



## Review article

# Chitosan, conducting polymer, and metal oxide nanocomposites: Mechanistic insights and sustainable approaches for water treatment

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## ABSTRACT

The role of nanomaterials in sustaining life and different environmental challenges related to water pollution needs to be addressed. This review article discusses the role of nanomaterials in the treatment and analysis of impure water. The focus of the review article is on the use of low-cost adsorbents, i.e., conducting polymers (CPs), Chitosan (CS), and Metal oxide nanoparticles (MO NPs) for wastewater treatment, evaluating their adsorption properties, mechanism, and feasibility for large-scale implementation. It highlights the growing concern about dye contamination in water and its removal using different adsorbents and photocatalytic agents. A comparison of nanomaterials' adsorption and photocatalytic capacity with different conventional water treatment technologies is made. This review examines the potential of CPs, CS, and MO NPs in water purification, focusing on their unique physicochemical properties that can enhance filtration systems and pollutant removal efficiency. The effect of surface modification of CS and CPs on water purification efficiency was investigated. The application of advanced technologies, including adsorption, oxidation processes, and biological treatments, was discussed in the context of their economic viability and environmental benefits. An insight into innovative and sustainable strategies for addressing water pollution, with an emphasis on cost-effective solutions and the promise of nanomaterials in revolutionizing water treatment processes, is discussed in detail. This review article highlights the innovative and cost-effective CPs, CS, and MO NPs-based strategies for wastewater treatment, emphasizing adsorption and photocatalytic efficiency to enhance pollutant removal capacity and sustainability.

## 1. Introduction

Water is considered one of the most precious molecules for sustaining life. Heavy metals are a significant environmental and public health concern due to their persistent toxicity and bioaccumulation capabilities

[1]. Control of heavy metal pollution in aquatic systems demands comprehensive monitoring and regulatory measures, along with pollution control strategies aimed at protecting marine ecosystems and human health [2,3]. Heavy/toxic metals like Arsenic, Mercury, Cadmium, Chromium, and Lead are recognized for their severe ecological

**Abbreviations:** ARS, Alizarin Red S; BET, Brunauer, Emmett and Teller; BG, Brilliant Green; BTCA, 1,2,4,5-Benzene Tetracarboxylic Acid; CAT, Catalase; COFs, Covalent Organic Frameworks; CP, Conducting Polymer; CPs, Conducting Polymers; CPF, Chitosan Nanoparticles Fibrous carrier; CR, Congo Red; CS, Chitosan; CNTs, Carbon Nanotubes; CVD, Chemical Vapor Deposition; DNNSA, Dinonylnaphthalene Sulfonic Acid; EDS, Energy Dispersive Spectroscopy; EDTA, Ethylene diamine Tetra Acetic acid; EIS, Electrochemical Impedance Spectroscopy; EY, Eosin Yellow; FESEM, Field Emissive Scanning Electron Microscopy; FTIR, Fourier Transform Infrared Spectroscopy; GO, Graphitic Oxide; GSH, Glutathione; G-C<sub>3</sub>N<sub>4</sub>, Graphitic Carbon Nitride; HRTEM, High-Resolution Tunneling Electron Microscope; HTCC, N-[2-hydroxy-3-trimethyl-ammoniumpropyl] Chitosan Chloride; IC, Indigo Carmine; MAA, Mercaptoacetic Acid; MBT, 2-Mercaptobenzothiazole; MG, Methyl Green; MNMs, Metal Nanomaterials; MMT, Montmorillonite Clay; MO, Metal Oxide; MO, Methyl Orange; MOFs, Metal Organic Frameworks; MNPs, Metal Nanoparticles; MWCNTs, Multi Walled Carbon Nanotubes; NMs, Nanomaterials; NO, Nitric Oxide; NPs, Nanoparticles; NCs, Nanocomposites; OMMT, Organically Modified Montmorillonite; PANi, Polyaniline; PANi-GO, Polyaniline-Graphene Oxide; PANiHM, Polyaniline Hollow Microspheres; PAMpDA, Polyaniline-co-m-phenylenediamine; PAMpDA@Fe<sub>3</sub>O<sub>4</sub>, Polyaniline-co-m-phenylenediamine@Fe<sub>3</sub>O<sub>4</sub>; PFO, Pseudo First Order; PPM, Polypyrrole methane; PPY, Polypyrrole; PPCPs, Personal Care Products; PSO, Pseudo Second Order; PTp, Polythiophene; RBBR, Remazol Brilliant Blue R; RGO, Reduced Graphene Oxide; Rh-B, Rhodamine-B; SBB, Sudan Black B; SCMNP, Silica-coated Magnetic Nanoparticles; SEM, Scanning Electron Microscopy; SDS, Sodium Dodecyl Sulphate; SOD, Superoxide Dismutase; TFC, Thin Film Composite; TPP, Triphosphosphate; ZVM, Zerovalent Metallic.

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and health impacts [4]. Dye contamination in aquatic environments is an emerging environmental concern, driven by the widespread use of synthetic and natural dyes in different industries [5]. Addressing dye contamination presents several challenges, including the complex nature of dye compounds, the need for effective treatment technologies, and the economic implications of implementing pollution control measures [6,7]. Effective strategies for managing dye contamination include the adoption of advanced treatment technologies, such as adsorption, oxidation processes, and biological treatment methods [8].

This review paper outlines the role of different natural and synthetic adsorbents in the purification of impure water and thus will help in maintaining a balance between biological systems, ecological processes, and environmental preservation [1]. It provides a summary of the economic and technical viability of the addition of low-cost adsorbents in enhancing wastewater treatment. The review article reveals several low-cost materials like CS, CPs, and MO NPs individually or in combination for their adsorption characteristics and evaluates the feasibility of their application to provide an efficient and viable solution to problems concerning water pollution [9]. Health and regulatory agencies increasingly pay attention to the fate and behavior of commercial nanostructures in aquatic environments. Because of their specific properties, nanomaterials (NMs) find wide applications with potential risks to environmental and human health [10]. Although continuous research efforts are being undertaken, the regulatory mechanisms will be the key to proper risk management to ensure safety by using NMs [11, 12]. The application of nanotechnology can already be considered a great promise to attain better efficiency in water filtration and decontamination processes. Owing to the characteristics of NMs, it is possible to exploit them to enhance the efficiency of pollution removal, improve the efficiency of filtration systems, and provide more efficient water treatment. Herein, the potential of nanotechnology to revolutionize water purification methods was discussed in detail [13].

Advanced technologies for water treatment are in demand because of the vast and complicated nature of contaminants within water resources. Therefore, nanotechnology opens new routes for improving filtration and decontamination by manipulating at the atomic and molecular levels [14]. Nanotechnology utilizes NMs properties that are quite dissimilar from their macro analogs, particularly the surface area, increased reactivity, and selective adsorptive functions [15]. Different types of NMs, like CS, CPs, and MO NPs, and their applications in filtration systems are discussed in the subsequent sections.

Metal NPs, including metal, MO, carbon-based materials like Silver, Titanium dioxide, Graphene, etc., were utilized to attain higher surface areas and catalytic characteristics for degradation of impurities or overcoming the resistance created during filtration [16]. Nanocomposites (NCs) are composites of NMs and traditional media, such as membranes, meant to enhance the efficiency of filtration processes as well as contaminant removal [17]. Nanofibers and Nanotubes find applications in membrane technologies to increase permeability and achieve increased filtration efficiency [18]. Nanotechnology affords different mechanisms, including adsorption, photocatalysis, and chemical reactions, which facilitate water remediation [19,20]. NMs such as Titanium dioxide might employ energy from light to oxidize harmful contaminants that will then degrade organic compounds into less harmful forms.

The advantages of nanotechnology in water treatment systems include improved efficiency, energy conservation, and specificity in the control of targeted contaminants [21]. A critical review article is required, focusing on the role of CS, CPs, and MO NPs in the impure water purification process. Few challenges still exist in industrial-scale water treatment for the complete removal of different substances, mainly organic dyes and ions associated with heavy and toxic metals. Those impurities usually resist and thus remain dissolved for longer periods in the aquatic ecosystems, even when the water is released into those water bodies, creating persistent environmental hazards. A potential solution to this problem is promising and is rather an ethical

application of metal nanoparticles (MNPs) or MO NPs. These NPs can improve the efficiency of pollutant removal with advanced mechanisms such as catalysis and adsorption [22,23].

The increasing involvement of metal nanomaterials (MNM) in the latest prevailing fast-changing technological trends has also been witnessed primarily in water treatment. MNMs have excellent catalytic activities, including photo-catalysis, and high adsorption reduction efficiency. The surfaces of NMs can potentially be functionalized to enhance performance. Additionally, MNMs are very biocompatible, which is significantly relevant in using these particles for applications involving environmental remediation and similar technologies [24,25].

Different types of treatment of industrial wastewater pollutants were reported. A few are, i) electrical treatment and Photocatalytic reduction involve electrocoagulation, electro-oxidation, electro-dialysis, microbial fuel cell development, followed by photocatalytic degradation of organic dye in the presence of semiconductor catalyst, ii) Precipitation involves adding precipitants example lime softening agents to impure water so that dissolved impurities (like heavy metals, phosphates, or fluorides) react and form insoluble solid precipitates, iii) A combination of ion exchange method and reverse osmosis method for the purification of impure water, iv) Nanofiltration using porous nanomaterials followed by adsorption of impurities on solid adsorbent support.

In coagulation, coagulants help neutralize the charge on colloidal suspensions, encouraging even the tiniest particles to bond together. Chemical precipitation is a useful technique for the treatment of dissolved metal ions from industrial wastewater by converting them into insoluble particles. Ion exchange cleans wastewater by swapping out ions of similar charge with resins. These resins have active groups embedded in a polymer matrix that promote effective ion movement. Nanofiltration eliminates ions that contribute to osmotic pressure, with pore sizes ranging from 0.01 to 0.035  $\mu\text{m}$  [25]. Heavily contaminated water requires pre-treatment before this process. Reverse osmosis employs pressure to filter out dissolved solids and particles, including bacteria, thanks to its tiny pores measuring 0.0001  $\mu\text{m}$ . It requires just the right amount of pressure and minimal maintenance. Adsorption is a widely used technique for removing natural and organic contaminants by facilitating chemical interactions between two different phases. It effectively eliminates these contaminants by promoting chemical reactions between them [26].

This review article presents a comprehensive and novel synthesis of current advancements in CS, CPs, and MO NPs-based NCs for sustainable water treatment applications. Unlike previous reviews, this work uniquely integrates the synergistic interactions between natural biopolymers and inorganic/organic NMs, highlighting how such hybrid systems enhance adsorption, photocatalysis, and antimicrobial activity. The article not only categorizes these materials based on their structural and functional roles but also evaluates their performance in removing a wide range of pollutants, including dyes, heavy metals, pharmaceuticals, and pathogens.

