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Wastewater treatment using chitosan and its derivatives: A mini review on latest developments

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Abstract

Effluents and contaminants released from the industries are needed to be treated before releasing them to water bodies. Most common effluents from these industrial wastes are organic compounds, dyes and heavy metals. Heavy metals and their associated anions, as well as organic material, have been separated from wastewaters in industries using a variety of methods. Adsorption is an effective method for water treatment as they are less energy consuming and cost effective. Biopolymers such as chitosan, cellulose, keratin are used for the process of adsorption as they are present abundantly and recyclable. Chitosan is a deacetylated product of chitin. Chitosan and its derivatives are extremely essential due to their abundant availability, low cost, environmental friendliness, and biodegradability and can be widely applied in wastewater treatment. -NH2 and -OH groups are present in chitosan and provide chitosan an opportunity to make physical and chemical modifications. Modifications of chitosan into hydrogels and nanocomposites provide wider applications in wastewater treatment.

Keywords: chitosan; hydrogels; nanocomposites; water treatment

Introduction

Clean and safe drinking water is an essential requirement for supporting life, healthy living and it is also a basic element for domestic, industrial and agricultural usage. The drastic growth of industries has led to serious dangers of land, air, and water pollution, in which water pollution in particular due to the direct discharge of effluents of many industries such as paint industries, metal plating, food industries, pharmaceutical industries and battery manufacturing (Mohammadzadeh Pakdel and Peighambardoust, 2018*)*. Most of the effluents that get discharged into water resources are highly polluted by heavy metal ions, dyes, and organic materials and they getting mixed with groundwater creates imbalance in the ecological system. Even though the earth is covered with 70% water, 97% of it is salty water and for human use only 3% is available (Ahmad *et al*., 2019*)*. This shortage of resources of water and large volumes of wastewater produced by industrial activities highlights the need for strong research in advanced wastewater treatments. The conventional methods for wastewater treatment includes coagulation–flocculation (Cheng *et al*., 2021), oxidation (Miklos *et al*., 2018), membrane processes (Couto *et al*., 2018), evaporation (Menon *et al*., 2020), ion exchange (Zhang *et al*., 2019), electro–precipitation (Ramírez-Estrada *et al*., 2018), floatation (Nippatla and Philip, 2019) and reverse

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osmosis (Volpin *et al*., 2018). Majority of conventional methods have drawbacks and are insufficient to deal with the wastewater treatment problem. One of the efficient conventional methods are membrane processes and electrochemical processes, but since they are limited to finishing treatments or for specific effluents and have a very high cost of treatment, they cannot be widely applied in wastewater treatment. Precipitation processes are applicable for wastewater treatment but have drawbacks such as technical limitations and also produce large amounts of contaminated sludge (Desbrières and Guibal, 2018). Adsorption is a well-known technology, which is an economical and effective method for the decontamination of water (Crini *et al*., 2018). Adsorption using activated carbon is commonly used for treating wastewater, but high cost of treatment and energy consumption makes researchers investigate low–cost adsorbents such as biopolymers (Kalia and Avérous, 2011). Properties like biocompatibility, non–toxicity, environmentally friendly, biodegradability and lesser cost makes biopolymers an attractive option for water treatment. Keratin (Saha *et al*., 2019), cellulose and chitosan (Olivera *et al*., 2016) are some extensively studied biopolymers for wastewater treatment.

Chitosan is a derivative of chitin, which is a linear copolymer of β-(1-4)-linked N-acetyl-2-amino-2-deoxy-d-glucose (acetylated, A-unit) and 2-amino-2-deoxy-d-glucose (deacetylated, D-units) (Kaur and Dhillon, 2014). Numerous properties of chitosan such as biocompatibility, biodegradability, hydrophilicity, nontoxicity, and antimicrobial properties (Rabea, 2014) makes chitosan highly applicable in the pharmaceutical sector (Shariatinia, 2019). Due to the presence of several reactive groups, chitosan shows various chemical and biological properties such as viscosity, solubility in different media, mucoadhesive, optical and structural characteristics (Shukla *et al*., 2013). Effortlessness in modification, potential to form films (Liu *et al*., 2018), gels, nanoparticles (Li *et al*., 2010), microparticles (Hoseini *et al*., 2020) and beads (Ngah *et al*., 2008) as well as affinity towards metals, amino acids and dyes also makes it an important applicant in various sectors and industries as shown in Figure 1.

