Application of chitosan-based materials in wastewater treatment

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Abstract. Chitosan, as a natural polysaccharide material, plays a great role in the field of wastewater treatment due to its good biodegradability, antimicrobial activity, and other properties. However, it may have some differences in the processes of adsorption, flocculation, and membrane separation. We comprehensively describe the application effect and mechanism of action of chitosan-based materials on the above three major water treatment processes. In the adsorption process, chitosan-based adsorbents such as hydrogels and chitosan beads are prepared by cross-linking, impregnation, and the introduction of functional groups. These chitosan-based materials exerted efficient removal performance in the adsorption of heavy metals, organic dyes, anionic pollutants, etc., which was attributed to their large specific surface area and abundant functional groups. In the flocculation process, the chitosan-based materials were modified by grafting and modifying their shape, while heavy metal ions, organic dyes, and algae were removed using processes such as chelation and charge neutralization. In the membrane separation process, a number of nanofiltration and microfiltration membranes were created by coating chitosan ions, and physicochemical modification was done for the separation of heavy metal ions and emulsified oil droplets in water to improve the fouling resistance and retention rate of the membrane materials. This paper reviews the application of chitosan-based materials in wastewater treatment and proposes that in-depth studies on their toxicity and reusability should be conducted in the future, with a view to playing a role in areas such as the removal of malodorous water odors.

Keywords: chitosan-based materials, adsorption, flocculation, membrane separation, water treatment.

1. Introduction

Chitosan is a natural polysaccharide material obtained by deacetylation of chitin, which exists in the cell walls of certain fungi and can be extracted and prepared in large quantities from the shell layers of marine crustaceans (WanNgah and Isa, 1998; Rinaudo, 2006). Chitosan has a wide range of applications in biomedicine, agriculture, textiles, and environmental remediation due to its wide range of sources, safety, and non-toxicity, as well as excellent biodegradability, adsorption, and antimicrobial activity (Wan Ngah et al., 2011; Wang and Zhuang, 2022). With deepening research on chitosan and its modified materials, chitosan-based materials show great application prospects in emerging applications such as water treatment and cosmetics.

Rapid economic development has led to a serious increase in the pollution of municipal sewage and industrial wastewater with heavy metals and organic matter, which will threaten human health if not properly treated. Traditional methods such as adsorption and flocculation, although widely used, have shown certain limitations due to their high cost and difficulty in avoiding the effects of secondary pollution (Shannon et al., 2008). Chitosan-based materials play an increasingly important role in the fields of environmental protection and sustainable development due to their unique adsorption properties and eco-friendly characteristics. Pakdel et al. (2018) used chitosan to synthesize hydrogels for the adsorption and removal of pollutants, such as heavy metals, from water; Yang et al. (2016) researched the application of chitosan and its derivatives as a flocculating agent for treating different types of wastewater. We can find that chitosan-based materials is differentiated in the process of wastewater treatment and can be used as an adsorbent, flocculant, and separation membrane to remove pollutants from water bodies. However, most of the current reviews focus on the preparation scheme of the material, and very few reviews focus on the process of applying the material.

This paper reviews the current state of research on the use of chitosan in wastewater treatment, with an emphasis on the adsorption, flocculation, and membrane separation processes of chitosan-based materials for eliminating contaminants such as heavy metal ions, dyes, and nutritional salts from wastewater. It also serves as a foundation for future research on ecologically friendly composites based on chitosan and its derivatives.

2. The role of chitosan-based materials in wastewater treatment process

2.1 Chitosan-based materials as adsorbents

Adsorption is a physical wastewater treatment method with the advantages of low cost, easy operation, and high efficiency, so it is widely used in industrial wastewater treatment, domestic sewage treatment, and water source protection. Chitosan has the advantages of natural environmental protection, high adsorption capacity, low cost, and regulating performance. It can effectively adsorb heavy metal ions, organic dyes, anions, and other pollutants.

2.1.1 Adsorption of heavy metal ions

Heavy metal ions like lead, mercury, chromium, copper, arsenic, and cadmium pose serious threats to aquatic ecosystems. Leveraging its extensive specific surface area and rich amino and hydroxyl functional groups, chitosan proves adept at adsorbing these ions onto its surface and pores, effectively purifying water.

