For chitosan, measurement of the DD is crucial in determination of its chemical structure, physical properties and interrelation. It is also essential in providing a basis from which the functions and applications can be predicted and optimized [12]. Therefore, a suitable method of obtaining reliable DD values is essential for diverse researchers working in those fields. To date, several techniques have been developed and applied for determination of the DD of chitosan, such as proton nuclear magnetic resonance (^1H NMR, solid ^{13}C NMR and ^{15}N NMR), UV spectrophotometry, conductometric and potentiometric titration, differential scanning calorimetry (DSC) or CHN elemental analysis among others [13].

Molecular weight and viscosity

Knowledge of the molecular weight of chitosan is of fundamental importance for the understanding of its applications and its role in living systems. The molecular weight of ChSN depends largely on the conditions of deacetylation and can be determined by

several methods such as chromatography, light scattering and viscometry [14]. The latter is the simplest and most popular method to determine molecular weight of chitosan. The average MW is determined by the Mark–Houwink–Sakurada equation which relates this parameter with the intrinsic viscosity: $[\eta] = k \text{ MW}^\alpha$, where k and α are empirically determined coefficients. However, this method has the disadvantage of not being absolute because it relies on the correlation between the values of intrinsic viscosity with those of molecular weight. ChSN is available commercially with molecular weight ranging from 5 to 1,200 kDa.

Viscosity of chitosan increases with increase in its molecular weight and concentration. Increasing the degree of deacetylation also increases the viscosity. This can be explained by the fact that high and low deacetylated chitosan have different conformations in aqueous solution. ChSN has an extended conformation with a more flexible chain when it is highly deacetylated because of the charge repulsion in the molecule. However, the chitosan molecule has a rod-like shape or coiled shape at low degree of deacetylation due to the low charge density in polymer chain. The viscosity of chitosan solution is also affected by factors such as concentration and temperature. As the chitosan concentration increases and the temperature decreases, the viscosity increases. Chitosan viscosity decreases with an increased time of demineralization due to depolymerization.

Physical characteristics of chitosan

The principle physical parameters of ChSN polymer and their methods of determination are summarized on table 2:

Table 2. Main physical properties of chitosan with corresponding measuring methods.

Property	Values	Measuring methods
Deacetylation degree	66 – 99 %	Fourier Transform Infrared [15] Potentiometric Titration [16]
Macromolecular weight	5 – 1.200 kDa	Gel Permeation Chromatography [17] Viscosimetry [18]
Particle size	< 30 μm	Optical microscopy [19]
viscosity	< 5cps	Capillary test [20]
Density	1.35 – 1.4 g/cm ³	Density meter [21]
Melting point	152-159 °C	Differential Scanning Calorimetry [22]
Crystallinity	57 %	X-Ray diffraction [23]
Ash value	> 1 %	Gravimetric analysis [24]
Moisture content	> 8.5 %	Gravimetric analysis [25]

Chitosan chemistry

The chemical behavior of ChSN includes [26]:

- Linear amino polysaccharide with too highly nitrogen content

- Insoluble in organic solvents and water
- Highly soluble in acidic solutions
- Reactive amino and hydroxyl groups for chemical activation and cross-linking
- At weak base, deprotonated amino group acts a powerful nucleophile
- Enable to form hydrogen bonds intermolecularly.
- Forms salts with inorganic acids and organic.
- Form chelates and complexes with many transitional metal ions
- Cationic biopolymer with rise charge density (one positive charge per glucosamine residue).
- Flocculating agent that interacts with negatively charged molecules.

The presence of large amounts of protonated $-NH_2$ groups on the chitosan structure accounts for its solubility in acid aqueous media since its protonation constant (pK_a) value is approximately 6.5 [27]. Chitosan solubility depends on different factors such as polymer molecular weight, degree of acetylation, pH, temperature, and crystallinity. When around 50% of all amino groups are protonated, chitosan becomes soluble [28]. This behavior is explained by the fact that deacetylation leads to an increase in the number of glucosamine units and a modification in the crystalline structure of the polymer. On the contrary, a reduction in the crystallinity is observed for high acetylated ChSN samples [29]. Chitosan can easily form quaternary nitrogen salts at low pH values. So, organic acids such as acetic, formic, and lactic acids can dissolve chitosan. The most commonly used solvent for chitosan is 1 % acetic acid at about pH 4.0. Chitosan is also soluble in 1 % hydrochloric acid and dilute nitric acid but insoluble in sulfuric and phosphoric acids.

As seen in Figure 5, the reactive groups found in chitosan are a primary amino group (C2) and primary and secondary hydroxyl groups (C6, C3). Glycosidic bonds and the acetamide group can also be considered functional groups. These functional groups allow for a great number of modifications, producing polymers with new properties and behavior. ChSN derivatives have been produced, aiming to improve chitosan's properties, such as solubility or biodegradability, or to introduce new functions or properties. For instance, solubility has been improved in water aqueous media by deacetylation, depolymerization or quaternisation among other processes [30].

Interactions of chitosan with textile materials

The backbones of chitosan and cellulose are similar which has been postulated as a reason for their mutual affinity.

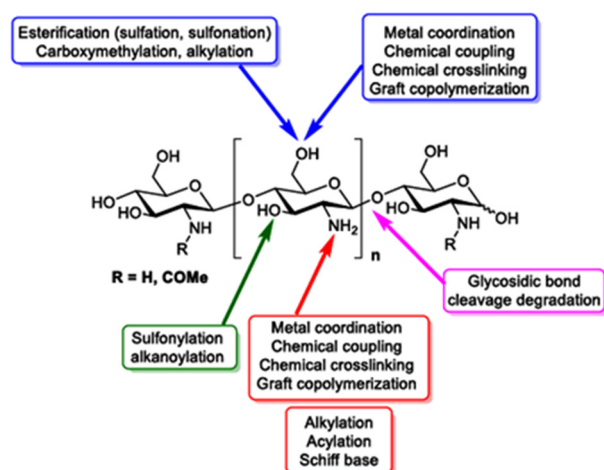


Figure 5. Chemical modifications of ChSN functional groups [31].

In acidic solution, chitosan's amine groups are positively charged; while in alkaline conditions, chitosan is uncharged and its solubility in water is diminished. It has been observed that chitosan adsorbs on textile cellulosic materials (cotton, viscose, jute ...) in both acidic and alkaline conditions [32]. The more possible cellulose-chitosan intermolecular interactions are based on hydrogen bonds and Van der Waals forces. However, ionic and/or covalent bonds can be formed under special conditions and cellulose treatments [33]. For some applications, the irreversible binding of ChSN to the cellulose substrate is extremely important, especially when the bio-activity of a material should appear only at the interface on the biological medium. The functional groups of cellulose, such as carboxylic and aldehyde groups, contribute to the binding. The $COOH$ groups provide electrostatic attraction between cellulose and ChSN as an adsorbent, while aldehyde groups allow covalent binding of ChSN via Schiff base formation [34]. These chemical interaction mechanisms between chitosan and cellulosic fiber were investigated by Grande et al. [35] using FTIR spectra analysis. The strong absorption in the region $3500-3100\text{ cm}^{-1}$ signifies the strong hydrogen bonded ChSN and cellulosic material and also the existence of primary $-NH_2$ and secondary $-NH$ groups. The absorption peak that appeared at 1640 cm^{-1} for chitosan-treated cellulose suggested the formation of Schiff base ($C=N$ double bond) between aldehydic carbonyl group of cellulose and amino group of ChSN corroborating the formation of a donor-acceptor interionic complex between the cellulose and ChSN.