One of the key strengths of this review lies in its interdisciplinary approach—bridging material chemistry, environmental engineering, and nanotechnology—to propose design strategies for next-generation water purification systems. However, despite the depth of analysis, certain limitations exist. For instance, the discussion is largely focused on laboratory-scale studies, with limited insights into scalability, real-field deployment challenges, long-term stability, and regeneration efficiency of the nanocomposites. Furthermore, regulatory and toxicity-related concerns are touched upon but warrant deeper investigation. These limitations underscore the need for future research to focus on pilot-scale demonstrations, life-cycle assessments, and eco-toxicological evaluations. By addressing these gaps, the promising potential of CS and CPs-based NCs in water treatment can be more effectively translated into real-world sustainable solutions.

## 2. NCs in water purification technology

NMs are mostly characterized by properties that may differ from those of the parent bulk material. Materials at the nanoscale present unique electrical, mechanical, optical, and magnetic properties. These arise primarily from quantum effects and increased surface interactions, which may potentially modify behavior and functionality. Beyene et al. highlighted that industrialization, urbanization, and climate change led to global freshwater scarcity. In response, scientists and governments are exploring nanotechnology-based water treatment methods. NCs with high surface areas effectively remove pollutants such as bacteria, viruses, and toxins, essential for water purification [27]. NPs perform better pollutant removal for different water-borne toxins, including heavy metals, organic contaminants, inorganic anions, and even pathogenic microorganisms. The most advanced and approved NMs used in water and wastewater treatment processes include carbon nanotubes (CNTs), MO NPs, zerovalent metallic NPs (nZVM), and others like COFs, MOFs, g-C<sub>3</sub>N<sub>4</sub>, etc. These materials provide suitable properties, including high surface areas, reactivity, and the possibility of targeting specific pollutant fractions, enhancing the efficiency of purification processes. [28–30]. Shasha Liu's team developed a Thin Film Composite (TFC) biofilm enhanced with biogenic Ag NPs (BioAgO-6, ~6 nm). These membranes offer improved hydrophilicity, water permeability, and salt rejection over chemically synthesized counterparts. They retain 95 % of silver after 50 days in water, maintain 90 % rejection performance after four months, and exhibit durable antibacterial activity against pathogens like *Pseudomonas aeruginosa* and *Escherichia coli*, effectively reducing biofouling in nanofiltration [28]. Johnson et al. investigated the incorporation of NMs as fillers in polymeric nanofiltration membranes to improve water transport and fouling resistance while preserving selectivity. They investigate the role of NMs in modifying the structural and chemical properties of membranes, which impacts transport efficiency, selectivity, and fouling. The study underscores the significance of 0D, 1D, and 2D nanofillers in enhancing membrane performance and functionality [30]. When one or more dimensions of such phases approach the nanoscale, interfaces between different phases have an increasing importance. Thus, engineered interfaces between phases in NCs hold the key to improved performance in advanced applications of NMs [31,32]. NCs have exceptional efficacy in the elimination of numerous pollutants from water through very selective mechanisms like chelation, adsorption, and ion exchange. The mechanisms enable effective bacterial, viral, and pollutant removal.

B. Raneesh et al., [33] provide a comprehensive overview of advancements in MO-based NCs in research. They discuss the methods of preparation and characterization, along with a variety of applications, such as core/shell structures, multilayer thin films, and silicon-mixed oxide systems. The authors also highlight practical applications, including gas sensors and hydrogen production, which are significant for photovoltaic and photocatalytic technologies. MO NPs, when combined with CPs and CS, ensure high reactivity and targeted interactions with the contaminants. MO NPs improve their photocatalytic and adsorption properties toward the adsorption and decomposition of pollutants. Carbon-based NCs, depending on carbon content, such as CNTs or graphene, g-C<sub>3</sub>N<sub>4</sub>, show high adsorption efficiency for the pollutants due to their large surface. Polymer-based NCs depend on the embedding of NPs in polymer matrices, with which the properties may be tailored for specific interactions toward pollutant-removing functions. Finally, membrane NCs with embedded NPs into membrane structures enhance their filtration performance and contaminant rejection [33]. The matrix material is the most critical factor of NCs, as its properties directly affect the application and properties of the whole composite. There are primarily three types of matrix materials used for NCs, i.e., metal, ceramic, and polymer matrix.

Metal matrix NCs contain metal matrices with nanometer-sized particles that are added to them for better thermal stability, mechanical strength, and conductivity. Such properties make the metal

composite suitable for application in aerospace, automotive, and electronics. Ceramic matrix NCs contain a ceramic matrix reinforced with NMs. The doping of NPs into the ceramic matrix can enhance its toughness, thermal conductivity, and wear resistance significantly. By incorporating nanoscale reinforcements into a ceramic matrix, ceramic matrix NCs are most suitable in such high-temperature and abrasive environments as cutting tools and aerospace components.

Polymer matrix NCs consist of polymer matrices embedded with nanofillers like MO NPs. These nanofillers improve the mechanical, thermal stability, and barrier properties of the polymer to a great extent. The applications of polymer matrix NCs exist in almost every field, be it in packaging, the automotive field, or biomedical applications [34]. In each of these matrix materials, new properties are introduced to the NCs that can be selected to custom-engineer specific application needs through the intentional addition of nanoscale reinforcements. Jiahua Zhu et al. provide a study of the preparation and applications of magnetic NCs [35]. They highlight the potential of these materials for environmental remediation, particularly in the adsorption of heavy metals such as Chromium, Arsenic, Palladium, and Mercury. The study examines various fabrication methods, adsorption behaviors, and key factors that influence practical applications, including contaminant concentration, adsorption kinetics, and solution pH. The magnetic separation-based NMs have high adsorption behaviors of heavy ferrous-based metal ions as a novel NCs adsorbent [35]. This combines the properties of both magnetic and NCs in the purification process. Some of the key areas that affect the adsorption power of NCs are the concentration of adsorbent and pollutants, adsorption kinetics, pH of electrolyte, etc. [36,37]. Manoj B. Gawande and his team explore the emerging role of Silica-coated Magnetic NPs (SMNPs) as innovative and recyclable nanocatalysts for sustainable industrial applications. They highlight the unique advantages of SMNPs, such as recent advancements in their design, functionalization, and stabilization [37].

Integration of NMs with CS, CPs, and MO NPs is the key to advancing these technologies for water treatment and dealing with complex challenges in purification and management [38–41]. Karthikeyan Ramamurthy et al. investigated the hepatotoxicity of Sudan Black B (SBB), a textile azo dye, on embryonic zebrafish. SBB exposure caused dose-dependent toxicity, with 100 % mortality at 0.8 mM, along with growth defects and liver damage. Biochemical analysis revealed increased SOD, CAT, NO, and LDH levels, reduced GSH, lower glucose levels, and higher lipid accumulation. Inflammatory gene expression changes indicated a response to SBB-induced toxicity [41].

## 3. CPs in water purification technology

The macromolecular structures consist of long chains of repeating units known as monomers [42–44]. CP exhibits different properties like mechanical, optical, electrical, wettability, etc. The molecular structure and the degree of doping of the polymer decide its properties [45]. CP is mostly organic material, comprised of extended  $\pi$ -conjugated systems. Such conjugated systems include polyaniline, polypyrrole, polythiophene, polyacetylene, etc., which have been engineered with electrical properties through several chemical modifications and doping processes. CP thus comprises versatile materials that can be modified through changes in their structure and chemistry to find applications in electronic equipment and sensors, among other advanced technologies [46,47]. The properties of CP that make them efficient for various applications include their optical, mechanical, electrical, magnetic, and microwave absorptive properties. In microwave absorbers, materials absorb microwaves and are very important in photodetectors and defense coating, as they give immunity to outer monitoring. The magnetic behavior of CPs offers useful information about charge-bearing groups and unpaired spins. An electrical property of CPs is affected by doping. Doped conducting polymers show increased conductivity compared to the original polymer due to the introduction of charge carriers. The mechanical properties of CPs are recognized for their flexibility,

lightweight characteristics, and good mechanical strength. Doping enhances their mechanical properties. The unique optical properties of CP are characterized by the ability to absorb and emit light both in the visible and near-infrared regions. Such properties can be modified with doping or structural changes.

Different properties of CPs, like microwave absorbing, mechanical, optical, electrical conductivity, wettability, and magnetic properties. Conducting polymers like polyaniline (PANI), polypyrrole (PPy), polythiophene, PEDOT, etc., are also termed as microwave absorbing materials (MAMs). Mixing of CPs with elastomers, biopolymers, graphene, CNTs, or metal oxides enhances their mechanical strength and toughness. Optical properties of CPs depend on nature of dopants, temperature, electrical potential, etc. Electrical conductivity of CPs depends upon nature of dopants and nature of CPs. Wettability properties of CPs determine how these materials interact with liquids (especially water), which directly affects applications in sensors, coatings, biomedical devices, energy storage, and anti-corrosion layers. Presence of  $\pi$ -conjugated backbones + doping-induced charge carriers give rise to unusual magnetic properties of CPs.

CP NCs have come to find more intensive applications in recent years, well suited to advanced technologies such as sensors, energy storage devices, and electronic components, pointing to the increasing versatility and effectiveness of these materials in a wide range of applications [48–50]. The electrical, electronics, and optoelectronics sectors hold significant applications of conjugated polymer composites due to synergistic effects prevailing in their components. The synergistic effect of elements enables the composite to meet challenging performance demands in several applications. CP composites enabled electrical devices feature high conductivity and flexibility, making them suitable for flexible electronics, interconnects, and conductive coating [51]. In electronics, carrier transport properties are improved along with electrical characteristics, which can be modulated to be applied in organic light-emitting diodes, organic transistors, and organic photovoltaics [52]. Optoelectronics opens up the avenue to control optical properties through a composite formation that leads to new developments in light sensors, photodetectors, and optical modulators [53].

The mechanical, conductivity, and stability of the CP are affected by internal interactions-including interactions between chain segments of the polymer chain and inter-chain interactions, namely, those relating to different polymer chains [54,55]. Doping or photoexcitation can readily induce a transition from nonlinear excitation regimes in conjugated polymers to metallic states. Localization of  $\pi$  bonds follows this transition and leads to the formation of rather complex excitations such as polarons, solitons, and bi-polarons. This process of doping or photoexcitation introduces charge carriers or excitations, providing interaction for these charge carriers or excitations with the electronic structure of the polymer, followed by establishing self-localized states showing peculiar electronic and optical properties [56,57]. CP-based NCs are synthesized by different techniques like Chemical Vapor Deposition (CVD), Electrochemical polymerization, and solution-based synthetic approaches to polymerization, including oxidative coupling and metathesis, each of which provides unique advantages and is selected based on the desired properties of the final polymer, such as its electronic characteristics, morphology, and scalability for specific applications [58]. Nesrine Harfouche et al. developed polyaniline-graphene oxide (PANI-GO) composite films via electrodeposition in a neutral potassium nitrate solution on platinum and iron electrodes. The neutral bath was chosen for better GO dispersion stability and reduced Fe corrosion. The coatings were analyzed using EIS, UV-Vis, FTIR, and SEM. Corrosion tests in 1.0 M HCl highlighted the composite's potential as an anti-corrosion coating [58].