For single heavy metal-polluted water bodies, Sessarego et al. (2019) used THPS crosslinked functionalized chitosan to prepare novel phosphorus salt crosslinked chitosan (PCC) to improve its adsorption capacity for Cr (VI), which was activated for Cr (VI) adsorption at pH 6. Therefore, PCC can be used for wastewater treatment in a wider pH range than chitosan. An alginate/melamine/chitosan aerogel prepared by Gao et al. (2021) showed an optimum adsorption of Pb (II) of 1331.6 $mg\cdot g^{-1}$ at pH 5.5, which was able to overcome the competition of coexisting divalent metal ions.

For composite heavy metal-polluted water bodies, Liu et al. (2019) utilized a special SiO_2 leaf-like structure to avoid the aggregation and loss of chitosan and enhance the contact of functional groups with heavy metal ions, which resulted in the maximum adsorption of As (V) and Hg (II) by the composite materials of 198.6 $mg\cdot g^{-1}$ and 204.1 $mg\cdot g^{-1}$, with a removal rate of more than 60% in 2 min. Song et al. (2021) used a biochar/chitosan composite adsorbent for the efficient removal of Cr, Cu, Se, and Pb metals from oil sands treated water, showing high removal efficiencies for Cu (97.5%) and Pb (94.3%). Karimi et al. (2022) prepared magnetic chitosan/alumina/Fe_3O_4 nanocomposites by EDTA functionalization, with maximum adsorption capacity in the order of Cd > Cu > Zn, and the removal of Cu, Cd, and Zn ions at pH 5.3 was 99.98%, 93.69%, and 83.81%, respectively.

We can conclude that chitosan-based materials are mostly applied to the removal of complex heavy metals due to the complexity of environmental pollution and the diversity of pollutants. In the above process, most of them are dominated by chemical adsorption, and the functionalization modification makes the reaction rate of functional groups increase, thus enhancing the adsorption efficiency. Subsequently, relevant composite materials can be introduced to make up for the shortage of their physical properties.

2.1.2 Adsorption of organic dyes

Organic dyes, organic solvents, petroleum-based chemicals, and other organic debris can be found in bodies of water. Organic dyes, which are very poisonous and difficult to degrade, are mostly found in the wastewater of the paper, leather, plastics, textile, and metal sectors, and are difficult to treat using chemical and biological procedures. Furthermore, chitosan-based polymers have a high affinity for various colors as well as a high adsorption capacity.

For food dyes, Sellaoui et al. (2021) discovered that chitosan films were more effective in adsorbing FD&C yellow No. 5 dye at temperatures ranging from 298 to 328K and pH levels between

4-7. Jiang et al. (2023) synthesized a hydrophilic magnetic Fe_3O_4 embedded chitosan-crosslinked-polyacrylamide composite that had better water solubility in acidic solution, adapted to a wider pH range, and had an adsorption capacity of 359.71 mg·g^{-1} for the food dye sunset yellow, which was much higher than that of other adsorbents.

For anionic and cationic dyes, Karimi-Maleh et al. (2021) developed novel chitosan beads impregnated with 1-butyl-3-methylimidazolium bromide. These beads achieved rapid removal efficiency by removing up to 86% of methylene blue cationic dye within just 25 minutes. Chang et al. (2020) prepared a CTS/PAA/GO composite polyionic physical hydrogel where linear PAA improved swelling and adsorption properties while GO enhanced mechanical strength as well as adsorption properties (Fig. 1). This hydrogel was capable of effectively absorbing both cationic model dye MB and anionic model dye FY3 with equilibrium adsorption capacities reaching approximately 296.5±31.7 mg·g^{-1} and 280.3±23.9 mg·g^{-1} respectively.