It is known that the binding of chitosan to wool is due to ionic interactions, such as carboxyl groups in wool forming salts with the free amino groups in chitosan and hydrogen bonding interactions between hydroxyl and amide groups of the wool with the hydroxyl groups of the chitosan [36]. These chitosan-protein fibers interaction was confirmed by Silva et al. [37] using X-ray photoelectron spectroscopy (XPS)

analysis of chitosan coated soybean fabrics. They noted that after the addition of chitosan there was a positive shift in the binding energies of the C–O/C–N and O–C–O/C=O peaks. The presence of chitosan significantly increases the relative percentage of binding energy area of these two peaks displays also a positive shift of the carboxylic component peak from 288.5 to 288.9 eV.

The above mentioned weak ionic interactions between chitosan and proteinic fibers limit the application of the former to proteinic textile materials. Chemical modification of chitosan can create a strong chemical bonding between chitosan and wool, which consequently increases the washing durability. Glyoxal or glutaraldehyde or citric acid has been used as crosslinking agents. Chitosan has also been grafted to wool after acylation with succinic anhydride and phthalic anhydride [38].

Biological properties of chitosan

Chitosan is not only a naturally renewable resource but also a low-cost polymer and very versatile material. Much of the recent interest in chitosan and its derivatives arise from the fact that they combine several favourable biological characteristics, including biodegradability, biocompatibility and non-toxicity, making them valuable materials for pharmaceutical, biomedical as well as textile applications. Besides these properties, chitosan-based materials present antibacterial, antiviral, hemostatic, antioxidant and anti-inflammatory activities [39].

• Hemostatic effect

One of the biggest challenges for the modern medicine is massive blood loss, which is responsible for the highest number of deaths resulting from various injuries. Until now, no functional, food or drug administration-approved artificial blood substitutes have been developed. Thus, the ability of fast and efficient inhibition of bleeding is an issue of a great importance. Currently, the most popular methods of hemostasis restoration is the application of an hemostatic agent. An ideal hemostatic agent should have antimicrobial property. This can be achieved by applying a textile matrix (dressings and sponges) containing antibiotics that are effective against most bacteria species. Due to its proved antimicrobial, antioxidant and anti-inflammatory properties, chitosan has been considered as an attractive candidate for hemostasis in medical applications [40]. In general, ChSN can enhance the hemostatic activity by attracting the negative charge erythrocytes and red blood cells, plasma, activation and aggregation of platelets, and blood coagulation [41]. As a result, chitosan is normally used as hemostatic dressing for wound healing in clinic instead of a blood-contact medical device. The hemostatic property was attributed to the polycationic characteristic of chitosan

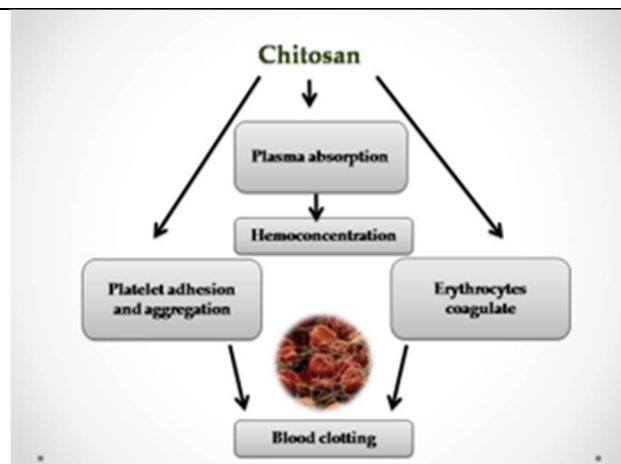


Figure 6. Mechanism of chitosan hemostatic effect [43].

and its non-specific binding to cell membranes resulting from the positively charged amino groups along the molecular chains [42]. In fact, the positively charged chitosan enhanced platelet adhesion, aggregation and activation of intrinsic blood coagulation (Figure 6).

• Antiviral activity

There are reports of antiviral properties of chitosan and its modified forms. In the report of Ai et al. [44] chitosan extracted from the larvae of *Musca domestica* exhibited inhibition of infectivity of two baculo viruses, *autographa californica* multiple nucleopolyhedro virus (AcMNPV) and *bombyx mori* nucleopolyhedro virus (BmNPV). In another study by Davis et al. [45], 53 kDa chitosan showed a reduction in the growth of surrogate enteric viruses, feline calicivirus FCV-F9 and murine norovirus MNV-1. Derived 3,6-O-sulfated chitosan was shown, by Gao et al. [46], to exert broad antihuman papillomavirus (HPV) activities in vitro, by targeting the viral capsid protein, known to play a role in viral replication inside a host cell. Li et al. [47] showed that sialyllactose (SL)-incorporated chitosan based materials are able to inhibit viral adhesion of influenza virus to a host cell. The SL-chitosan binds to hemagglutinin protein on the surface of an influenza particle, which is responsible for viral attachment to the host cell surface via binding with surface glycoligands. Guo et al. [48] reported on antiviral mechanism of a combined biological agent consisting of chitosan oligosaccharide and cytosinepeptidomycin (CytPM-ChSN). CytPM-ChSN effectively inhibited tobacco mosaic virus (TMV), suppressed viral RNA and capsid protein accumulation. Davydova et al. [49] reported that low molecular weight derivatives of chitosan especially with decreasing degree of polymerisation showed antiviral property against TMV by inhibiting the development of necrosis caused by the virus.

Figure 7 summarizes the general antiviral mechanisms of actions of the chitosan and chitosan derivatives. Broadly, chitosan based materials act on

non-enveloped viruses (murine norovirus-1, MS2, feline calicivirus F-9, human papillomavirus, hepatitis A virus, Coxsackie virus B4, phi X174) through viral capsid interaction. One or more mechanisms of action may take place. The positively charged chitosan has been postulated to bind to viral nucleic acid and reduce the viral genome integrity [51]. With respect to enveloped viruses, chitosan may stimulate the immune system to inhibit Newcastle virus, bind to envelope glycoprotein to inhibit viral entry (of HIV-1, influenza) and/or inhibit the virus growth (of herpes simplex virus) in a nonspecific manner. The antiviral actions of such therapeutics and their respective physicochemical constructs can be exploited to eradicate the SARS-CoV-2, another variant of enveloped virus.

- **Antioxidant property**

The body maintains an oxidation balance under normal physiological conditions. When the antioxidant capacity is not adequate to combat the sudden increase in free radicals, the surplus free radicals lead to cell injury, metabolic disorders of the cellular macromolecules, and the occurrence of skin and soft tissue diseases. The antioxidant properties of ChSN are attributed to the amino and hydroxyl groups in its molecular chain, which can effectively scavenge excessive free radicals in the human body [52]. The antioxidant activity of chitosan mainly depends on its relative molecular weight and the level of acetylation. Chitosan shows a greater ability in scavenging free radicals having relatively low molecular weights and higher levels of acetylation [53]. In addition, chitosan derivatives containing Schiff's base and a quaternary ammonium salt exhibit stronger antioxidant capability than chitosan due to the presence of hydroxyl and halogen groups [54]. The antioxidant capacity of ChSN can be regulated by adjusting its molecular weight, acetylation level, and the extent of chemical modification, thereby conferring tremendous application prospects in medical cosmetology and the treatment of soft tissue diseases [55].