Ten most commonly used methods of preparation of CPs used by Physicists, Chemists, NMs and Material Science background researchers include host-guest synthesis, chemical, electrochemical, photochemical and concentrated emulsion methods of synthesis of CPs were used by

Chemistry background researchers. The methods like plasma polymerization, soluble precursors, solid-state synthesis, metathesis synthesis and pyrolysis methods were used by physics and Materials Science background researchers.

**Chemical synthesis:** Initially, the monomers undergo oxidation or reduction before being polymerized to produce CPs. **Electrochemical polymerization:** Electroactive monomers are oxidized in anodic conditions or reduced in cathodic conditions to form polymers. This approach is straightforward, cost-effective, and efficient. **Photochemical synthesis:** This method is quick, environmentally friendly, and economical for creating the desired conducting polymers. **Metathesis** is a type of reaction where parts of two different compounds come together to form new molecules. **Emulsion polymerization** is a process that occurs in different phases, involving water, latex particles, and monomer droplets, driven by radical polymerization. **Inclusion polymerization:** produces composite materials at the molecular level, allowing for the creation of innovative low-dimensional composites with significant potential. **Solid-State Polymerization:** This process simply stretches out polymer chains by heating them in the absence of oxygen and water. Using a vacuum or inert gas removes by-products while keeping temperature, pressure, and diffusion under careful control. **Plasma Polymerization:** This process forms thin films without pinholes and cross-links materials which eventually turn out to be insoluble, thermally stable, chemically inert, and mechanically robust. **Pyrolysis:** Pyrolysis is breaking up organic materials at extremely high temperatures to analyse polymers present in plastic and rubber, etc., thus doing less sample preparation research contributes greatly to the environment.

CP can be doped with inorganic, organic, or polymeric dopants to increase conductivity, processability, and stability. Dopants may be categorized into broad classes according to their mechanism: self-induced dopants change the electronic structure without an external condition, neutral dopants maintain charge neutrality, redox dopants allow charge transfer by redox, ionic dopants add mobile ionic species, and non-redox dopants affect conductivity by mechanisms other than redox [59,60]. For conjugated polymers, different doping methodologies are used, like charge-injection doping, in which charge carriers are introduced through an applied voltage to allow the direct generation of charge carriers in the polymer matrix. In Radiation-induced doping, doping is done using high-energy radiation, like UV or X-ray irradiation, to produce charge carriers or form reactive sites in the polymer structure [61]. In the *in-situ* doping process, dopants are mixed into the polymer at the time of synthesis or polymerization. So, there will be perfect homogeneity and distribution of dopants in the polymer matrix [62]. In chemical doping, the conjugated polymer is doped with dopants through a chemical reaction. In this doping process, dopants interact with the polymer, thereby changing its electronic structure and, hence, its conductivity [63]. Domini et al. synthesized polyaniline (PANI-HA) doped with DNNSA, enhancing solubility and processability in organic solvents. Methanolic base solutions were more effective than aqueous ones for counter-ion exchange, with deprotonation also occurring in neutral methanol. Re-protonation using acids or salts (e.g., Zinc nitrate) introduced various counter-ions while preserving conductivity and electroactivity, as confirmed by UV-visible-NIR spectroscopy [63]. The electrochemical doping technique uses an external electrochemical potential to dope with charge carriers created in the polymer through oxidation and reduction reactions at the interfaces between the polymer and the electrodes. Each has different advantages, so they can be chosen according to special demands for improving the electrical properties, stability, and processability of the polymer [64]. Thanh-Hai Le and colleagues [64] investigated the conductivity mechanisms as well as the electrical and electrochemical properties of CPs. Their study emphasized essential factors that influence these properties, mainly focusing on the size and morphology of the materials as critical parameters. Additionally, they offered a comprehensive discussion on recent trends in research related to electrochemical capacitors and sensors, highlighting the most significant studies published since 2003.

Electrochemical doping technique included the charge injection (charges are injected into the polymer backbone by applying an external potential), radiation-induced (uses high-energy radiation to generate charges or reactive species that dope the CPs), *in-situ* (the polymerization of monomers (like aniline, pyrrole, thiophene) is carried out in the presence of a dopant), Chemical (Oxidative (p-type) and Reductive (n-type) doping) and electrochemical methods (applying a potential (oxidizing or reducing) injects charges into polymer backbone) of doping of CPs.

It was found that CPs, as well as their NCs, are highly effective and efficient as adsorbents in the removal of pollution in the environment. This may be attributed mainly to its varied physical as well as chemical, and redox properties [65,66]. Different studies were carried out on the employment of modified CP to remove pollutants. For instance, it has been proven that PANi-PPy nanofibers are ideal for the effective removal of Cr(VI) from polluted environments. This capability arises from the synergistic effects of the electronic structure of the polymer and the surface properties [67,68]. Madhumita Bhaumik and her research team [67] developed polypyrrole-polyaniline (PPy-PANi) nanofibers as effective adsorbents for removing chromium in the form of Cr(VI) from aqueous solutions. Batch adsorption studies showed that Cr(VI) uptake was highly pH-dependent, achieving optimal performance characterized by the pseudo 2nd order kinetic model and the Langmuir isotherm, indicating monolayer adsorption. The nanofibers displayed a high maximum adsorption capacity of 227 mg/g, maintained efficiency in multi-ion environments, and exhibited stability over two adsorption-desorption cycles. It is widely recognized that CPs and their composites act as powerful adsorbents for a wide range of impurities and pollutants. Their outstanding performance in adsorption processes can primarily be attributed to the enhanced electrical conductivity of these materials, which allows interactions with various pollutants, besides their adjustable physical and chemical properties that could be tailored for specific environmental applications. The proposed mechanisms for degrading organic dyes from water are hydrophobic interactions, hydrogen bonding,  $\pi$ - $\pi$  stacking, acid-base interactions, and electrostatic forces. All these mechanisms facilitate effective adsorption and the removal of organic dyes. The primary mechanisms for the removal of heavy metal ions are reduction processes, chelation, ion exchange, and electrostatic attraction. The reduction processes convert metal ions to their less soluble, less toxic forms. Chelation uses chelating agents that form stable complexes with metal ions. Ion exchange involves an exchange with another ion in an adsorbent. Electrostatic attraction is based on the charge interactions between metal ions and the adsorbent. Each method is selected based on the specific properties of the contaminants and the adsorbent material [69].

Ehsan Nazarzadeh Zare and his team synthesized poly(aniline-co-m-phenylenediamine) (PAMPDA) through radical oxidation polymerization of aniline and m-phenylenediamine. They then created a super-paramagnetic NCs, poly(aniline-co-m-phenylenediamine)@Fe<sub>3</sub>O<sub>4</sub> (PAMPDA@Fe<sub>3</sub>O<sub>4</sub>), using PAMPDA and iron salts (Fe(II) and Fe(III)) via in situ co-precipitation. The study investigated the ability of PAMPDA and PAMPDA@Fe<sub>3</sub>O<sub>4</sub> to remove toxic heavy metal ions—specifically Pb (II), Cd(II), and Co(II)—under varying conditions, such as time of contact, adsorbent dosage, pH, and original concentration. Co(II) showed the highest sorption, and the data were analyzed using two isotherm models and kinetic equations [70]. Available in various forms, these nano-adsorbents have been utilized to efficiently remove heavy metal ions and organic dyes from wastewater. Their diverse configurations allow for tailored approaches in optimizing the efficiency of adsorption toward the specific contaminant challenges in various contexts of the environment [69,70].

### 3.1. PANi-based NCs in water purification

Mohamed Laabd and his team [71] studied a PPy composite from cellulosic agricultural waste, specifically Argan nut shells, synthesized

through chemical oxidative polymerization. They explored its potential as an eco-friendly adsorbent to remove three polycarboxybenzoic acids: hemimellitic acid, trimellitic acid, and pyromellitic acid in single, binary, and ternary systems. Using the Langmuir approach, they effectively described the adsorption data. The comparison between single and multi-component systems revealed an antagonistic effect. The results showed that the affinity of the acids for the PPy/NA composite followed the order: pyromellitic acid > trimellitic acid > hemimellitic acid. An increasing interest has been drawn on CP, particularly PANi, toward their potential applications as advanced adsorbents for a wide range of pollutants and contaminants due to their excellent adsorption capacities [71]. PANi is reported to have effective adsorbent properties, which are highly relevant due to its wide surface areas, tuneable conductivity, and functional groups. It is then possible for PANi to show very high maximum adsorption capacities for different types of pollutants, be it heavy metals, organic dyes, and other toxic residues [72].

#### 3.1.1. PANi-based NCs for the degrading dye

Marjan Tanzifi et al. described that the particle size of 70 nm PANi nano-adsorbent was used for the effective removal of MO dye using a neural network-based model in optimizing the parameters of batch adsorption. Such a model significantly improved the efficiency of the adsorbent. At 65°C, the capacity of adsorbing the MO dye increased from 3.34 mg/g to 32.04 mg/g when the initial MO dye dosage was raised from 10 to 100 mg/L, and at 25°C from 3.28 to 30.28 mg/g. The data of adsorption successfully explained by the maximum monolayer capacity of 75.9 mg/g could be analyzed for interpreting kinetics and equilibrium behavior using the pseudo-2nd order model following Langmuir isotherm [73]. Teng & Bryan et al., in their study, used the PANi/iron oxide hybrid (PANi/Fe<sub>3</sub>O<sub>4</sub>) for Congo Red (CR) dye removal, and it was found that PANi/Fe<sub>3</sub>O<sub>4</sub> composite prepared through a physical mixing technique using acetone was confirmed by FTIR analysis. The best condition for the highest adsorption capacity and removal efficiency of CR dye is at a 70/30 ratio of PANi to Fe<sub>3</sub>O<sub>4</sub>. The performance of the composite significantly improved by merely coating Fe<sub>3</sub>O<sub>4</sub>. Optimal adsorption conditions are observed at a temperature of 45 °C, 0.1 g of adsorbent, and 20 ppm of dye [74]. The removal percentage cycle of CR dye is shown in Fig. 1.