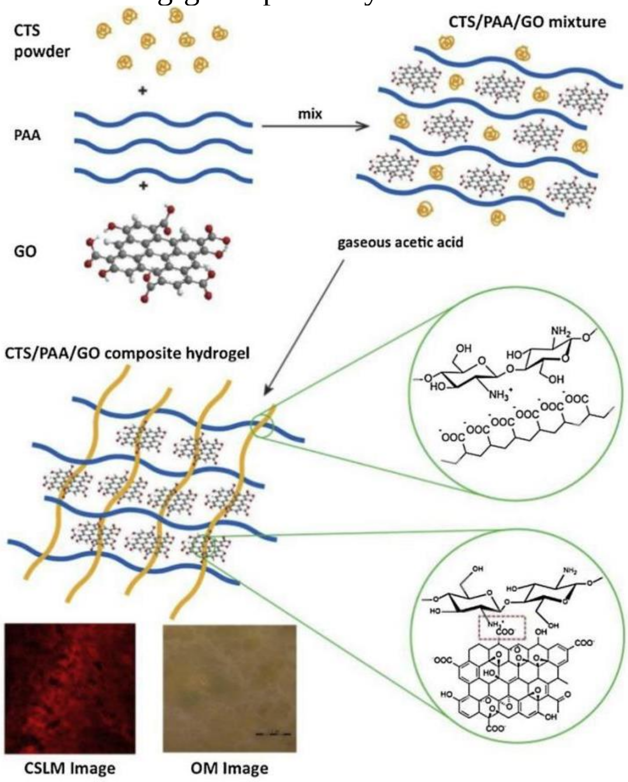

Fig.1 Formation mechanism of CTS/PAA/GO hydrogel (Chang et al., 2020).

In summary, it can be seen that there are certain limitations in the removal of organic dyes using only chitosan materials, and composite materials are needed to make up and improve their mechanical and chemical properties, which can further optimize the adsorption process and improve the adsorption efficiency and recycling rate. Subsequently, we can try to combine other wastewater treatment technologies, such as biodegradation, advanced oxidation, etc., to construct a joint treatment system of multiple methods to achieve more efficient and comprehensive wastewater purification.

2.1.3 Adsorption of anionic pollutants

Chitosan has a certain adsorption capacity for anionic pollutants in water such as cyanide, nitrate, and phosphate. The removal efficiency of anionic pollutants is influenced by various factors, such as the nature of chitosan, the concentration of pollutants, and water conditions.

For the adsorption of phosphate, Dai et al. (2020) prepared CS/CGG hydrogels from chitosan and cationic guar gum, and the adsorption of phosphate was a multilayered adsorption process with q_e of 1.27±0.01 mg-P/g, 5.57±0.01 mg-P/g, and 10.72±0.03 mg-P/g for 25 mg/L, 50 mg/L, and 100 mg/L KH_2PO_4 solutions, respectively. Xu et al. (2020) used bentonite and chitosan to prepare the adsorbent La-BC-5, which showed a maximum removal rate of 99.7% at low phosphate concentration, remained 89.1% after five regeneration cycles, and showed good immunity to coexisting anions (Fig. 2).

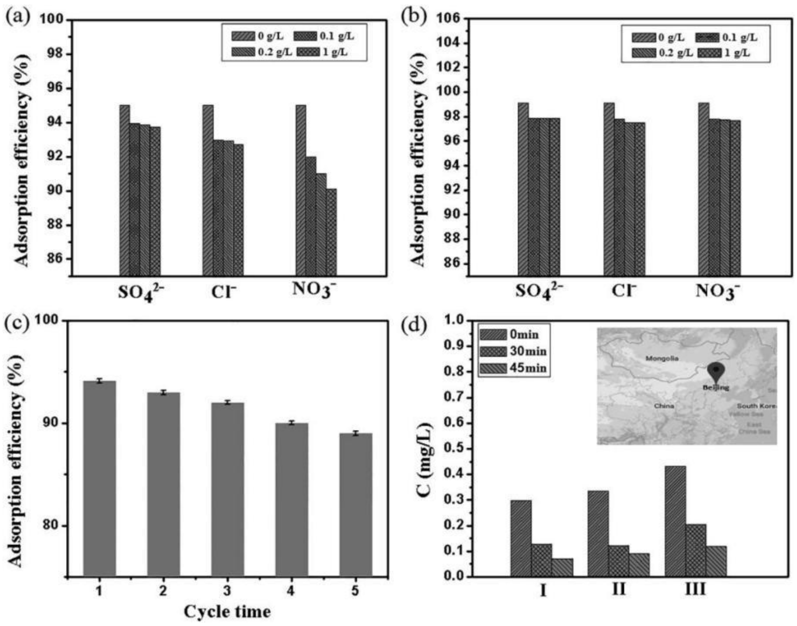

Fig. 2 (a, b) The effects of various coexisting anions on the adsorption efficiency, (c) relationship between cycle time and phosphate adsorption efficiency of La-BC-5, and (d) surface water adsorption in different sections of Huaisha River, Beijing (Xu et al., 2020).