- **Anti-inflammatory effect**

Inflammation is the first protective response to infection or injury of human body driven in a tissue compartment by a specific set of immune and inflammatory cells with the aim of restoring its structural and functional integrity after exposure to an adverse stimulus [56]. It is known that chitosan and its derivatives, which interact with eukariotic cells, stimulate the synthesis of anti-inflammatory cytokines, causing different directions in the effect on immunity, both stimulating and inhibitory. Thus, it was established that chitosan contributes to the decreased synthesis of anti-inflammatory cytokines by human astrocytoma cells, without affecting time their vitality at the same. It also prevents the development of chronic inflammation, which can

cause severe diseases [57]. When studying experimental bronchial asthma in mice after intranasal introduction of ChSN, the anti-inflammatory effect of water soluble forms of chitosan manifested itself in a decreased degree of infiltration of the bronchial epithelium by leukocytes and inhibition of the synthesis of various anti-inflammatory factors of the immune-response [58]. Davydova et al. [59] tested the anti-inflammatory activity of chitosan with high molecular weight (115 kDa) and low MW (5.2 kDa), and both chitosan samples presented an intensified induction of anti-inflammatory IL-10 cytokine in blood and suppression of colitis progress. The authors concluded that the main contribution to anti-inflammatory activity of chitosan was driven by structural elements comprising its molecule, but not depending on MW. Oliveira et al. examined the inhibition of pro-inflammatory cytokines and anti-inflammatory activities of chitosan [60]. From the achieved results, a reduction of TNF- α (pro-inflammatory cytokines) in 3~10 days of cells cultured on ChSN material and significant increase of anti-inflammatory cytokines IL-10 and TGF- β 1 were presented.

- **Antimicrobial activity**

The antimicrobial activity has been considered the most essential and influential bioactivity of chitosan and employed not only to the preparation of biomedical materials but also to the functionalization of other materials including textiles. The mechanism of antimicrobial activity of chitosan is not yet fully understood although numerous researches have been carried out so far. The antimicrobial effect of chitosan is much higher comparing to chitin due to the numbers of the amine groups that is responsible for cationic property of chitosan. Positively charged chitosan at acidic condition might interact with negatively charged residues of carbohydrates, lipids, and proteins located on the cell surface of bacteria, which subsequently inhibit the growth of bacteria [61]. Thus, the electronic property of chitosan plays a very important role in the inhibition mechanism of microorganisms (Figure 8). The high density of positive charge on the structure of chitosan or its derivatives generates strong electrostatic interaction that is affiliated with DDA. With this theory, chitosan is more promising for the inhibition of Gram-negative than Gram-positive bacterium since the negatively charged cell surfaces interact more with positively charged chitosan [62-63]. However, many researches demonstrated that the chitosan was a more efficient inhibitor against Gram-positive compared to Gram-negative microorganism in their experimental results [64-65].

The use of chitosan an antibacterial finish of textile materials is discussed in the next section.

ANTIBACTERIAL FINISHING OF TEXTILES WITH CHITOSAN

In the textile industry, there is a growing demand for new medical textiles and textiles related to health care. The development of research in this field was largely due to the events associated with the rapid propagation of dangerous pathogens that may cause epidemics. Until now, many chemicals have been used in antimicrobial textiles such as phenols and thiophenols, antibiotics, heterocyclic compounds with anionic groups, nitro compounds, urea, formaldehyde and amine derivatives, which show bactericidal effect, but in most cases are toxic to humans and harmful to the environment. That is why the textile industry is looking for new, ecofriendly solutions that are non-toxic to people and are safe for the environment. The most studied natural polymer for antimicrobial finishing of textiles is chitosan. ChSN is a wide-spectrum biocide with high antimicrobial efficacy against both Gram-positive and Gram-negative bacteria, as well as fungi and yeasts [67]. Chitosan inhibits the growth of a wide variety of bacteria and fungi, showing broad high bacteria killing rate and low toxicity toward mammalian cells [68].

In textile field, the most common way to apply chitosan to fabrics is by wet thermal curing, involving relatively high temperature with energy consumption, costs and possible fabric degradation; moreover, the addition of toxic reagents, such as glutaraldehyde, is requested as cross-linking agent. In recent research works, ultraviolet radiation in the presence of a suitable photo initiator was proposed as an alternative process to graft chitosan molecules to textile fibres by radical process [69-70]. In detail, in case of a cellulosic substrate such as cotton, the same cellulose molecule can be involved in the reaction by the formation of radicals, which can react with those formed on the chitosan molecule, conferring a strong fastness to the finishing. Moreover, UV grafting is a fast and eco-friendly process, carried out at room temperature, with lower cost than the traditional thermal process. Cotton, silk and synthetic fabrics were considered as substrates. Obtained results showed that chitosan UV curing yielded strong antimicrobial properties, reaching near 100 % reduction on all considered fabrics. In order to have a good treatment fastness, chitosan has to be diluted with acetic acid solution before spreading on fabrics and an impregnation time of 12 h at an ambient temperature or 1 h at 50 °C is necessary before the curing to ensure a good penetration inside the fibres [71]. The homogeneous distribution of chitosan on fabrics was confirmed by dyeing tests with an acid dye and by SEM analysis (Figure 9), which showed the optimal distribution of the finish on single fibre surface.

Erdogan [73] studied the antibacterial effectiveness against *E. coli* ATCC 8739 of cotton fabrics coated with crayfish and shrimp chitosan/nanosilver

composite. The antibacterial efficacy of chitosan/nano-Ag coated fabric samples against *E. coli* ATCC 8739 was tested by the JIS L 1902-2015 standard test method (it is a quantitative method used to test the ability of antibacterial finished fabrics to inhibit microbial growth and kill microorganisms). The crayfish and shrimp chitosan formed a colorless film and coated the nano-Ag particles homogeneously on the cotton fabric. Antibacterial activity values were calculated as 3.10 and 5.74 for crayfish and shrimp chitosan coated cotton fabrics and as 5.37 and 5.10 for crayfish and shrimp chitosan+nano-Ag coated cotton fabrics, respectively. Their experimental results showed that chitosan/nano-Ag coating exhibited a good antibacterial activity (99.99 % reduction) against *E. coli* ATCC 8739 and can be used in the manufacture of garments such as medical textiles, baby clothes and underwear.

Several textile fabrics made from natural and synthetic materials were coated with ChSN to achieve antimicrobial property. Table 3 summarizes results gathered from literature of textile substrates treated with chitosan to impart them antibacterial activity.

CHITOSAN AS AN ANTIVIRAL AGENT

A normal textile fabric does not have any antiviral properties but the incorporation of chitosan into textiles can make them antiviral. ChSN make the treated textiles antiviral by employing either of the following two mechanisms or by combining them. In the first mechanism, the applied biopolymer makes the surface energy of a textile surface relatively low. Doing so will stop the viral transmission via textiles by restricting viruses to the textile surface. This low surface energy of textiles can also destroy the outer lipid barrier of a virus which will make the viral genome inactive by making it unable to penetrate into a host cell [81]. In the second approach, when a virus comes in contact with a textile surface treated with ChSN, the latter bind with the outer layer of the virus and inhibit its vital mechanisms. Chitosan oxidize and dissolve the lipid or the glycoprotein layer and enter inside the virus structure and adhere to the genome (i.e. with the virus DNA or RNA) and deactivate the same by breaking it into fragments. As a result of these interactions, the disintegration of the virus ensues, manifesting itself in the leakage of the viral genome and a loss of infectivity, leaving the viral particle inactive on the treated textile surface [82]. A schematic diagram explaining the antiviral mechanism through virus destruction (i.e. through the second approach) is shown in Figure 10.