Also, Y. Zhao et al., explained that hierarchical MWCNTs/Fe<sub>3</sub>O<sub>4</sub>/PANi composite was synthesized and showed magnetic separability. The kinetics for the adsorption of MO by the composite followed the pseudo-2nd order model, and adsorption showed high pH dependency. For MO adsorption, the Langmuir model could better describe the isotherm than the Freundlich model, and the composite showed very high adsorption

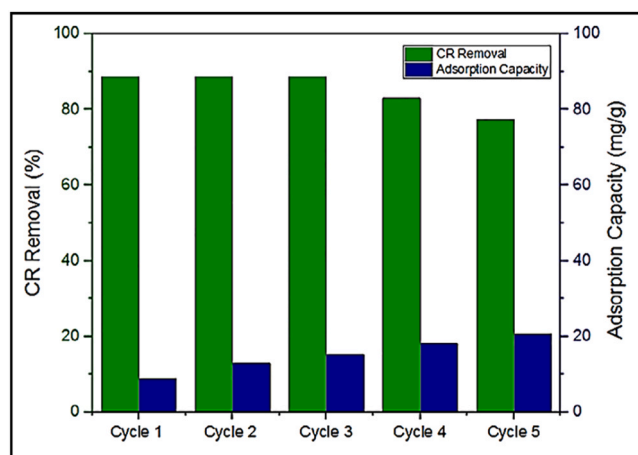


Fig. 1. The impact of the number of sessions on the CR adsorption capacity and removal percentage of the PANi/Fe<sub>3</sub>O<sub>4</sub> composite. [Reprinted with permission from (<https://doi.org/10.1016/j.matpr.2021.01.766>). Copyright (2021) Elsevier].

capacity for both MO and CR, which is due to the N-containing groups in PANi. PANi also resisted the dissolution of  $\text{Fe}_3\text{O}_4$  NPs, so enhancing the stability and reusability of the composite in solution as discussed in Fig. 2. Fig. 2 A illustrates the adsorption rate of MO using MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi NC. The surface of MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi contains numerous active adsorption sites, allowing for rapid adsorption within the initial 160 min. The longer equilibration time is attributed to the saturation of these sites, which restricts the diffusion of MO. Fig. 2 B presents the pH dependence of MO adsorption through MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi. The trend observed in Fig. 2 A and 2B indicates a negative correlation between MO adsorption and pH values above 4.0, while a positive correlation is noted for pH values below 4.0. Consequently, with an optimal adsorption efficiency at pH 4.5, further kinetic and isothermal studies were conducted at this pH level. Fig. 2 C compares the MO adsorption isotherms across  $\text{Fe}_3\text{O}_4$ , MWCNTs, PANi, MWCNTs/ $\text{Fe}_3\text{O}_4$ , and MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi. This comparison shows that the adsorption capacity of the MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi NCs surpasses that of all its components and their binary combinations. Fig. 2D explores the reusability of the MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi. After four cycles, the adsorption efficiency for MO has only slightly decreased. These findings confirm that MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi is highly effective for practical applications in dye removal, demonstrating excellent recyclability and regeneration properties [75].

Mousumi Mitra et al., discussed *in-situ* oxidative polymerization synthesis of PANi and PANi/RGO composites at various concentrations of RGO. These materials efficiently degraded Methyl Green (MG), Rh B,

and CR under visible light. The composite of PANi/RGO presented a superior photocatalytic activity over that of PANi and RGO individually due to the synergistic effect added by the sheet-like morphology of RGO. Among the composites, PG2 that contained 5 wt% RGO presented the most efficient photocatalytic performance, with rate constants about 6.1 and 2.3 times superior for MG, 4.1 and 2.2 times greater for Rh-B, and 3.3 and 2.2 times better for CR than those involving PANi and RGO [76]. The above data of concentration plots of different dyes is systematically explained in Fig. 3. Table 1 depicts the potential of PANi-based NCs in photocatalytic efficiency for degrading different organic dyes present in impure water. Different attempts were made to improve the performance of PANi-based NCs to enhance their adsorption capacity for degrading different organic dyes in impure water (Table 1) [77–116].

Djelloul et al. [77] developed binary  $\text{ZnO}-\text{Al}_2\text{O}_3$  and synthesized PANi-supported adsorbents through *in situ* oxidative polymerization. The integration of the PANi matrix onto the  $\text{ZnO}-\text{Al}_2\text{O}_3$  composite enhanced porosity, facilitating efficient dye removal. Adsorption equilibrium data aligned well with the Langmuir isotherm model, revealing a maximum adsorption capacity of  $442.8 \text{ mg}\cdot\text{g}^{-1}$ . Thermodynamic analysis indicated that the process was spontaneous and exothermic. Additionally, the adsorbent demonstrated excellent reusability, maintaining efficiency over five recycling cycles. Zhao et al. [82] developed a multi-wall (MW) CNTs-based composite decorated with  $\text{Fe}_3\text{O}_4$  NPs and modified with PANi. The composite efficiently adsorbed methyl orange (MO), with adsorption kinetics following pseudo 2nd order kinetics. It showed high adsorption capacities of  $446.25 \text{ mg/g}$  for MO and

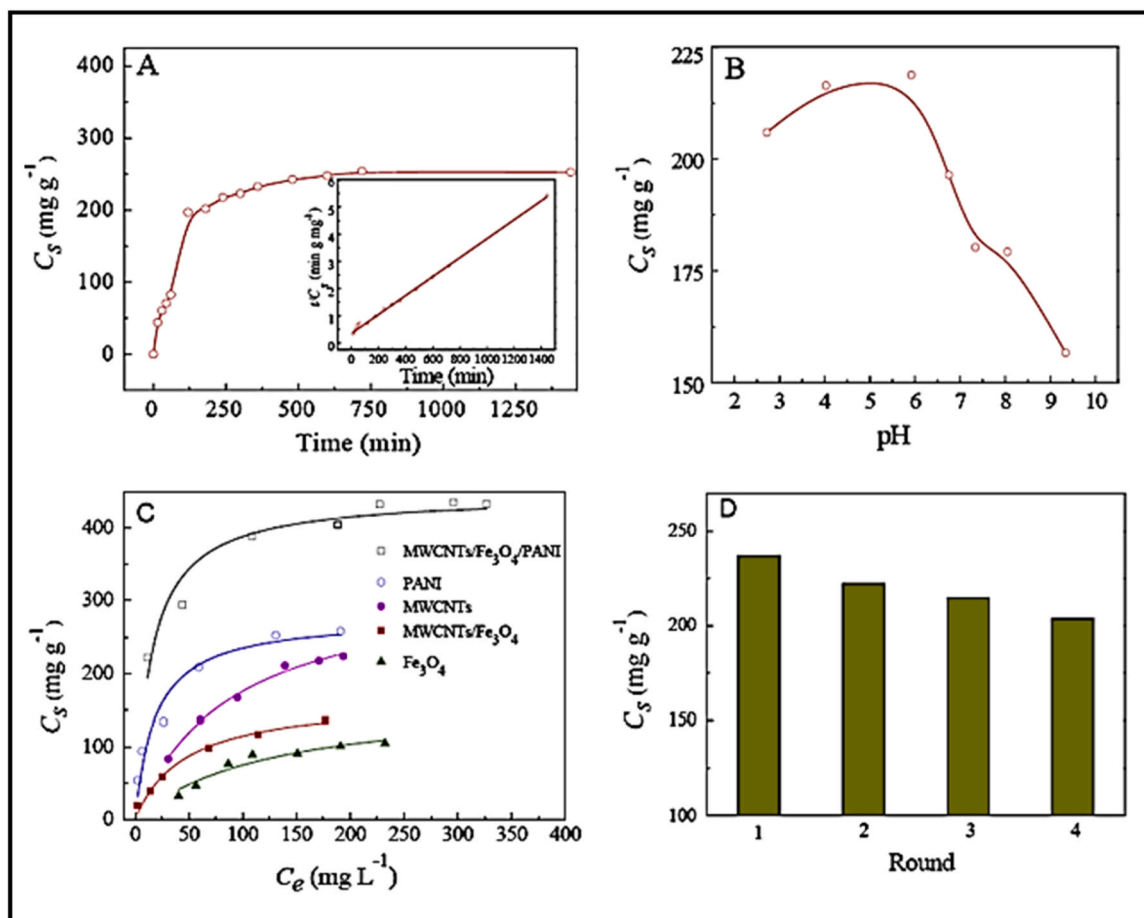
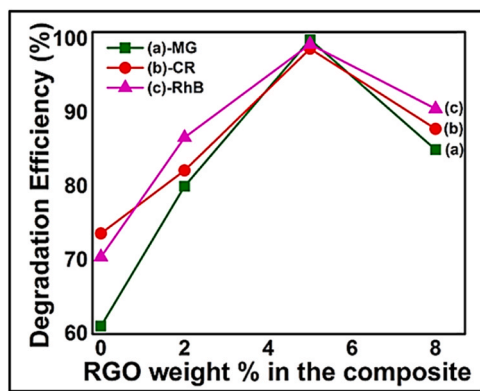


Fig. 2. (A) The graph and inset show pseudo-2nd-order kinetic plots for the adsorption of MO on MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi at pH 4.5 with an initial concentration of  $[\text{MO}] = 150 \text{ mg L}^{-1}$ . (B) The effect of pH on MO adsorption to MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi at an initial concentration of  $[\text{MO}] = 150 \text{ mg L}^{-1}$ . (C) Adsorption behavior of MO on  $\text{Fe}_3\text{O}_4$ , MWCNTs, PANi, MWCNTs/ $\text{Fe}_3\text{O}_4$ , and MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi at pH 4.5. (D) Recycling of MWCNTs/ $\text{Fe}_3\text{O}_4$ /PANi in the removal of MO at pH 4.5 with an initial concentration of  $[\text{MO}] = 150.0 \text{ mg L}^{-1}$ . The mass-to-volume ratio is  $0.4 \text{ g L}^{-1}$  and the temperature was  $298 \text{ K}$ . [Reprinted with permission from {<https://doi.org/10.1016/j.jcis.2015.03.015>}. Copyright {2015} Elsevier].



**Fig. 3.** Concentration ratio plots of (A) MG, (B) Rh B, and (C) CR in aqueous solutions against specific time intervals under various conditions by using the PANi/RGO composite. [A copyright of ACS covered under CC BY, Source: <https://doi.org/10.1021/acsomega.8b02941>].

417.38 mg/g for Congo red (CR). The nitrogen groups from PANi enhanced adsorption, while PANi also stabilized the  $\text{Fe}_3\text{O}_4$  NPs, preventing their dissolution.