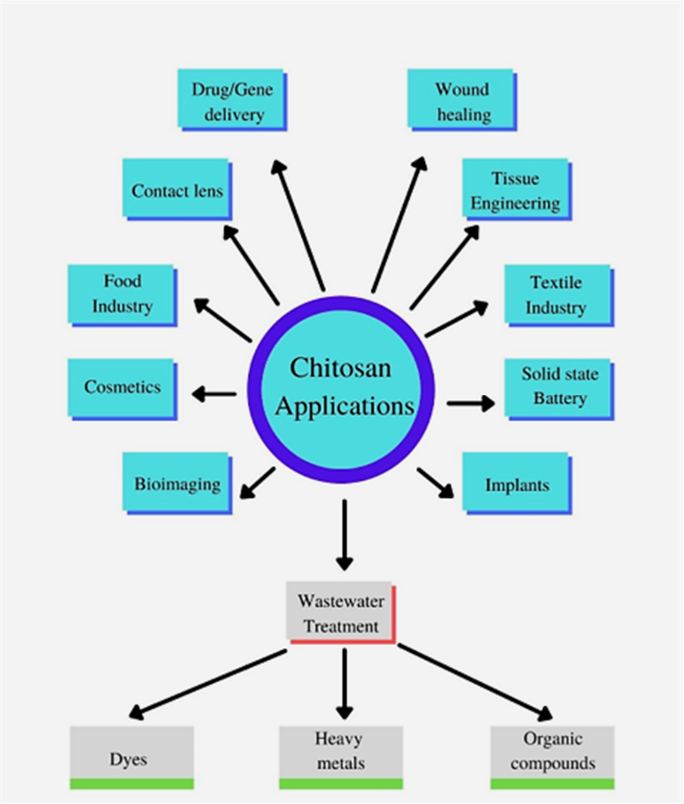

Figure 1. Applications of chitosan in various fields

In recent years, chitosan is explored as a wastewater treatment agent and several studies proved its adsorption capability and usage in treatment of textile wastewater, rice mill wastewater and egg processing industry wastewater (Thirugnanasambandham *et al.*, 2014). The cheaper cost of production, easy availability of raw materials for the extraction of chitosan along with the eco-friendly nature makes chitosan a highly potential wastewater treating agent.

Chitosan chemical properties

Chitosan is a highly basic polysaccharide which is a deacetylated product of chitin. Chitin is the second most abundant natural polysaccharide which functions as a structural polysaccharide. The molecular weight of chitosan is influenced by the degree of deacetylation where the commercially available chitosan has degree of deacetylation to an extent of 70–90% (Islam *et al.*, 2020). Chitosan varies from chitin and cellulose by its properties and N-deacetylation of chitosan makes it soluble in dilute aqueous acetic and formic acids. Solubility of chitosan is a major factor that limits its applications and the solubility in acids is due to the easy protonation of amino groups (Doshi *et al.*, 2017). The solubility of chitosan is found to be improved in neutral water by reducing its molecular weight (Fu and Xiao, 2017). The molecular weight distribution and weight average molecular weight (Mw) of chitosan are determined by HPLC and light scattering respectively (Muzzarelli *et al.*, 1987; Wu, 1988). The presence of amino (–NH2) and hydroxyl (–OH) groups which work as reaction sites makes chitosan an excellent natural adsorbent for wastewater treatment (Chang and Juang, 2004). However, the solubility of chitosan in the acidic medium and limited surface area restricts the use of pure form of chitosan as an adsorbent, so various modifications such as crosslinking, grafting, and imparting magnetic property are used to make chitosan applicable in wastewater treatment (Sheth *et al.*, 2021). Physical and chemical modifications are also made possible due the presence of reactive groups which in turn enhances the absorption potential of chitosan.