To date, the incorporation of chitosan into the textiles can be done at different stages and in different ways. ChSN antiviral finishing on the textile material can be done onto the textile fibre, yarn and fabric depending upon the application technique.

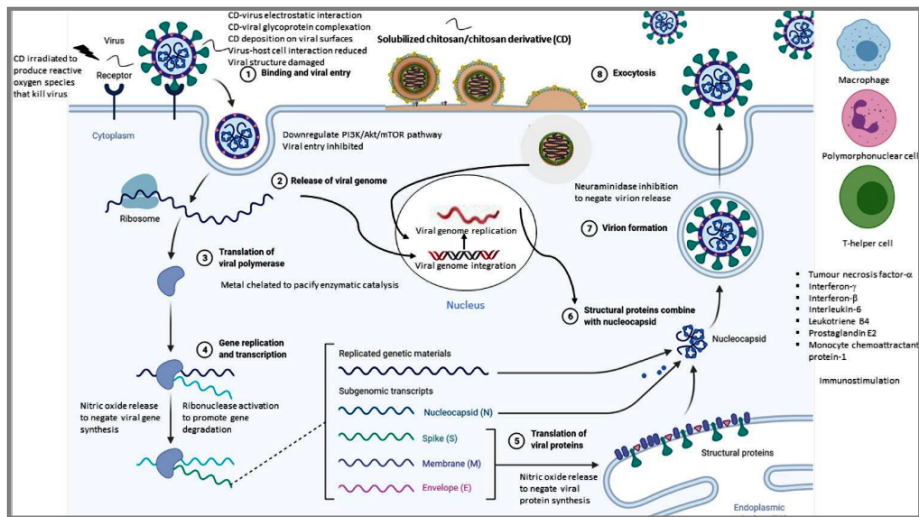


Figure 7. General antiviral mechanisms of actions of the chitosan and its derivatives [50].

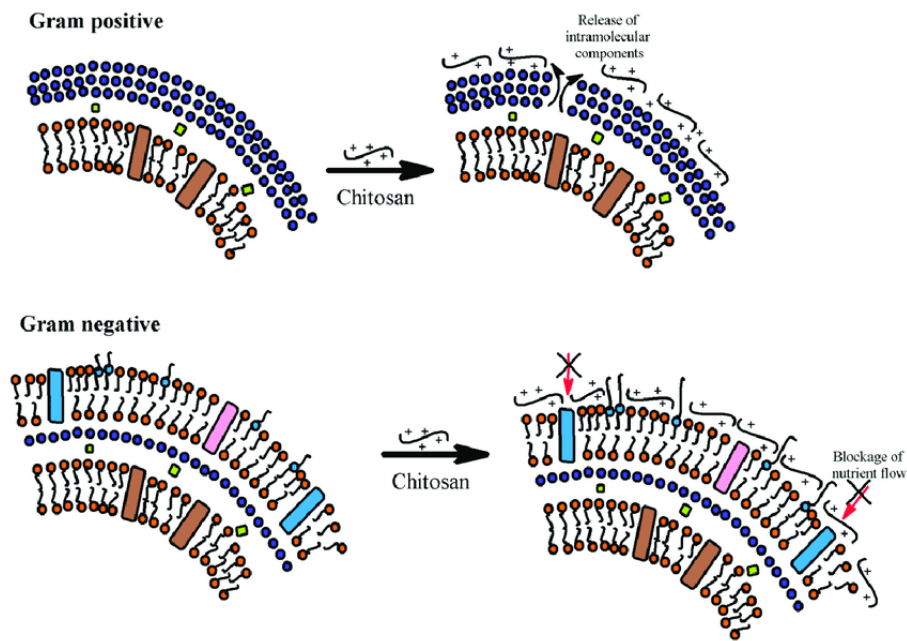


Figure 8. Action modes of chitosan on Gram positive and Gram negative bacteria [66].

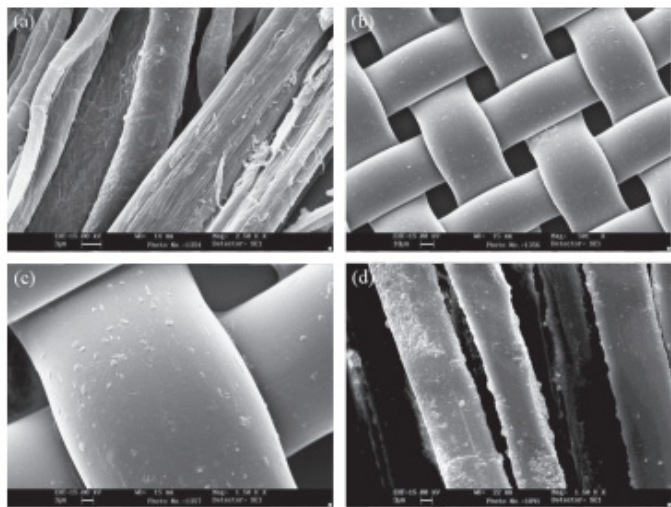


Figure 9. Chitosan UV grafted to fibres: cotton (a), PET (b), PA (c) and silk (d) [72].

Table 3. Antibacterial efficiency of chitosan treated textiles.

ChSN material	Substrate	Bacteria	Efficiency	Test method	Reference
ChSN/ZnO nanoparticles	Cotton	S. aureus E. coli	98.48 % 99.88 %	Shake flask method (ASTME2149-01)	[74]
ChSN	Silk	E. coli ATCC 11229	100 %	ASTM E 2149-01	[75]
ChSN/Copper nanoparticles	Polyester	E. coli ATCC 25922 S. aureus ATCC 25923	99.9 % 99.9 %	AATCC 100-2004 AATCC 100-2004	[76]
ChSN nanoparticles	Cotton	B. subtilis ATCC 6633 Proteus ATCC 33420	15 mm 15,2 mm	Disk diffusion method	[77]
Acidic ChSN solution	Viscose	S. aureus E. coli Candida glabrata (fungi)	97 % 94 % 85 %	ASTM E 2149-01	[78]
Chitosan/propolis Composite	Wool	S. aureus Pseudomonas aeruginosa Aspergillus niger	90 % 80 % 78 %	Plate count method	[79]
ChSN/Cu nanoparticles	Polyester/Cotton (50 %/50 %)	S.aureus E. coli	3 mm 3 mm	Disk diffusion method AATCC 100–1999	[80]

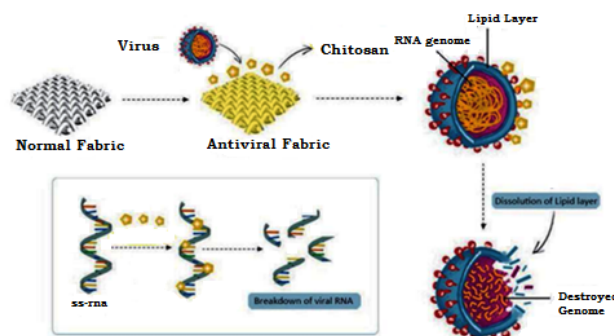
Table 4. Chitosan based textile materials and antiviral activity efficiency