For the adsorption of nitrate, Bian et al. (2022) prepared a novel core-shell composite (GCS@LDH), where the wrinkles on the surface of the GCS and the LDH plates provided a large specific surface area, and the hydroxyl groups and metal hydroxides of LDH synergized with the active groups of the GCS to improve adsorption, with a maximum adsorption capacity of 29.1 $mg \cdot g^{-1}$ at pH 4 and 298K and a The corresponding removal rate was 97%. Chen et al. (2020) synthesized chitosan-ethylene glycol hydrogel (CEGH) by repeated freeze-thawing, the nitrate adsorption process was affected by pH and temperature, and the polarity of CEGH was the main removal mechanism, which functioned under acidic conditions through hydrogen bonding interactions with a maximum adsorption capacity of 49.04 $mg \cdot g^{-1}$.

For specific anion-contaminated water bodies, improving the adsorption efficiency and interference resistance of the adsorbent is the basic guideline for determining the chitosan modification program. In addition, by comparing the maximum adsorption capacities of the two nitrate removal materials, the adsorption mechanism of hydrogen bonding interactions can be deeply explored with a view to improving the adsorption effect.

2.2 Chitosan-based materials as flocculants

Chitosan has chelating and adsorption effects on many substances, which can combine with suspended particles and colloidal particles in water to form larger flocs and make them coagulate and precipitate, thus realizing the removal of suspended matter. Chitosan flocculant is not only effective for solid particles but also has an adsorption effect on some organic matter and heavy metal ions.

2.2.1 Flocculation of heavy metal ions

Chitosan is rich in functional groups such as amino and hydroxyl groups that enable chitosan to form complexes with heavy metal ions and form stable precipitates in water. García et al. (2018) synthesized dendritic chitosan, which has chelating and coagulating effects, and separates seven metal ions at the same time with an optimal window of 97-140.2 $mg \cdot L^{-1}$ at pH 4.7, and the flocculation process does not affect algal lipids and fatty acids. Tang et al. (2020) Sulfated chitosan-based flocculants prepared by graft copolymerization can effectively remove dissolved heavy metals from water through chelation of sulfonic acid groups, adsorption of other functional groups, and co-sedimentation of flocs and exhibit efficient flocculation performance, thermal stability, and solubility in the treatment of hematite colloidal particles. Sun et al. (2019) prepared bifunctional chitosan-based flocculants PCA, PCAXC, and PCAAF, which removed Cu, Cr, and Ni from water by chelation and flocculation, with Cu and Cr removal rates of 85.1% and 76.1%, respectively, for PCAXC, and Ni removal rate of more than 75.7% for PCAAF. Sun et al. (2020) grafted acrylamide and thioglycolic acid onto carboxymethyl chitosan to prepare acid-resistant bifunctional MACA copolymers [CMCTS-g-P (AM-TGA)], which are rich in hydroxyl and amine groups (Fig. 3) and can be easily modified to chelate and trap heavy metals.

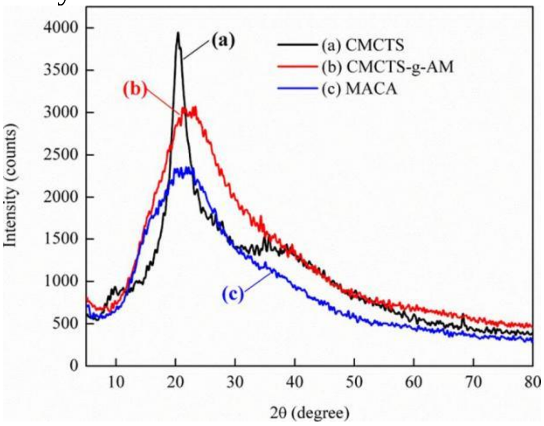

Fig. 3 FTIR characterization: (a) CMCTS, (b) CMCTS-g-AM, and (c) MACA (Sun et al., 2020).