Effluents and chitosan affinity

The affinity of chitosan towards metals and proteins are explored for the possible application of treating heavy metals and organic compounds in water. Heavy metals are usually treated using physical, chemical and biological methods. Heavy metals are naturally present in groundwater in minor quantities which is required by living organisms but higher concentrations of several heavy metals are highly toxic (Mazhar and Ahmad, 2020). The heavy metals along with their routes of entry, harmful effects on human health and the standard permissible concentrations are mentioned in Table 1.

Table 1. Routes of entry, hazardous effects and standard permissible concentrations of some metal ions (Barakat 2011), (Sheth *et al.*, 2021), (Sarode *et al.*, 2019)

Metal	Route of Entry	Health hazards	Permission level (ppm)
Arsenic	Inhalation and ingestion	Cancer, irritation of respiratory system, liver and kidney damage, loss of appetite, nausea and vomiting.	0.01
Copper	Inhalation and ingestion	Cancer and hazardous effects on aquatic animals	2
Lead	Inhalation, ingestion, and absorption through skin	Brain damage, dysfunction of kidney, liver, and central nervous system	0.01
Chromium	Inhalation, ingestion, and absorption through skin	Lung damage and irritation or respiratory system	0.05
Cadmium	Inhalation, ingestion.	Kidney damage, renal disorder, human carcinogen	0.003
Mercury	Inhalation, ingestion, and absorption through skin	Arthritis, kidney disease, damage in circulatory and nervous system	0.006
Nickel	Inhalation.	Lung, liver and kidney damage	0.07

Organic pollutants are currently one of the most important environmental issues, and their removal from aqueous solutions is a major concern since they become persistent in the environment. Dyes from textile industry, pesticides and herbicides from agricultural lands, drugs, etc. are some common organic materials found in water bodies and their adsorption is influenced by the size, chemical structure, and polarity of the molecules (Vidal and Moraes, 2019). Drugs after acting on an organism will get excreted from the system in the form of urine which in turn ends up in water bodies. These accumulated pharmaceutical substances may affect the living organisms and in worst conditions create drug resistant microorganisms. This highlights the need of treating pharmaceutical products by the process of adsorption using synthetic or natural polymers (Chauhan *et al*., 2019). The presence of hydroxyl and amino groups in chitosan and its ability to form hydrogen interactions with functional groups of drugs makes chitosan a major biosorbent for the treatment of pharmaceutical contaminants (Karimi-Maleh *et al*., 2021). Other than drugs, endocrine disruptors are the other common organic contaminants and are widely studied in recent years due to its hazardous effects on human endocrine systems. In general, the process of adsorption used for removing bisphenols and chlorophenols is highly efficient and cost-effective (Sun *et al*., 2010). Other than chitosan the most commonly utilised material is active carbon, however clays or clay-based composites have lately been produced (Tsai *et al*., 2006; Hameed, 2007).