Type of chitosan	Substrate	Application method	Virus type	Efficiency	Reference
ChSN/Carboxymethyl-cellulose composite	Polyamide fabric	Layer by Layer coating	Coronavirus MHV-3	99,99 %	[85]
hydroxypropyl chitosan / O-carboxymethyl-N, N, N-trimethyl ammonium chloride chitosan	Cotton fabric	press-rolling process	Ebola hepatitis	99 % 99 %	[86]
ChSN Layer	Polypropylene face mask	Middle Layer	Enterovirus (EV71)	97.95 ± 0.61%	[87]
ChSN solution	Poly(lactic acid) face mask	Coating	HCoVOC43	97.9 %	[88]
ChSN/Citric acid solution	Cotton fabric	Impregnation	Herpes simplex virus1 (HSV-1)	99.9 ± 0.50 %	[89]
ChSN/ Sialyllactose	Fibers	Wet spinning	influenza virus (H1N1)	99.6 %	[90]

For ease of application and continuous production, antiviral application onto the fabric is preferable. For fabrics like cotton, wool, silk, and other manmade fibers, ChSN antiviral treatment is done by surface treatments, like the exhaust method, the pad-dry-cure method, and the coating technique, alone or in combination, depending on the antiviral composition and the fabric quality. Chitosan biopolymer can also be incorporated in manmade fibers before the fibre extrusion (Figure 11). The exhaust method is the most common and popular method of chitosan finishing of textile materials. This method is also well suited for the antiviral finishing of natural fibres and polyamide fabrics. The process is very similar to the dipping technique and application is carried out below the boiling temperature. For polyester fabric, the high-temperature exhaust method is preferable. The process is very similar to disperse dyeing and the application is carried out at 120-130 °C. The pad-dry-cure method is another way of applying ChSN onto the textile material. The fabric is padded with the ChSN composition in an aqueous medium along with a suitable binder, followed by drying and curing for proper fixation.

Several research works reported the use of chitosan as an antiviral agent in textile materials. Examples of these studies are highlighted in Table 4.

Nevertheless, it is well known that various chemical and physical procedures are used in the preparation of chitosan from chitin. This processing significantly affects the final chemistry of the chitosan and the product quality. The variability of ChSN sources and processes of preparation has led to production of wide range of chitosan polymers with different physicochemical properties, such as differences in degree of deacetylation, molecular weight, crystallinity, and residual ash and protein contents. Such variability in chitosan physicochemical properties could lead to inconsistent and conflicting reports regarding its antiviral performance [91].

**Figure 10.** Antiviral mechanism through virus destruction on ChSN treated textile fabric [83]

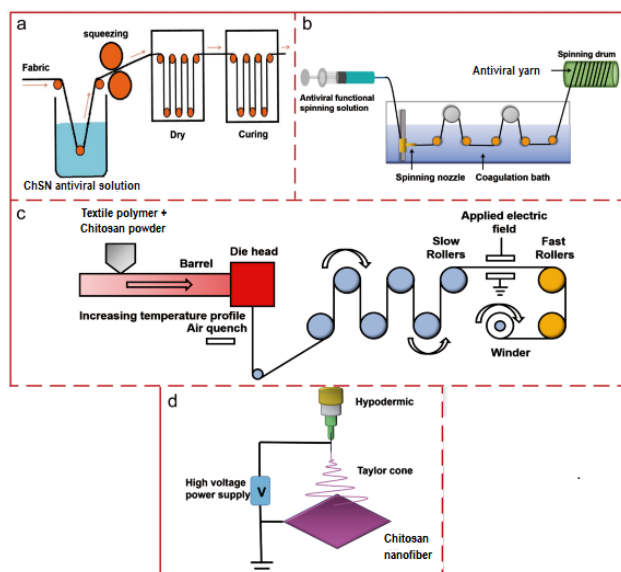


Figure 11. Methods of processing chitosan based antiviral textiles: (a) Antivirus textile soaking and rolling process, (b) Preparation of antiviral yarns by wet spinning, (c) Antiviral ChSN textiles prepared by melt spinning, (d) Equipment for electrostatic spinning of ChSN nanofibers [84].

ECO-FRIENDLY DYEING WITH CHITOSAN

The dyeing process is a major step in fabric processing, especially for consumer esthetic test [92]. All dyeing methods require quite high energy, large amount of water and salts until the dye molecules distribute and diffuse uniformly. In addition, these processes release a high amount of complex effluents containing all chemicals together [93]. Therefore, the loss of untreated dye and salt and highly efficient effluent treatment all have very high equivalent cost. Moreover, the drawbacks also impact on aquatic environment-related health. The use of chitosan or modified chitosan has beneficial effects on dyeing process which reduces the amount of untreated dye in the bath without using any salt. As a result, a simple effluent treatment would be enough to process released effluent from dye house [94]. Chitosan contains a high proportion of amino groups which provide more dye sites for anionic (reactive, acid and direct) dyes to be absorbed by textile fibers and fabrics through Van Der Waals forces and electrostatic attraction. Therefore, ChSN treatment can improve tinctorial properties of textiles such as affinity to anionic dyes, color strength and fastness [95-96] in salt free dyeing baths [97].

Cotton fibers are extensively used in fabric production because of their exceptional characteristics of comfortability, air permeability, biodegradability, no static electricity build up and excellent hygroscopicity. Reactive dyes are usually used to dye cotton fibers owing to their brightness, diversity of colors, good wet fastness and easier applicability [98]. When applied to a cotton fabric in an alkaline environment, the dye reactive group forms a bond linking dye molecule and

that of the fiber. Therefore, the dye form part of the fiber and will be less prone to removal by washing as compared to other dyestuffs which stick to the fabric by adsorption [99]. Conventional dyeing process of cotton fabric with reactive dyes requires the use of large amount of electrolyte as exhausting agent [100]. These electrolytes lead to enormous environmental problems because they are neither exhausted nor destroyed thus remaining in the discharged dye liquor [101]. To reduce the use of electrolytes in dyeing, different options have been explored. These options include surface modification of cellulose fabrics by reacting with compounds having cationic groups or managing dye and salt addition during the exhaustion process [102]. The surface of cotton fiber can be modified to increase dye-fiber interactions by introducing cationic sites either by aminization or cationization. This will overcome the lack of affinity of reactive dye to cotton fiber, therefore providing the possibility of salt free dyeing [103]. One such approach is cationization of cotton with $-NH_2$ natural amino groups of chitosan (Figure 12).