It was observed that PANi@ZnO- $\text{Al}_2\text{O}_3$  NCs show 442.8 mg/g adsorption capacity for Congo Red (CR) dye at pH 5.5 and follows pseudo-2nd order kinetic [77] but when  $\text{Al}_2\text{O}_3$  was replaced with  $\text{MnO}_2$  the adsorption capacity of NCs for crystal violet (CV) dye decreases to 119.25 mg/g at 10.0 pH again following pseudo-2nd order kinetic [78]. PANi/ $\text{Fe}_3\text{O}_4$  NCs show an adsorption capacity of 264.9 mg/g for Acid Blue dye in the pH range 2–12 following 2nd order kinetics [84]. Adding MWCNTs in these NCs increases its adsorption capacity to 446.25 mg/g for MO dye and 417.38 mg/g for Congo Red (CR) dye [82]. While the addition of AgO NPs in PANi/CNTs NCs decreases its adsorption capacity to 98.7 mg/g for Brilliant Blue G dye [106]. It was observed that the addition of GO in PANi does not significantly increase the adsorption capacity of NCs i.e., 142.8 mg/g for Brilliant green (BG) dye at 7.0 pH [89]. The addition of  $\text{Fe}_3\text{O}_4$  in Graphene/PANi NCs slightly increases its adsorption capacity to 248.76 mg/g for CR dye at 9.6 pH [94]. The addition of activated carbon in PANi slightly improves its adsorption capacity to 285 mg/g for MO dye at 6.5 pH [87]. The addition of binary MO NPs that is NiO and  $\text{MnO}_2$  NPs resulting in PANi/NiO/ $\text{MnO}_2$  NCs does not significantly improve the adsorption capacity of NCs that is 248.4 mg/g for MO dye at 7.0 pH [80]. Researchers have tried to improve the efficiency of PANi by capping it with gum acacia, but it showed only 35.41 mg/g for MB dye at 10.0 pH [90]. The addition of  $\text{Fe}_3\text{O}_4$  in PANi slightly improves its adsorption capacity to 361.78 mg/g for MO dye [95]. An attempt was also made to modify PANi with almonds, but its adsorption capacity decreased to 9.8 mg/g for Orange G dye in the pH range of 2–11 [96]. The highest adsorption capacity was observed for PANi-Hollow-Microsphere (PANiHM)/ $\text{MnO}_2$ / $\text{Fe}_3\text{O}_4$ , which is 1142.13 mg/g for methyl green (MG) dye at 6.5 pH [110]. Dutta et al. [110] developed a novel composite material comprising polyaniline hollow microspheres (PANiHM) combined with manganese dioxide ( $\text{MnO}_2$ ) and iron oxide ( $\text{Fe}_3\text{O}_4$ ) NPs. This composite was specifically engineered for the efficient removal of hazardous dyes, such as methyl green (MG) and CR, from polluted water. The PANiHM/ $\text{MnO}_2$ / $\text{Fe}_3\text{O}_4$  composite was characterized by a notably high BET surface area of 191.79  $\text{m}^2/\text{g}$ , underscoring its exceptional adsorption potential. The composite demonstrated impressive maximum adsorption capacities ( $Q_{\text{max}}$ ), achieving 1142.13 mg/g for MG and 599.49 mg/g for CR dye.

Sujay Das et al. successfully explained the BET surface area of F-PANi2 xerogel was 236  $\text{m}^2/\text{g}$ , which was a porous 3D hierarchically fibrillar network. UV-vis and XPS analyses confirm that PANi in hydrogels is in the emeraldine salt state. Gel robustness was demonstrated through compressive stress-strain testing. Due to amine and imine groups in the F-PANi fibers, xerogel demonstrates excellent

adsorption properties for Cr (VI) and anionic dyes (EY, RB2, and MO) in acidic conditions. According to Sujay Das, the F-PANi2 xerogel effectively absorbs anionic dyes, while cationic and neutral dyes do not get absorbed. This is illustrated in Fig. 4. The experiments were conducted for 30 min at a temperature of 25 °C, testing pH levels of 3, 7, and 9. The highest adsorption values for (Eosin Yellow) EY, RB2, and MO dyes were recorded at pH 3, while a significant decrease in adsorption capacity was observed at pH 9 (Fig. 4A). The electrostatic interaction between the positively charged surface of F-PANi and the negatively charged dye molecules was strongest at pH 3, leading to the highest removal percentage. At pH 7, the addition of folic acid to PANi enhanced the adsorption of anionic dyes. However, at pH 9, PANi was entirely in the EB form, resulting in minimal adsorption of the anionic dyes, as shown in Fig. 4b. UV-vis spectroscopy was used in Fig. 4c to monitor the adsorption behavior of EY dye in the F-PANi2 xerogel as ethanol was desorbed. The results showed that, after a period of exposure, the supernatant displayed an increase in absorbance intensity due to the EY dye, indicating that the dye molecules were dissolving into the solution. The xerogel's regeneration capability was further assessed through multiple adsorption-desorption cycles. As illustrated in Fig. 4d, even after five consecutive cycles, the xerogel maintained up to 83 % of its original EY dye removal efficiency, demonstrating its strong recyclability. These findings confirm that F-PANi2 xerogel is both robust and recyclable for EY dye adsorption, making it a promising candidate for sustainable water treatment applications [117].

Mondal and Rana have shown that the scalability of PANi nanotubes is good for potential usage in removing dyes from contaminated water and oil-water separation. The PANi fibers have a range of aspect ratios depending on the dopant acids used. The highest aspect ratio of PANi fibers comes from doping with B4CA. The PANi nanostructures synthesized here have shown an adsorption ability of up to 300 mg/g for anionic dyes in aqueous solutions. Another part of the PANi fibers, which can be tuned to modulate the aspect ratio, can also be modulated to vary the capabilities of dye adsorption. Indigo Carmine (IC) was selected as a model dye for further investigation through adsorption at room temperature, as shown in Fig. 5. When comparing the adsorption behavior of IC to other selected dyes, it was noted from Fig. 5b that the adsorption capacity of IC is significantly greater than that of the other dyes. The finding was derived from a UV-Vis spectroscopic study, where the absorbance of each dye was tracked over various time intervals. A detailed analysis of the uptake rates of seven dyes using different nanostructures is presented in Fig. 5c. The results indicate that IC dye exhibits superior adsorption compared to the other dyes under the same experimental conditions [118].

### 3.1.2. PANi-based NCs for the removal of heavy metal ions

These include metal and metalloid ions such as  $\text{Pb}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Hg}^{2+}$ ,  $\text{Cr}^{6+}$ ,  $\text{Co}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{As}^{3+}$ ,  $\text{F}^-$ ,  $\text{U}^{6+}$ , and  $\text{Cu}^{2+}$  ions [119]. These pollutants are very soluble and non-biodegradable; therefore, they tend to accumulate in biotic and abiotic systems, which creates severe threats to ecosystems and human beings. Their continuous presence hampers the biological cycle and is responsible for environmental contamination. However, the effectiveness of removal from the system depends upon appropriate treatment methods. Because NCs offer a very large number of sites for chemical interaction due to their increased surface area, they have been proven highly effective in the removal process of heavy metal ion species. Surface functionalization, which increases affinity for specific target ions, also makes them the best candidates in the application of wastewater treatment for the adsorption of heavy metals [120]. The adverse effect of these ions on human health due to higher exposure to the human body is seen in Fig. 6.

Table 2 depicts conducting Polymer, PANi-based NCs for adsorbing toxic/heavy metals [121–146]. Cellulose/PANi NCs show 189 mg/g adsorption capacity for  $\text{Cr(IV)}$  ion at pH [121]. PANi in the presence of plant-based products like kapok fiber shows a very small adsorption capacity of 63.6 mg/g for  $\text{Pb(II)}$  ion [124]. A ternary composite of

**Table 1**  
The incredible potential of PANi NCs in photocatalytic efficiency for facilitating the degradation of different dyes.

| PANi-based NCs                                          | Target Dye                             | pH               | Adsorption Capacity(mg/g)   | Kinetics | References |
|---------------------------------------------------------|----------------------------------------|------------------|-----------------------------|----------|------------|
| PANi@ZnO–Al <sub>2</sub> O <sub>3</sub>                 | CR                                     | 5.5              | 442.8                       | PSO      | [77]       |
| PANi/ZnO/MnO <sub>2</sub>                               | CV                                     | 10               | 119.25                      | PSO      | [78]       |
| (PANi)/activated silica gel (ASG)                       | Brilliant Green (BG)/MO                | 7                | 136.98(BG)/<br>161.29(MO)   | PSO      | [79]       |
| PANi/NiO/MnO <sub>2</sub>                               | MO                                     | 7                | 248.4                       | PSO      | [80]       |
| PANi/ZrO <sub>2</sub>                                   | MB                                     | 7                | 77.51                       | PSO      | [81]       |
| MWCNTs/Fe <sub>3</sub> O <sub>4</sub> /<br>PANi         | MO & CR                                | 4                | 446.25(MO)<br>417.38(CR)    | PSO      | [82]       |
| Cu <sub>2</sub> O/ZnO-PANi                              | CR                                     | 6                | 100                         | PSO      | [83]       |
| PANi/Fe <sub>3</sub> O <sub>4</sub>                     | Acid Blue 40(AB40)                     | 2–12             | 264.9                       | PSO      | [84]       |
| PANi/<br>Fe <sub>3</sub> O <sub>4</sub> /Silica         | Diclofenac                             | 7                | 90 %                        | PSO      | [85]       |
| PANi/Ag                                                 | BG                                     | 12               | 20.92                       | PSO      | [86]       |
| Polyaniline/Activated carbon                            | MO                                     | 6.5              | 285                         | PSO      | [87]       |
| PANi/TiO <sub>2</sub>                                   | MB                                     | 3                |                             | PFO      | [88]       |
| PANi/Graphene Oxide (GO)                                | BG                                     | 7                | 142.8                       | PSO      | [89]       |
| Gum acacia-capped polyaniline                           | MB                                     | 10               | 35.41                       | PSO      | [90]       |
| PANi/Bi <sub>2</sub> WO <sub>6</sub>                    | CR                                     | 5.5              | 142.92                      | PSO      | [91]       |
| PANi/GO/L-Cysteine                                      | CR                                     | 7                | 56.57                       | PSO      | [92]       |
| PANi/ZnO                                                | Methyl Orange (MO)                     | 6–9              | 87.5                        | PSO      | [93]       |
| Graphene/PANi/<br>Fe <sub>3</sub> O <sub>4</sub>        | Congo Red                              | 9.6              | 248.76                      | PSO      | [94]       |
| PANi/Magnetic Oxide (FO)                                | MO                                     | 7                | 361.78                      | PSO      | [95]       |
| PANi-modified almond (PANi@AS)                          | Orange G<br>(OG)                       | 2–11             | 9.8                         | PFO      | [96]       |
| MgFe <sub>2</sub> O <sub>4</sub> /PANi                  | Methyl red                             | 7                | 98.04                       | PSO      | [97]       |
| CaFe <sub>2</sub> O <sub>4</sub> -PANi                  | Victoria Blue                          | 8                | 266.67                      | PSO      | [98]       |
| Iron oxide-PANi-PNC                                     | Eriochrome Black-T (EBT)               | 2–5              | 285.71                      |          | [99]       |
| (Fe-Mn-Zr/PANi-NC)                                      | Methyl orange & Eosin Yellow           | 5                | 217.39 (MO)<br>117.65 (EY)  | PSO      | [100]      |
| MnFe <sub>3</sub> O <sub>4</sub> -PANi                  | MO                                     | 6                | 175.44                      | PSO      | [101]      |
| Fe <sub>3</sub> O <sub>4</sub> /ZnO/PANi                | Magnetically removable catalyst        | NA               |                             | NA       | [102]      |
| PANi/PVA-NiCu                                           | MB                                     | 13               |                             | PSO      | [103]      |
| PANi/ZnO                                                | MO                                     | 11               |                             | PSO      | [104]      |
| (Si–Al)–Cu <sup>2+</sup> /PANi                          | Acid Blue 25<br>(AB25)                 | 2                | 135.86                      | PSO      | [105]      |
| Ag-PANi/CNT                                             | Brilliant Blue G (BBG)                 | 4                | 98.7                        | PSO      | [106]      |
| PANi-DCC@GO                                             | red K-2G                               | 6.5              | 733.3                       | PSO      | [107]      |
| PANi/SiO <sub>2</sub>                                   | MB                                     | 11               | 6.97                        | PFO      | [108]      |
| PANi/GO/MOF-Fe <sub>3</sub> O <sub>4</sub>              | MO                                     | 7                | 239.78                      | PFO      | [109]      |
| (PNHM)/MnO <sub>2</sub> /Fe <sub>3</sub> O <sub>4</sub> | methyl green (MG) & CR                 | 6.5              | 1142.13 (MG)<br>599.49 (CR) | PSO      | [110]      |
| Fe <sub>3</sub> O <sub>4</sub> @PDA@PANi                | Tartrazine (Tar)and Sunset Yellow (SY) | 3                | 113 (Tar)<br>102.9 (SY)     | PSO      | [111]      |
| PANi-TiO <sub>2</sub>                                   | MO                                     | 5                | 62                          |          | [112]      |
| PANi/montmorillonite clay (PANi/MMT)                    | Acid Green 25                          | 10               | 12.5                        | PSO      | [113]      |
| CS-GO                                                   | Acid Red & Brilliant Green             | 2 (AR)<br>6 (BG) |                             | PSO      | [114]      |
| PANi/Fe <sub>3</sub> O <sub>4</sub> @C                  | Reactive blue 198 (RB198)              | 5.5              | 108.69                      | PSO      | [115]      |
| C-CS@ZnO                                                | MO                                     | 8.8              | 185.2                       | PSO      | [116]      |