Adsorption mechanism of chitosan

Adsorption is a common water treatment method that has gained interest among environmentalists and scientists in recent years due to its ability to provide high-quality effluent treatment at a low cost of operation. It is a widely accepted equilibrium separation process that is an efficient and cost-effective method for water treatment as well as analytical separation techniques on both pilot scale and large scale (Zubair *et al*., 2020). The process of adsorption can be executed with various materials composed of biodegradable substances, organic substances, minerals, activated carbons, nanoparticle beads, and more. The most cost-effective way of using the process of adsorption is the use of naturally found organic materials such as biopolymers such as chitosan or cellulose. The entire process of using biological materials for the process of adsorption in order to remove pollutants such as metals, dyes, metalloids, etc. is called as biosorption which can include living or dead organisms and sometimes biopolymers such as chitin and chitosan (Kalyani and Hemalatha, 2016). Chitosan has certain physical and chemical features which assist them in the process of adsorption, the functional groups of the compound acts as a potential scavenger for ionic metals and pollutants in the water treatment process (Nguyen and Nguyen, 2019). The structure of chitosan has lone pair electron's on -NH2 and -OH groups which leads to a process called chemisorption which further depends and changes based on the pH of a solution (Vakili *et al*., 2014*)*. Researchers have used nanoparticles or nano chitosan for the removal of heavy metals which involves the mechanism of removal by electrostatic interaction, ion–exchange, metal chelation and also certain formation of ion pairs *(*Bhatnagar and Sillanpää, 2009*)*. Studies have shown that the uptake of ions such as Pb(II) and Cu (II) takes place by the magnetic chitosan nanocomposites with the help of -NH2 and -OH functional groups which is due to the electrostatic attraction between charged ions that are opposite in nature (Liu *et al*., 2009; Yuwei and Jianlong, 2011). In case of the removal of different dyes from wastewater materials physical adoption, dye interaction, chemical bond formation mechanisms work. The most common mechanism of adoption of acidic dyes by nano sized chitosan is due to the interaction that takes place between negatively charged dyes ions and the amino groups of chitosan (Cheung *et al*., 2009). The mechanism of absorption changes with the ph. of the solution such as if the dye is to be adsorbed from an alkaline solution the probable mechanism that takes place by the chitosan material is chelating interaction and not electrostatic interaction (Abdelmouleh *et al*., 2004). Newly discovered ways are also coming up with chitosan in current scenarios where technology such as activated carbon-based adsorption are studied with chitosan (Abdelmouleh *et al*., 2004). Different adsorption mechanisms of chitosan are shown in Figure 2.