Ashenafi et al. [105] modified the cotton fabric surface with chitosan through cationization reaction. Then, chitosan modified textile was dyed with Reactive red-31 dye without salt and compared with the conventional dyed cotton. Chitosan modified cotton showed 13.1 % improvement in dye exhaustion, 10.6 % in dye fixation and 21.5 % in the total dye utilization as compared to the conventional sample. Also, the color strength of the modified cotton fabric was better than that of conventional sample which is 18.88 and 18.02 respectively. In addition, both the dyeing time and temperature were reduced by 10°C and 20 minutes, respectively in the cationized cotton dyeing technique. Compared with untreated cotton dyed with a conventional dyeing procedure, the chitosan modified cotton fabrics dyed without electrolytes have adequate and quite comparable colorfastness to

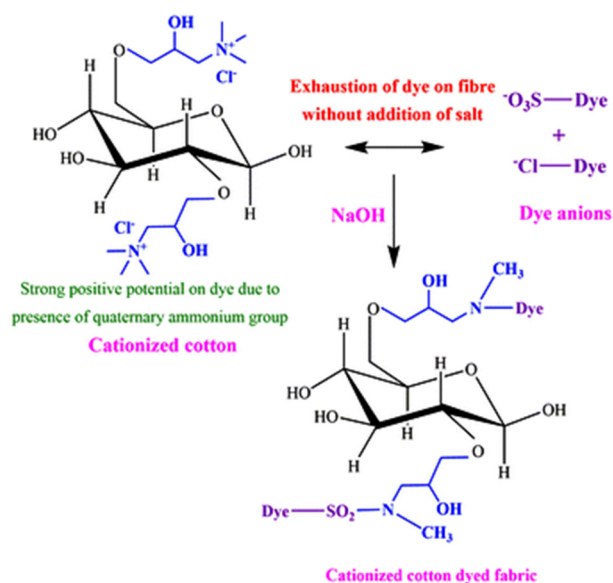


Figure 12. Dyeing mechanism of reactive dye on cationized cotton [104].

washing, dry rubbing and wet rubbing fastness but slightly reduced light fastness. Besides, the chitosan modified dyeing process greatly reduced the amount of water usage and time required to adequately rinse and remove hydrolyzed reactive dye. Similar results were recently obtained by Rehman et al. [106] when they dyed chitosan treated cotton with five ultimate reactive dyes: Drimarene Blue, Drimarene Red, Drimarene Yellow, Drimarene Lemon and Drimarene Fuschia. Their results showed an increase in the shade depth (measured by color strength K/S) of chitosan treated samples as compared to untreated samples. Color fastness properties like washing fastness and rubbing fastness were also improved because of greater dye fixation. But, light fastness was not significantly affected because it depends upon the inherent dye structure hence it remained almost the same.

In another work, Karthikeyan and Ramachandran [107] carried out salt-free reactive dyeing of cotton fabrics pretreated with emulsions of chitosan nanoparticles. Nano-emulsions with different chitosan concentrations: 0.01 %, 0.05 %, 0.1 %, 0.3 % and 0.5 % (w/v) were used for cotton fabrics padding at a pick up weight of around 80 %. Then, nanochitosan treated cotton samples were dyed in a dye bath containing 3 % of CI Reactive Red 6. They concluded that when the nanochitosan concentration increases, the dye uptake also increases and the cotton fabric treated with 0.5 (w/v) chitosan nanoparticles had higher K/S values. Moreover, the crocking and washing fastness values of all such dyed samples are quite satisfactory and slightly superior to those of conventional dyed sample. This was probably due to the fact that cotton fabrics coated with nanochitosan have uniform depth of shade after dyeing because chitosan can cover the neps formed in immature cotton fibers.

According to Aitova [108], the surface characteristics of wool fabric depend on the various types of treatments, such as plasma, enzymatic and chitosan treatments. All these processes induce high dyeing efficiency and shrink-resistance in wool fabric. For example, the plasma, enzyme and ChSN combination give the best K/S values compared to untreated dyed fabric. Since plasma pretreatment modifies the surface, enzymatic treatment can remove the hydrophobic layer of wool more efficiently, which makes dye migration higher after the application of chitosan.

Polyester fibers show a hydrophobic character and swell to a very small extent in water making access by dyestuff molecules to the fiber difficult. This together with the absence of active chemical groups makes it difficult to dye polyester with dyestuffs other than disperse dyes. In an attempt to dye polyester with reactive dye, Najafzadeh et al. [109] coated alkali treated polyester samples with nano-chitosan. Then surface modified fabrics were dyed with Bezactive green S-4B reactive dye. They affirmed that

increasing nano-chitosan content resulted in higher dye adsorption. Dyed polyester fabrics showed good dry-rubbing and washing fastness and moderate wet-rubbing fastness. According to these results, reactive dyeability of polyester at low temperatures in the absence of a carrier provides a more environmentally friendly alternative to traditional polyester dyeing methods.

Several other studies reported the use of chitosan and its derivatives to improve the dyeability of textile fabrics with anionic dyes. These interesting research works are summarized on Table 5.

However, in general synthetic dyes are allergic, carcinogenic and detrimental to human health [120]. In addition, synthetic dyeing processes involve the use of huge quantities of hazardous chemicals. For these reasons, the interest in use of non-allergic, non-toxic and eco-friendly natural dyes (such as Roselle, Lawsonia inermis L, Cochineal, Curcuma and so others) for textile applications is growing these last years as a result of the stringent environmental standard imposed in many countries. But, it is well known that the majority of natural dyes generally show very low affinity toward textile fibers and possess poor color fastness properties [121]. To overcome these problems, chitosan biopolymer can replace the salts such as ferrous sulphate, sodium sulphate, sodium carbonate and sodium chloride which have been widely used for dyeing of textiles with natural dyes to improve the fastness properties and dye absorption rate. Textile dyeing with natural dyes using biocompatible and biodegradable modification agents such as chitosan is the cost effective environmental friendly approach in the field of dyeing industry and emphasized that the modification of the fabric is one of the best routes to improve the affinity between dye and fabric [122].

In the recent work of Verma et al. [123], chitosan coated cotton fabric was dyed with the natural dye Marigold. They found that ChSN treated samples exhibited high percent dye absorption (50.25 %), good colour strength (10.51) and acceptable wash fastness grade (4). In another work, Mansour et al. [124] investigated the use of chitosan to improve the dyeability of cotton fabrics with Hibiscus sabdariffa L. (Roselle). The results of this study highlighted that cotton fabrics treated with ChSN have better depth of shade (K/S = 11) than those untreated fabrics (K/S = 4) dyed with Roselle. The cotton fabrics treated with chitosan not only provide better depth of shade but also give better wash and light fastness than those of the untreated fabrics dyed with Roselle.

In order to dye cotton fabric with natural dye sodium copper chlorophyllin (SCC), Zhao et al. [125] used ChSN as a double-layered fabric coating to serve as bio-mordant. They found that the maximum q_e (adsorption amount) of SCC calculated from the Langmuir isotherm model was raised from 4.5 g/kg to 19.8 g/kg. The dye uptake of the treated fabric was

improved from 22.7 % to 96.4 % at 1 % dye concentration. By a second chitosan layer cured on the dyed fabric via the cross-linking method the wash fastness of the cotton fabric dyed with SCC can be improved from 3 to 5.

In another research, Ke et al. [126] investigated the application of Cochineal natural dye on cotton fabrics treated with chitosan. Their experimental results showed that treating cotton with 15 g/L ChSN and baking at 130 °C offered the higher Cochineal adsorption. Dyeing kinetics curves indicated that with the increase of the dyeing temperature, dye diffusion coefficient increased and half dyeing time decreased. The washing and rubbing fastness of the treated cotton fabrics were all improved and rated above grade 3. Cochineal was also used by Safapour et al. [127] to dye wool yarns using chitosan-cyanuric chloride (ChSN-Cy). Results showed that ChSN-Cy (15 % on weight) treatment of wool had several beneficial effects which brought about remarkable improvement in dye uptake, decrease in optimum dyeing time and temperature, elimination of metal mordant from dyeing process due to bio-mordant role of ChSN-Cy, and development of antibacterial finish against pathogenic gram negative *E. coli* and gram positive *S. aureus* bacteria which was durable up to 20 washing cycles.