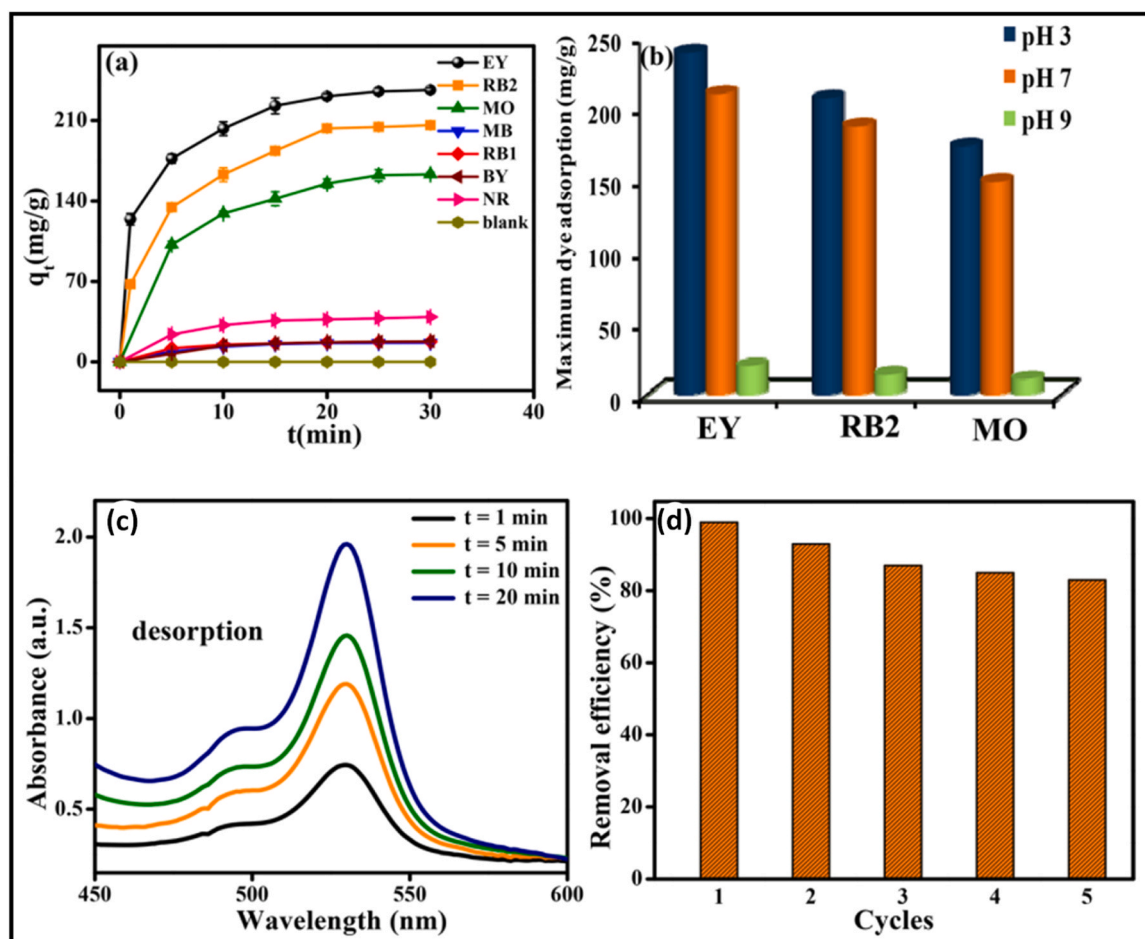
**Note:** PSO stands for Pseudo 2nd order and PFO Stands for Pseudo First order.

PANi/LDH@CNT depicts an adsorption capacity of 303.2 mg/g for molecular iodine [126]. The highest adsorption capacity was shown by PANi doped with 1,2,4,5-benzene tetracarboxylic acid (BTCA) and Zinc phosphate (BTCA-PANi@ZnP) i.e., 933.88 mg/g for Cr(IV) ions [128]. A. Hsini et al. introduced a novel BTCA-PANi@ZnP NCs using a straightforward two-step process. Thermodynamic analysis confirmed both spontaneous and endothermic adsorption. The NCs exhibited an impressive max. monolayer adsorption of 933.88 mg/g. Its high efficiency in adsorbing Cr(VI) ions highlights its potential for large-scale environmental remediation applications [128].

Lincheng Li et al. [133] developed a magnetic core-shell composite, Fe<sub>3</sub>O<sub>4</sub>@UiO-66@PANi, for efficient Cr(VI) removal from wastewater. This composite integrates the stability of UiO-66 with the active functional groups of polyaniline (PANi), achieving a remarkable adsorption capacity of 474.42 mg/g and a removal efficiency of 99.90 %. The study investigated the effects of initial pH, contact time, and Cr(VI) concentration on adsorption performance. The findings revealed that Cr(VI) removal occurs through a combination of electrostatic adsorption,

reduction, and chelation, positioning Fe<sub>3</sub>O<sub>4</sub>@UiO-66@PANi as a highly effective adsorbent for impure water purification. PANi@Ni(OH)<sub>2</sub> NCs shows an adsorption capacity of 625 mg/g for Cr(IV) ions at 4.0 pH [136]. Abdelghani Hsini et al. developed a PANi-coated WO<sub>3</sub> composite (PANi@WO<sub>3</sub>) to remove Cr(VI) from water, demonstrating optimal performance at pH 2. The adsorption process adhered to a pseudo-2nd-order kinetic model and fit the Langmuir isotherm, with the composite achieving an impressive maximum adsorption capacity of 549.37 mg/g, surpassing many other adsorbents [146].

Lincheng Li et al. formed a novel core-shell composite, Fe<sub>3</sub>O<sub>4</sub>@UiO-66@PANi, which was synthesized for the efficient removal of Cr (VI) ions from wastewater. Through a solvent-based and in-situ polymerization approach, the following results were achieved with an adsorbent capacity of 20 mg by using a pH value of 2.0, where 99.90 % of the 20 ppm Cr (VI) was removed while achieving a high capacity of 474.42 mg·g<sup>−1</sup>. Some mechanistic studies explain why the composite has a high adsorption capability, including reduction, electrostatic interactions, and chelation. Furthermore, because of the presence of active



**Fig. 4.** a) Removal of various dyes employing F-PANi2 xerogels at pH 7 and 25 °C, b) adsorption for the maximum amount of various dyes EY, RB2, and MO by F-PANi2 at various pH. c) Time-dependent UV-vis spectra of supernatant during desorption of dye from F-PANi2 in MeOH at 25 °C. d) Reusability test of F-PANi2 xerogels for dye removal up to five cycles at 25 °C. [Reprinted with permission from [ACS Sustainable Chem. Eng. 2017, 5, 9325–9337]. Copyright {2017} American Chemical Society].

groups in PANi,  $\text{Fe}_3\text{O}_4@\text{UiO}-66@\text{PANi}$  presented superior structural stability, acid tolerance, and regeneration capacity. Such properties represent that  $\text{Fe}_3\text{O}_4@\text{UiO}-66@\text{PANi}$  may be utilized as a potential candidate for the removal of Cr (VI) with high separation efficiency and reusability [147].

Yan Zhang et al. described new proton acid-doped PANi magnetic iron oxide core-shell structured NCs that have been designed to deliver good As(V) adsorption with high efficiency within the pH range of 4–11. The  $\gamma\text{-Fe}_2\text{O}_3@\text{PANi}-3$  composite delivered an adsorption capacity of 0.502 mmol/g for As(V). Kinetics implies that the pseudo 2nd order model describes the process, but includes outer and inner liquid film and intra-particle diffusion. Also, the adsorptive properties of a selection of reagents in solution were identified to identify several of the most effective at removing As(V) from solution. The charge and, hence, the reactivity of arsenate anions can be modified at various pH levels and would thus affect the efficiency of adsorption. Toward the achievement of optimum conditions with which to remove As(V), a set of experiments was performed at different pH values for optimum conditions of  $\gamma\text{-Fe}_2\text{O}_3@\text{PANi}-3$ . Along with this, the capacity and efficiency of such NCs for true Arsenate removal application by tracking the adsorption capacity over these cycles. That is also of paramount importance to know about the long-term possibility of using  $\gamma\text{-Fe}_2\text{O}_3@\text{PANi}$  as an adsorbent in ecological restoration [148].