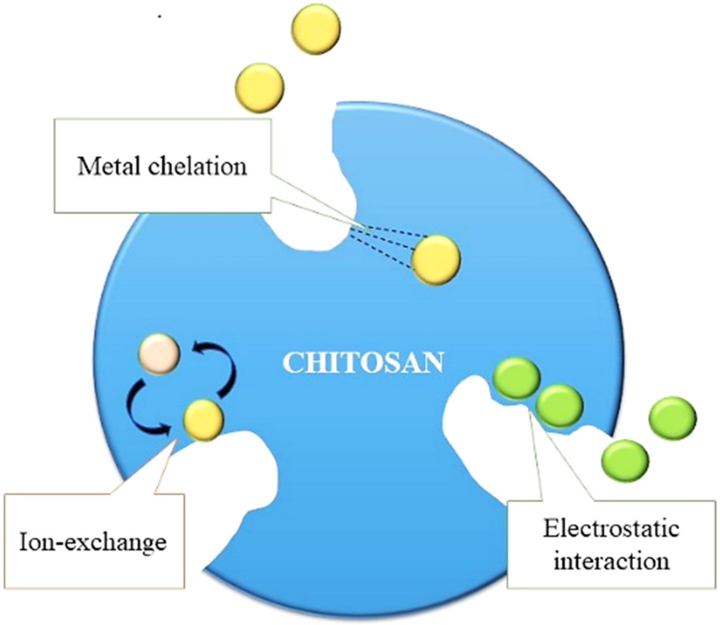

Figure 3. Various adsorption mechanisms of chitosan

Chitosan based hydrogels

The Chitosan in general has been studied for its potential uses and application in many things. The use and application of chitosan-based hydrogels are a new technique for many applications related to drug delivery, tissue engineering and wastewater treatment. The hydrogels of chitosan are prepared by moulding the physical and chemical aspects of the chitosan structure with the help of crosslinking, grafting, impregnation, incorporating of hard fillers, blending, interpenetrating and more (Kekes and Tzia, 2020). The use of these hydrogels for the water treatment has a potential use and prospects for pollution control. With the increasing number of pollutants in water bodies such as heavy metals, ions, dyes, and other organic discharges the process of biological water treatment of contaminated water has become important (Fu and Wang, 2011). Although there are different methods of treatment including osmosis, coagulation, flocculation, and more, the use of adsorption methods has higher potential than the rest of the methods. Chitosan hydrogels have advantages due to its properties of biodegradability, biocompatibility, and non-toxicity but it also comes with few drawbacks of low stability in acidic medium, less mechanical strength and also low thermal stability (Salehi *et al.*, 2016; Qi *et al.*, 2018b). Researchers have studied the use of chitosan hydrogels for the removal of toxic dyes and heavy metals from water bodies and have discovered positive results (Salehi *et al.*, 2016). The use of these chitosan-based hydrogels is for the removal of emergent pollutants. Igberase *et al.* (2014) in his study have used chitosan grafted polyaniline beads to study its adsorption properties for the removal of copper (II) from aqueous solution. Modification of hydrogels beads also have been studied for better rate of adsorption, a study was conducted which uses zirconium for the modification of hydrogel which was further used for the removal of boron (III) form wastewater (Kluczka *et al.*, 2018). There are different types of hydrogels that have been discovered including chitosan based composite hydrogels studied for the removal of dyes like methylene blue and malachite green (Kluczka *et al.*, 2018). Copolymeric chitosan-based hydrogels are also investigated recently for removal of crystal violet (CV), naphthol green, and sunset yellow from wastewater (Nagarpita *et al.*, 2017). Few other types of hydrogels have been in studies include interpenetrating chitosan-based hydrogels (Nagarpita *et al.*, 2017), blended chitosan-based hydrogels (Dergunov and Mun, 2009), and imprinted chitosan-based hydrogels (Rao *et al.*, 2006). With so many different hydrogels based on chitosan and its unique properties the prospects of nano-adsorbents for water purification have increased and also shows promising scope.

Chitosan nanocomposites

The potential use of chitosan for different industries including water treatment and purification has been studied abundantly lately. Its application in high end water purifiers is widely explored by various researchers. Chitosan being a biopolymer has shown different properties for its successful applications in the field of medicine, food technology, and marine. The characteristic properties of the compound incorporate features like biodegradability, adsorption, antimicrobial activities, non-toxicity, and more (Rinaudo *et al.*, 2020). Chitosan nano-biopolymers have a relatively new application for the adsorption of different heavy metals and dyes from water (Linghu and Wang, 2014). The use of chitosan derived nanoparticles for the adsorption of dyes, impurities and heavy metals in water has been mentioned in different literature providing the potential use in marine industry (Ngah *et al.*, 2011; Vakili *et al.*, 2014). Although there are various ways of employing Nano chitosan particles for the adsorption process, few chitosan material preparations have shown better results than other methodologies. Haider and Park (2009) in their research have used electrospinning methods for preparing nanofibers of chitosan and then were further used to adsorb Cu (II) and Pb (II) ions from an aqueous solution. The results showed adsorption of 263.2 and 485.4 mg g^{-1} for Pb (II) and Cu (II) respectively (Haider and Park, 2009). Other scientists tried to incorporate the gelation method for making Nano chitosan particles to test its adsorption for Pb (II) ions which showed maximum adsorption rate as 398 mg/g (Qi and Xu, 2004). Seyedi *et al.* (2021) explored the possibilities of chitosan particles to adsorption Cd(II) and was found to have a capacity of 358 mg/g (Zahedifar *et al.*, 2021). Other studies include the use of chitosan nanoparticles for the toxic dyes adsorption which was performed by Hu et al for-Acid Green 27 (AG27) dye of anthraquinone type (Hu *et al.*, 2006). Dyes like Acid Red 73 (AR73), Acid Orange 10 (AO10), Acid Orange 12 (AO12) and Acid Red 18 (AR18) were also tested upon by Wong *et al.* (2003). Incorporation and changes in chitosan can be done for its utilization for the absorption purpose for particular dyes. One such example is the use of zinc oxide nanoparticles incorporated in chitosan which was further used for adsorption of acid black 26 (AB26) and direct blue 78 (DB78) with capacities of 34.5 and 52.6 mg g^{-1} respectively for DB78 and AB26 (Salehi *et al.*, 2010). Major advancements on the synthesis of chitosan-based nanocomposites are mentioned in Table 2.