In an interesting previous study, Bhuiyan et al. [128] investigated the multifunctional treatment of jute fibers with chitosan and its effect on dyeing with the extract of the leaves of henna, commonly known as *Lawsonia inermis* L., and antibacterial characteristics of obtained jute fibers. The detailed study has demonstrated the double effects of ChSN on jute fiber. The treatment of jute with chitosan can appreciably enhance the uptake of dye by the fiber. Moreover, the color fastness property of dyed fabrics against washing and rubbing also exhibits within the acceptable range of good to excellent. On the other hand, the antimicrobial activities of jute fabric increase significantly due to the combined effect of natural dye henna and ChSN.

TEXTILE WASTEWATER TREATMENT BY CHITOSAN AND ITS DERIVATIVES

Textile wet processing operations (mainly dyeing and printing) produces high volumes of effluent wastewater often containing residual amounts of dyes. These dyes are extremely poisonous and potentially toxic together with a wide range of industrial polluting agents and they are linked to environmental contamination and fatal diseases. Raw or modified chitosan and their blends/composites have gained the focus of modern research for dyeing removal due to their excellent biosorbent properties, such as cationic and macromolecular structure with high adsorption capacity. Several dyes degradation methodologies using chitosan and its derivatives have been

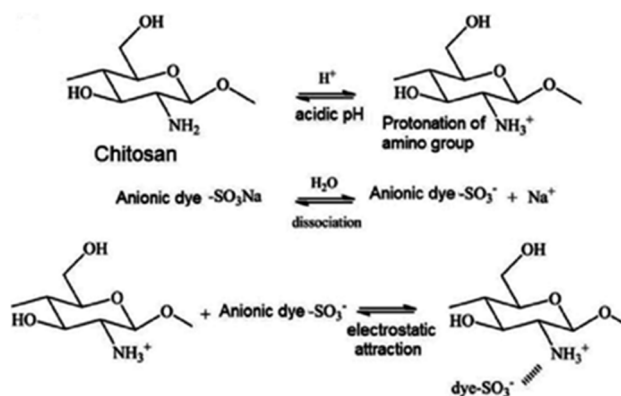


Figure 13. Adsorption mechanism of anionic dyes onto chitosan surface.

investigated including physicochemical (such as adsorption, coagulation-flocculation and separation/filtration) and biochemical (like bioremediation and bacterial degradation) processes [141]. These techniques were based on different adsorption mechanisms such as ion-exchange, complexation, coordination/ chelation, electrostatic interactions, acid-base interactions, hydrogen bonding, hydrophobic interactions, physical adsorption and precipitation [129].

The literature survey has indicated that raw chitosan was used in different physical forms in dyes removal, such as powder (particles and nanoparticles), hydrogels, beads, films and non-woven mats. Table 6 is a summary of interesting studies on dyes removal from textile effluents by different forms of chitosan and obtained maximal adsorption capacity (Q_m).

As we can see on table 6, the order of dye adsorption capacity by chitosan is acid > direct > reactive > basic > disperse dyes. In fact, it is well confirmed that chitosan is very effective for the adsorption of anionic dyes, especially in an acidic environment, following the ion exchange mechanism illustrated on Figure 13. However, the low adsorption capacity achieved in the case of basic and disperse dyes is attributable to unfavourable electrostatic interactions between the positively charged chitosan and positively charged dye molecules.

Although chitosan exhibits great potential as a dye adsorbent, it suffers from several limitations, such as high crystallinity and low hydrophilicity [138]. Chemical modification of chitosan molecules by crosslinking reactions (uniting the macromolecular chains with each other) or grafting (inserting functional groups) leads to the formation of chitosan derivatives with superior properties (enhancement of adsorption capacity and resistance in extreme medium conditions, respectively) [139].

The grafting of carboxyl amine and sulfur groups to ChSN has been regarded as an interesting method for improving some of the important specifications for enhanced dye adsorption, e.g. improving mechanical strength and its stability in acidic medium and increasing porosity as well as increasing surface area

[140-142]. The grafting of various functional groups onto the hydrogel network or the chitosan backbone can also improve chitosan's removal performance and selectivity for dye molecules and also used for controlling diffusion properties. The presence of new functional groups on the surface of the materials results in an increase of surface polarity and hydrophilicity and this enhances the biosorption of polar sorbates and improves the biosorption selectivity for the target dyes. Several cross-linking agents, e.g. epichlorohydrin ECH [143], sodium tripolyphosphate TPP [144] and glutaraldehyde GLA [145] revealed better results in preparing chitosan beads. Other cross linkers were also used to modify chitosan such as ionic liquids [146]. However, the adsorption capacity of this class of biosorbents remains not broad-spectrum: the amine and hydroxyl groups of ChSN indeed act as active sites for trapping mainly anionic pollutants while the effectiveness towards cationic dyes is rather scarce due to adverse electrostatic interactions. To overcome this problem, de Luna et al. [147] recently developed new composite chitosan-based hydrogels containing hyper-cross-linked polymer (HCLP) particles. HCLP particles were prepared by a modified Davankov procedure, consisting in the radical bulk polymerization of the poly[divinylbenzene (DVB)-vinylbenzyl chloride (VBC)] precursor followed by hyper-crosslinking of the obtained product by Friedel-Crafts reaction. Then, ChSN/HCLP hydrogels were obtained by phase inversion method in order to efficiently combine the dye biosorption ability of chitosan and the capacity of the porous particles of trapping dye molecules. Batch biosorption experiments revealed a synergistic effect between chitosan and hydrogels and the samples are able to remove both anionic and cationic dyes.

Results in terms of maximum adsorption capacities Q_m (in mg/g) for dyes removal obtained on different cross-linked chitosan are compiled on Table 7.

In last few years, a new generation of chitosan hybrid materials (composites and nanocomposites) have been synthesized and achieved improvement in adsorption properties. Chitosan was blended with other particles such as clay, graphene, dioxide titanium (TiO_2), halloysite, zeolites, magnetite, Zinc oxide (ZnO) and obtained composites gave excellent adsorption properties. Notably, the materials listed above are also well known adsorbents for removing wide range of hazardous compounds including dyes.

Therefore, composites of these materials with chitosan have often resulted in improved mechanical and chemical stability with large surface area which made them desirable adsorbents for large-scale dyes removal treatment.

In another work [162], Zhang et al. prepared zirconium-based chitosan microcomposite (Zr-ChSN) and employed as an efficient adsorbent for the removal of Orange II dye from aqueous solution. Equilibrium isotherms showed a good fit with the Langmuir isotherm and the maximum adsorption capacity was calculated as 926 mg/g indicating that the Zr-ChSN microcomposite exhibited excellent efficiency for the removal of Orange II dye. The mechanisms of adsorption were attributed to electrostatic attraction and exchange reaction between Zr-ChSN and dye $-SO_3$ anions.

It is observed that the adsorption properties of chitosan are drastically improved when it is made composites with metal oxides. For example, $Fe(OH)_3$ -loaded chitosan beads were developed for removal of anionic Congo red and methyl orange dyes from the solution [163].