All of the above chitosan-based materials have a variety of functional groups that can effectively remove heavy metals from water through chelation, adsorption, and flocculation and exhibit high efficiency in the water treatment process. They are characterized by low dosage and high efficiency. Subsequent research on the stability and durability of chitosan-based materials in practical applications should be strengthened to ensure their long-term sustainability.

2.2.2 Flocculation of organic dyes

The cationic nature of chitosan enables it to interact with the anionic groups of organic dyes to form flocculating clusters, which effectively aggregate and remove the organic dyes from the water. The flocculation process is relatively simple and inexpensive and has no significant effect on other components in the water.

The starch-acrylic acid-chitosan terpolymer synthesized by Xun et al. (2018) was able to efficiently remove acidic blue 113 (99.7% removal) from simulated wastewater and showed good flocculation effects over a wide range of flocculant dosages and wastewater pH. Liu et al. (2018) prepared chitosan and cerium polyferrosilicate (CTS-PFCS) flocculants for the three-dimensional chain-network structure with strip branching, where charge neutralization and chemisorption were the main mechanisms of flocculation in the treatment of direct red dye wastewater, showing superior decolorization performance and stability to PFCS for a wide pH range.

It can be seen that chitosan and its modified products as flocculants exhibit efficient flocculation performance in wastewater treatment and are suitable for a wide range of pH and wastewater conditions, with good general applicability.

2.2.3 Flocculation of algae

Chitosan can effectively aggregate algae into large flocs by charge neutralization and adsorption with the negatively charged carboxyl and sulfate groups on the surface of algae, and its flocculation effect is affected by factors such as pH, flocculant dosage, and algae species.

Nayak et al. (2018) harvested Spirulina HS_2 using four different flocculants at different concentrations and pH conditions, in which chitosan settled for 30 min at a dose of 10 $mg \cdot L^{-1}$ and pH 8.0 with the highest flocculation efficiency of 99.6%, and the chitosan-based harvested culture supernatant showed less inhibition of growth with a low dosage of flocculant and high flocculation efficiency. Augustine et al. (2019) optimized the collection process of microalgae by using chitosan magnetic nanoparticles and chemical magnetic nanoparticle composites without affecting the lipid and fatty acid composition of the algae.

Chitosan flocculant showed the advantages of high efficiency and low dosage in the harvesting and flocculation processes of algae without affecting the biochemical composition of the algae, which is good for the environment. The chitosan flocculation technology can be subsequently applied to actual algal water treatment to assess its feasibility and economy in large-scale applications.

2.3 Chitosan-based materials as separation membranes

Chitosan membranes can be applied in membrane separation processes such as ultrafiltration, nanofiltration, reverse osmosis, etc. due to their unique porosity, biocompatibility, and excellent separation properties. This material not only excels in the removal of suspended solids and heavy metal ions but can also be applied in areas such as water-oil separation.

2.3.1 Separation of heavy metal ions

Chitosan-based membranes can be modified and cross-linked to adjust their surface properties and pore structure, thus realizing the selective separation of different heavy metal ions.

Zakmout et al. (2020) developed a chitosan-coated polyethersulfone microfiltration membrane suitable for chromium recovery from tannery wastewater, with a removal rate of chromium up to 99% or more, which is far more efficient than that of cations such as Na and K. Yang et al. (2021) prepared positively charged composite nanofiltration membranes utilizing a series of gradient crosslinked chitosan composites, with a high removal rate of heavy metal ions up to 98.96% ($CuCl_2$), 99.04% ($FeCl_3$), and 98.3% ($ZnCl_2$) with more advantages in permeability and mechanical strength, which can be used to reduce water hardness and produce potable water. Hosseini et al. (2019) prepared a novel layer-by-layer cation-exchange membrane by coating a layer of chitosan-activated carbon nanoparticles on a polyvinyl chloride-based heterogeneous cation exchange membrane, which is smoother and more hydrophilic than the original membrane. It can be used to remove heavy metal ions from water by electrodialysis, and the performance of the membrane can be improved by ultrasonic regeneration.