Table 2. Major advancements on the synthesis of chitosan-based nanocomposites and their effluents

Chitosan nanocomposite modification	Effluent	Reference
Incorporation of SWCNT/Fe_3O_4/TiO_2 in to Chitosan	Chromium (Cr), As(V), Methylene blue and Congo red	(ZabihiSahebi *et al.*, 2019)
Sawdust-Chitosan nanocomposite beads (SDNCB)	Ni (II) and Cu (II)	(Kayalvizhi et al. 2022)
Graphene oxide/chitosan (GO/CS)	Methylene blue	*(Qi et al., 2018a; ZabihiSahebi et al.*, 2019)
Chitosan/MWCNT/Fe3O4 nanofibrous composite	Chromium (Cr)	(Beheshti *et al.*, 2016)
Polyacrylic acid grafted magnetic chitosan nanocomposite	Pb(II)	(Hu *et al.*, 2020)
Chitosan/polystyrene/ZnO nanocomposite	Nitrate ions	(Keshvardoostchokami *et al.*, 2017)
Chitosan/poly(vinylidene chloride)/Ag(CS/PVDC/Ag) bionanocomposite	Antibacterial agent (E. coli and G. Bacillus) and Metal ions (Pb, Cd, Fe)	(Al-Sherbini *et al.*, 2019)
Magnetic maghemite/chitosan nanocomposite	Methyl orange	(Jiang *et al.*, 2012)
Fe_3O chitosan carbon microbeads	Doxycycline	(Bai *et al.*, 2018)
Bio-silica/chitosan nanocomposite	Textile dye	(Darvishi Cheshmeh Soltani *et al.*, 2013)

Other ways chitosan is studied and optimized for its use in impurity adsorption is the use of modified chitosan such as chitosan nanocomposites (Aliabadi *et al.*, 2014). Studies have discovered that the adsorption property of chitosan for heavy metals are based on electrostatic interaction, ion–exchange, metal chelation (Bhatnagar and Sillanpää, 2009) and more whereas in case of adsorption of dyes phenomenon such as chemical bonding, ion–exchange, hydrogen bonds, hydrophobic attractions, van der Waals force, etc. takes place (Crini *et al.*, 2008).

Conclusions

The chitosan is a linear copolymer of β-(1-4)-linked N-acetyl-2-amino-2-deoxy-d-glucose (acetylated, A-unit) and 2-amino-2-deoxy-d-glucose (deacetylated, D-units) a derivative of chitin which formed by the partial deacetylation of chitin. The active groups present on chitosan are -NH2 and -OH groups which provide chitosan the potential to undergo chemical and physical modifications. Apart from that, chitosan has other beneficial properties such as being polycationic, non-toxic, biodegradable, having a high adsorption capacity, and having antibacterial properties. The most effluents present in wastewaters are heavy metals, organic compounds and dyes. Chitosan has higher affinity for these effluents due to the presence of the reactive groups present. These effluents are treated by the mechanism of adsorption. Adsorption is the result of specific interactions between the adsorbent and the adsorbate. These interactions are strongly linked to the chitosan's chemical structure. Chitosan made into hydrogels and nanocomposites provides wider application on wastewater treatment. Wastewater treatment using chitosan and its applications have potential for further research. Chitosan-metal biosorption or interaction has recently brought about solid-state polymer batteries and electronic devices. Furthermore, more research is needed to understand the effects of different immobilisation procedures on the rate and equilibrium uptake of contaminants by immobilised biomass. Currently, biosorption research is primarily focused on removing heavy metals and organics from water, with precious metal recovery from biosorbents receiving minimal attention (desorption).

Authors' Contributions

All the authors have contributed equally to the preparation of the final manuscript. All authors read and approved the final manuscript.

Ethical approval (for researches involving animals or humans)

Not applicable.

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Conflict of Interests

The authors declare that there are no conflicts of interest related to this article.

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