Table 5. Some applications of chitosan in anionic dyeing of textile materials.

Textiles	Dyes	Improved dyeing properties	References
Wool	Reactive : Thioxo-4-thiazolidinone	dyeuptake Fastness (light, rubbing and perspiration)	[110]
Viscose	Acid dyes : Red N-HFS and Blue N-HFS	Dye absorption Color pick up %	[111]
Cotton/Nylon 50:50 wt. %	Acid : Red 138	Shade depth, Color strength	[112]
Polypropylene	Acid : CI acid red 151	Dye absorption	[113]
Cotton	Reactive: C.I. Reactive red-31	Color strength Dye exhaustion Fastness (light, wash, rub)	[114]
Viscose	Acid Orange 7 and Methyl Red	Color strength Fastness to light, rubbing And perspiration	[115]
Silk	Reactive: Cibacron Blue FN-G	Color fixing rate Dyeing depth Fastness (wash, rub)	[116]
Jute	Reactive : Novacron Red FN2	Dyebath exhaustion Color strength	[117]
Polyamide 6	Reactives : CI Red 198 and CI Black 5	Dye uptake Fastness (wash, rub)	[118]
Polyester/cotton 50:50 wt %	Reactive : CI Red 198	Color intensity Perspiration fastness	[119]

Table 6. Application of chitosan as adsorbent for textile dyes.

ChSN form	Dyes	Qm [mg/g]	References
Particles	Astrazon GTLN Red: Basic	58.8	[130]
	Telon B Red: Acid	144.9	
	Remazol Yellow RR: Reactiv	98	
	Scarlet Red 23: Direct	142.8	
Powder	Acid Blue 9	210	[131]
Nanoparticles	Direct Blue-86	1097±55	[132]
	C.I. Reactive Blue 21	302±18	
Film	Tartrazine Azo	413.8	[133]
	Amaranth Azo	278.3	
Hydrogel	C.I. Reactive Black 5	122	[134]
	C.I. Disperse Orange 3	40	
Powder	Indigo	6.9	[135]
Suspension	Malachite Green: Cationic	14.2	[136]
	Crystal Violet: Cationic	16.2	
	Sunset Yellow: Anionic	160	
	Acid Yellow 17: Anionic	176	
	Direct Blue 7: Anionic	1172	
Powder	Dianix orange: Disperse	50	[137]

Table 7. Adsorption capacities for dye removal with different modified chitosans.

Modified chitosan	Dyes	Qm [mg/g]	Reference
Cyclodextrin-ChSN	Methylene Blue	2780	[148]
Epichlorohydrin- ChSN	Reactive Blue 2	2498	[149]
	Reactive Yellow 86	1911	
Edetate- ChSN	Reactive Yellow 84	1883.6	[150]
	Reactive Black 5	1296.6	
Urea amonium tartrate- ChSN	Congo Red	1597	[151]
Tripolyphosphate- ChSN	Reactive Black 5	1125.7	[152]
Glutaraldehyde- ChSN	Reactive Black 5	846.9	[153]
Quaternary ChSN	Reactive Orange	1060	[154]
Polyacrylic acid- ChSN	Methylene Blue	990	[155]
Aerogel ChSN	Methylene Blue	785	[156]
Network ChSN	Methyl Violet	411	[157]
	Congo Red	621	
Fe- ChSN	Acid Red 73	206	[158]
Hyper cross linked ghydrogels - ChSN	Indigo Carmine	118	[159]
	Rhodamine 6G	78	
N-maleyl- ChSN	Methylene Blue	66.89	[160]
	Crystal Violet	64.56	
Acrylamide- ChSN	Astrazone Blue	47	[161]

The adsorption capacities were 445.32 and 314.45 mg/g for Congo red and methyl orange, respectively at a pH 3. Similarly, Magnetic chitosan-graphene oxide MCGO nanocomposite was prepared in the work of Jiang et al. [164] as a multi-functional nanomaterial for Methyl Orange (MO) dye removal. The effect of the experimental parameters including adsorbent mass, pH value, contact time and concentration of MO on the adsorption capacity was investigated. The maximum adsorption capacity of MCGO for Methyl Orange dye was 398.08 mg/g.

Nanocomposites of chitosan and ZnO or Zeolite offer also very interesting dye removal results. In the study of Abul et al. [165], Chitosan Zinc Oxide Nano composite was used as an adsorbent for removal of sample that contains two reactive dyes: Reactive Black HN and Reactive Magenta HB from textile dying industry effluent. They found that by using 2 gm of composite per liter of effluent at ambient temperature and 60 minutes time of contact, it may be possible to remove 99 % of the original colors of the effluent. Later on, the adsorption of Methylene Blue dye (MBD) from aqueous solutions was investigated by Dehghani et al. [166] using a new

composite made up of shrimp waste chitosan and zeolite as adsorbent. They found that the Freundlich isotherm model exhibited better fitting ($R^2 = 0.999$) with experimental equilibrium data than the Langmuir one ($R^2 = 0.956$). According to Freundlich isotherm model, maximum adsorption capacity of the composite was 24.5 mg/g.

Integration of chitosan with clay material led to the development of more efficient dyes adsorbent system. Ngwabebhoh et al. [167] investigated the adsorption of nitrazine yellow dye by chitosan/montmorillonite (ChSN-MMT) composite as biosorbent. Box–Behnken methodology was applied to optimize the adsorption experiments. Maximum adsorption values were determined as 144.41 mg/g at pH 5. Recently, Kafil et al. [168] synthesised chitosan/carbon nanoflowers as a novel nanocomposite for Acid Black1 (AB1) and Congo red CR azo dyes removal. Maximum adsorption capacities of AB1 and CR were found to be 259.13 and 553.12 mg/g, respectively (according to Langmuir model under initial dye concentration of 400 mg/L at pH = 6.75).

CONCLUSION AND PERSPECTIVES

Textile is one of the most usable consumer products in our daily life although industrial processes are mostly hazardous. Along textile production chain the used finishing agents and chemicals are often toxic and non-biodegradable. According to investigated research works, the use of chitosan natural polymer could offer an opportunity to develop sustainability and eco-friendly textile finishing processes. Interestingly, ChSN and its derivatives offer a myriad of opportunities as an antimicrobial, anti-pollution and antiviral agent in textile products and processes.

Nevertheless, the main problem that arises is that the majority of manufactured chitosan has different physical properties including color, viscosity, and density. In addition, purity of the product differs from one manufacturer to another and produced chitosan is marketed under various grades including industrial, medical, pharmaceutical and food.

The global market for chitin and ChSN reached a volume of \$ 4.2 billion by 2021 [169], with a compound annual growth rate of 15.4 %, intensifying the need for a search of other sources to satisfy the growing market. In fact, the raw materials for bulk production of chitosan are usually from marine sources because of the abundance of the raw materials in the form of shellfish waste from the seafood industry. The production cost for the traditional crustaceans based chitosan is cheap (10 US dollar per kg to 1000 US dollar per kg) compared with fungal based ChSN [170]. In addition, marine sources may have seasonal variations in availability, and the heavily mineralized exoskeletons make for an arduous extraction process. Therefore, deep research investigations together within industrial efforts on ChSN alternative sources, such as insect biomass, mushroom and fungal bodies and microbial biomass, and their production processes may tackle such limitations.

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