The properties of chitosan composite membranes are obviously better than single chitosan separation membranes; not only the physical properties such as mechanical strength and membrane morphology are enhanced, but also the chemical properties such as hydrophilicity and selectivity are improved. Subsequently, in-depth research can be carried out on the regeneration performance of membrane materials to improve the reuse rate of chitosan membrane materials.

2.3.2 Separation of oil-water

Because of the special nature of oily wastewater, traditional water treatment techniques such as gravity separation, coagulation, and flotation are ineffective for the separation of emulsified oil droplets. The size of oil droplets is less than 20 μm, and membrane technology, with the development of membrane pore size from nanometer to micron, has a broad application prospect in oil-water separation.

Yagoub et al. (2019) used vacuum-assisted filtration to prepare nanoporous composite membranes consisting of cellulose nanocrystals, chitin nanocrystals, and chitosan on PET nonwovens with superhydrophobic properties, which were able to efficiently separate water-in-oil emulsions with high retention rates. Zhao et al. (2021) used oxidant-induced ultrafast cephalic cephalosol/chitosan co-deposition of natural catechols and chitosan on a porous polyvinylidene difluoride (PVDF) substrate to prepare a superhydrophobic microfiltration membrane (M-4 membrane) with ultrahigh pure water permeability and strong chemical stability, with a competitive flux 70% higher than that of the pristine PVDF membrane (Fig. 4), and the catechol/chitosan coating on the membrane surface acted as an energy barrier to the oil droplets, which significantly reduced the oil adherence. Zhou et al. (2019) reported a membrane consisting of a positively charged chitosan and a negatively charged sodium alginate of amphoteric nanofiber membranes with a strong hydrate layer, which can effectively reject various oils, including viscous crude oil, with a separation efficiency of up to 99% and also have a certain removal effect on water-soluble dyes and heavy metal ions.

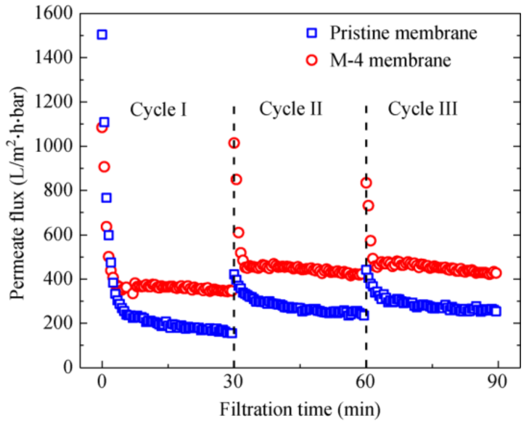

Fig. 4 Permeate flux during the three separation cycles of Tween 20-stabilized hexadecane-in-water emulsion (Zhao et al., 2021).

The discussed chitosan-based membranes display remarkable superhydrophobic characteristics, enabling effective separation of water-in-oil emulsions. This multifunctional capability is noteworthy. Conventional membrane materials, like polyvinylidene fluoride, polysulfone, and polypropylene, suffer from hydrophobicity and susceptibility to oil fouling, constraining their application. Consequently, hydrophilic-modified membranes are poised for deeper exploration.

3. Conclusion and perspectives

With the deepening of the research on the structure and properties of chitosan materials and the great attention paid by society to the issue of environmental sustainability, more and more chitosan-based materials have been applied to the field of environmental protection. Among them, in the treatment of water pollution, chitosan-based materials play a great role in the processes of adsorption, flocculation, and membrane separation due to their good renewability, degradability, and biocompatibility. This paper reviews the literature on chitosan-based materials in the field of wastewater treatment in recent years, describes the adsorption, flocculation, and membrane separation processes of chitosan and its derivatives on different pollutants, and summarizes the innovations in the preparation and modification methods. The following issues need to be further considered in future work:

- In order to determine the suitability of chitosan and its derived materials for use in contaminated waters, their toxicity needs to be evaluated.
- Multifunctional chitosan-based water treatment materials need to be designed in order to adapt to the increasing complexity of pollutants in water bodies.
- The reusability of chitosan material is an important property for its use as a green material and needs to be further investigated in recycling experiments.
- The removal of solid and liquid pollutants from water bodies by chitosan materials has been intensively studied, and its application in the removal of malodorous water odors can be further investigated.

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