Bin Zuo et al. explained that PANi NCs composed of  $\text{Fe}_3\text{O}_4@m\text{-SiO}_2@\text{CS}$  grafted with PANi (FSCP), proved efficient in the treatment of Cr (VI) from water, following the Langmuir isotherm model with

efficient adsorption capacity and faster kinetics than that described by a pseudo 2nd order model. At low pH, the active redox pair  $=\text{N}-/\text{-NH}-$  *in situ* facilitates the reductive formation of Cr (III) from Cr (VI). The adsorption mechanism must include electrostatic interactions with protonated groups of nitrogen and chelation with species containing nitrogen leading to adsorption of effective Cr (III) and Cr (VI) [149]. This research will be applied to evaluate the effectiveness, applicability, and feasibility of FSCP in overcoming chromium pollution on an environmental scale, especially regarding long-term rehabilitation efforts as depicted in Fig. 7.

### 3.2. Other CPs in water purification technology

Commonly used CPs are Polyacetylene (PA), Polypyrrole (PPy), Polythiophene (PTp), and PANi. Among those materials, heteroatom-based polymers like black polypyrrole, red polythiophene, and mottled polyaniline raise the electron cloud density, which further enhances the polymer's physical and chemical properties. PPy is regarded as one of the good candidates for an environmentally functional material because of its cheap, environment-friendly availability, excellent light absorption, electrical conductivity, and biocompatibility compared with others [150].

By Wenhui Zheng et al., using hydrothermal, *in situ*, polymerization, and modification techniques, a magnetic core-shell composite  $\text{Fe}_3\text{O}_4@\text{Polypyrrole}@\text{Sodium dodecyl sulphate}$  ( $\text{Fe}_3\text{O}_4@\text{PPy}@\text{SDS}$ ) was synthesized. In the adsorption experiments, five dyes, i.e., MB,

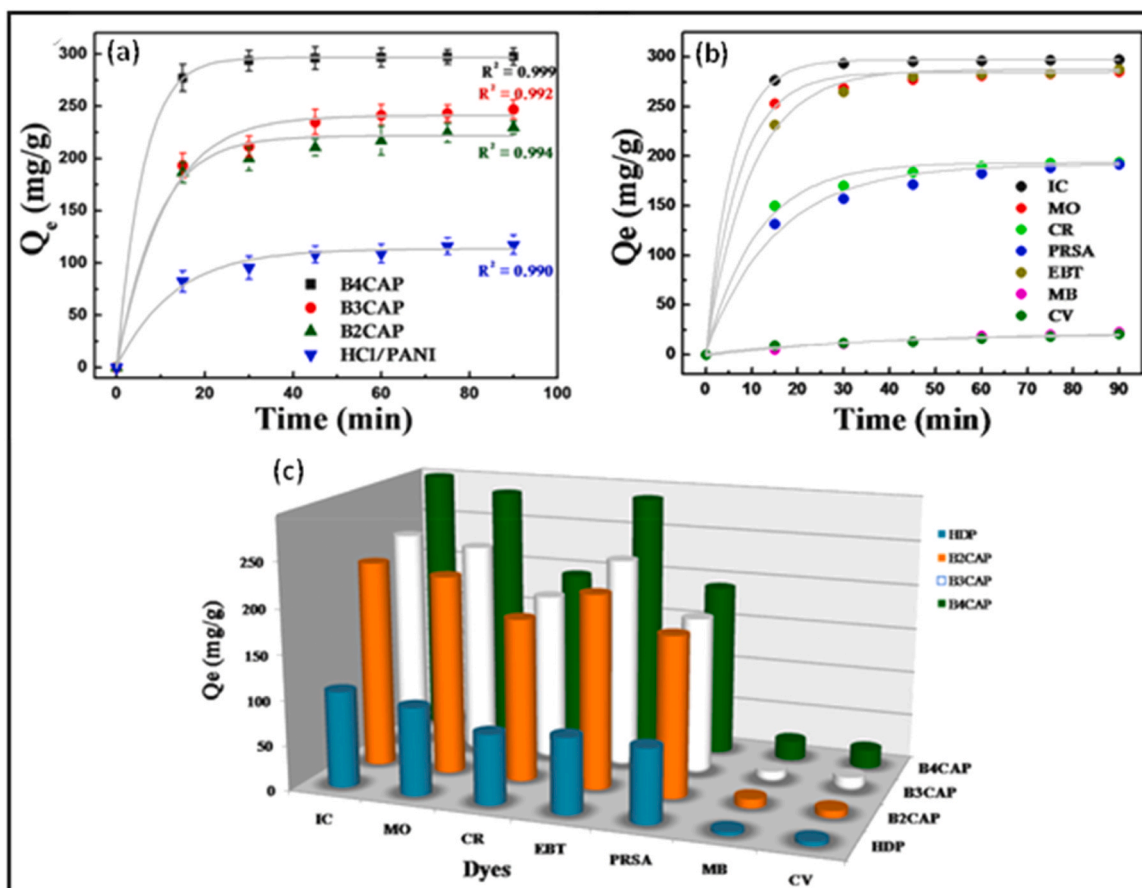


Fig. 5. (a) Adsorption of dyes by various PANi matrixes at room temperature with a similar PANI: Dye (w/w) ratio. (b) Rate of uptake by all the selected dyes using B4CAP. (c) Rate plot of uptake of different dye molecules by PANi nanostructures (B4CAP, B3CAP, B2CAP, and HDP). [Reprinted (adapted) with permission from {ACS Applied Polymer Mater. 2019, 1, 1624–1633}. Copyright {2019} American Chemical Society].

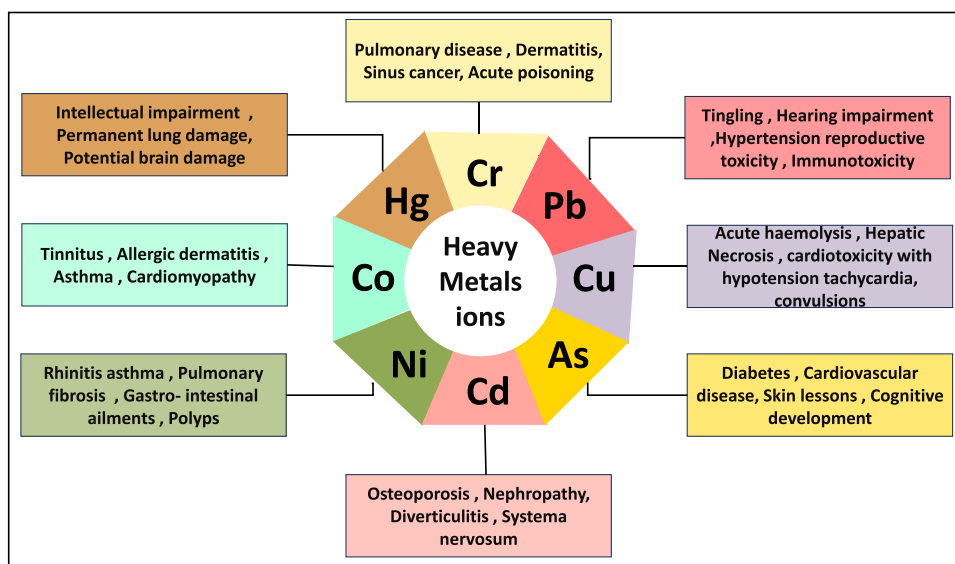


Fig. 6. Effect of exposure to heavy toxic metal ions on the human body.

malachite green, Rhodamine-B, Congo Red, Acid Red-1, and Mn (VII) heavy metal ions removed selectively using this composite. Temperature, contact time, initial concentration of the adsorbate, and different types of adsorbents were taken as factors to check their adsorption performance in great detail. Fig. 8 study investigated the effects of

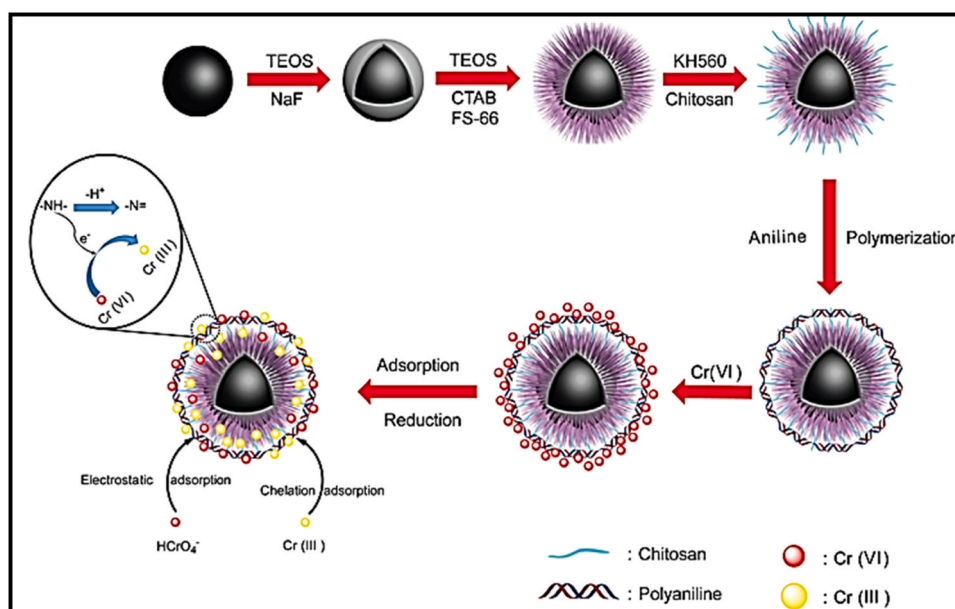
various parameters like the type of adsorbent, time of contact, concentration of debut adsorption stuff, and temperature on the efficiency of the adsorption process [151]. Table 3 depicts the use of PPY NCs for the removal of organic dyes and the adsorption of heavy metals [152–173]. It shows the effect of pH, target metal, adsorption capacity and kinetics

**Table 2**

CPs (PANI) based NCs for the removal of toxic/heavy metals.

| PANI-based NCs                                               | Target Metal          | pH   | Adsorption Capacity (mg/g)        | Kinetics | Reference |
|--------------------------------------------------------------|-----------------------|------|-----------------------------------|----------|-----------|
| Cellulose/PANI                                               | Cr (VI)               | 7    | 189                               | PSO      | [121]     |
| PANI/GO/MOF-Fe <sub>3</sub> O <sub>4</sub>                   | Na- Naproxen          |      | 40.64                             | PSO      | [122]     |
| Ca/Fe/LDH/PANI                                               | Pb (II)               | 6    | 110                               | PFO      | [123]     |
| PANI/kapok fiber                                             | Pb (II)               | 6    | 63.60                             | PSO      | [124]     |
| Fe <sub>3</sub> O <sub>4</sub> @PANI/IA                      | Cr (VI)               | 2–8  | 218                               | PSO      | [125]     |
| (PANI/LDH@CNT)                                               | I <sub>2</sub> (aq.)  | 7    | 303.20(Aq.)                       | PFO      | [126]     |
|                                                              | I <sub>2</sub> (Vap.) |      | 900 (Vap.)                        |          |           |
| PANI-IA-CuO                                                  | Cr (VI)               | 2–10 | 250                               | PSO      | [127]     |
| BTCA-PANI@ZnP                                                | Cr (VI)               | 6.8  | 933.88                            | PSO      | [128]     |
| $\gamma$ -Fe <sub>2</sub> O <sub>3</sub> @PANI               | As (V)                | 4–11 | 90.4                              | PSO      | [129]     |
| Core-shell ThO <sub>2</sub> -PANI                            | Cr (VI)               | 5    | 141                               | PSO      | [130]     |
| PANI/MnO <sub>2</sub> /TiO <sub>2</sub>                      | Cr (VI)               | 2    |                                   | PSO      | [131]     |
| MWCNTs @PANI@TiO <sub>2</sub>                                | As (III)              | 2–9  |                                   | PSO      | [132]     |
| (Fe <sub>3</sub> O <sub>4</sub> @UiO-66@PANI)                | Cr (VI)               | 2    | 474.42                            | PSO      | [133]     |
| PANI/LDH                                                     | Cr (VI)               | 6    | 110                               | PSO      | [134]     |
| PPy-PANI nanofibres                                          | Cr (VI)               | 2    | 227                               | PSO      | [135]     |
| PANI @Ni(OH) <sub>2</sub>                                    | Cr (VI)               | 4    | 625                               | PSO      | [136]     |
| PANI-kapok fiber                                             | Pb (II)               | 6    | 63.60                             | PSO      | [137]     |
| PANI/Ag (AMPSA)/ GO QDs NC                                   | Cr (VI)               | 2    | 59.96                             | PSO      | [138]     |
| PANI/CuO                                                     | Cr (VI)               | 3    | 15.62                             | PSO      | [139]     |
| CuFe <sub>2</sub> O <sub>4</sub> /PANI                       | Hg <sup>2+</sup>      | 7    | 157.1                             | PFO      | [140]     |
| Biocarbon/PANI nanofiber                                     | U(VI)                 | 4    | 282.2                             | PFO      | [141]     |
| PANI/PPy/M-SBA-15                                            | Hg <sup>2+</sup>      | 6.7  | 346.2                             | PSO      | [142]     |
| (Fe <sub>3</sub> O <sub>4</sub> @ mSiO <sub>2</sub> @CS@PANI | Cr (VI)               | 2    | 249.6                             | PSO      | [143]     |
| PANI/LDOs                                                    | Cr (VI)               | 2    | 409.77                            | PSO      | [144]     |
| r-GO/PANI/ZnO                                                | Cr (VI) and Cd (II)   | 7    | 22.522 Cr (VI) and 12.033 Cd (II) |          | [145]     |
| PANI@WO <sub>3</sub>                                         | Cr (VI)               | 2    | 549.37                            | PSO      | [146]     |

**Note:** PSO stands for Pseudo 2nd order, and PFO stands for Pseudo First order.



**Fig. 7.** PANi and CS-based nano-structures as reusable adsorbents and reductants for removing Cr(VI) from impure water by adsorption/reduction-based technique. [Reprinted (adapted) with permission from {ACS Appl. Nano Mater. 2021, 4, 2, 1831–1840}. Copyright {2021} American Chemical Society].

involved in the process.

Thabiso Carol Maponya et al. investigated a modified polypyrrole/m-phenylenediamine (PPy-mPD) composite integrated with magnetite (Fe<sub>3</sub>O<sub>4</sub>) NPs. These NCs demonstrated remarkable efficiency in removing 99.6 % of hexavalent Chromium (Cr(VI)) from a 100 mg/L solution under conditions of pH 2.0 and 25 °C. The study also explored the performance of the NCs across a range of Cr(VI) concentrations (100–600 mg/L) and temperatures (15–45 °C) [154].

PPy/m-phenylenediamine-based NCs show a good adsorption capacity for Cr(VI) metal ions, i.e., 555.6 mg/g at 2 pH [154]. A ternary NCs of PPy, CoO, and Graphene shows an even higher adsorption capacity, i.e., 780.36 mg/g for Pb(II) and 794.18 mg/g for Cd(II) ions at 5.5 and 6.1 pH, respectively [155]. The highest adsorption capacity was observed in PPy/CH<sub>4</sub>/Fe<sub>3</sub>O<sub>4</sub> NCs, which is 922.49 mg/g for adsorbing Cr(IV) ions [158].

Das. et al. synthesized a thiol-functionalized PPy composite and

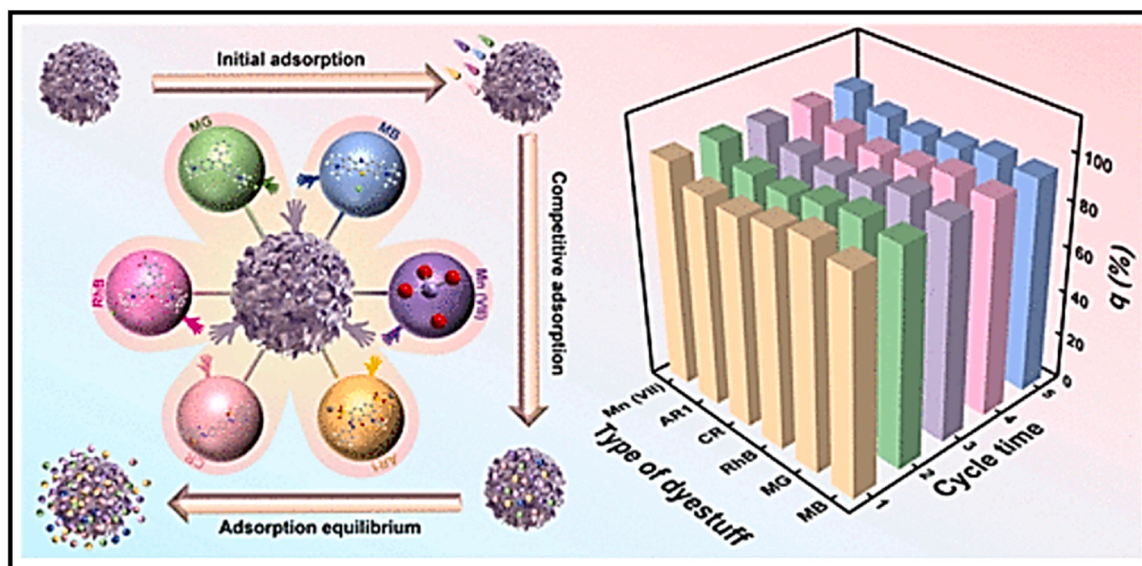


Fig. 8. Systematic results obtained from different adsorbent type, duration of exposure, adsorbate concentration, and temperature on the adsorption efficiency of NCs. [Reprinted (adapted) with permission from {Langmuir, 2023, 39, 29, 10098–10111}. Copyright {2023} of ACS].

Table 3

The Effectiveness of PPy-based NCs towards dye and toxic metal removal in water treatment.

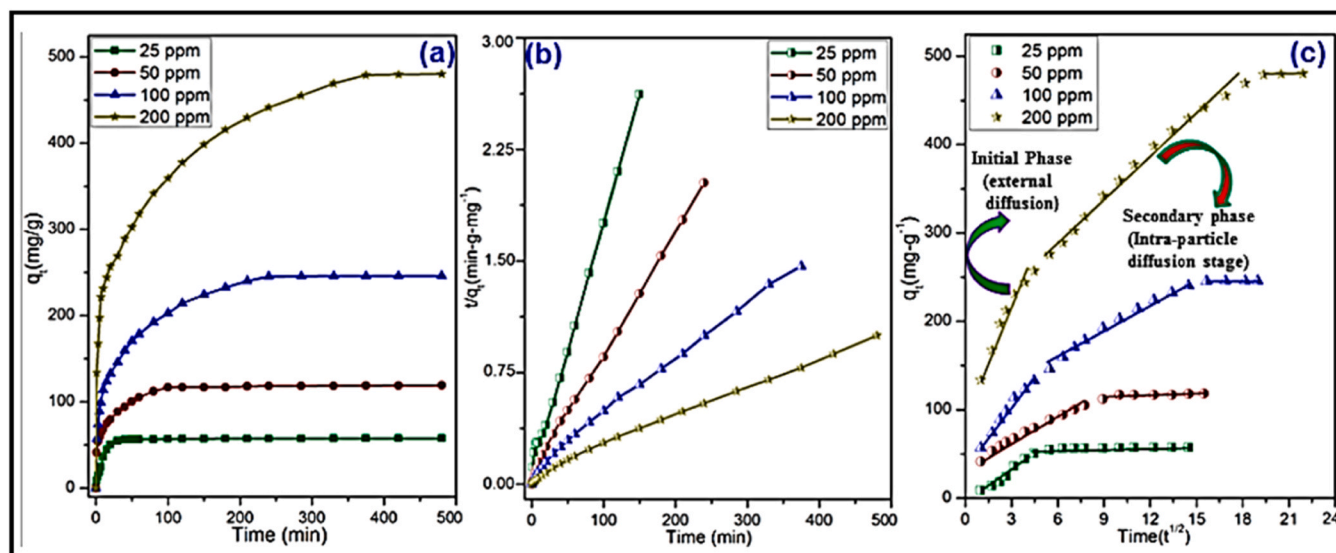
| PPy Based NCs                                                     | Target metal                           | pH                       | Adsorption Capacity (mg/g)        | Kinetics        | Reference |
|-------------------------------------------------------------------|----------------------------------------|--------------------------|-----------------------------------|-----------------|-----------|
| PPy/ monodispersed latex sphere                                   | Cr(VI)                                 | 2                        | 343.64                            | quasi 2nd order | [152]     |
| PPy-Fe <sub>3</sub> O <sub>4</sub> -SW                            | Pb(II)                                 | 5                        | 333.33                            | PSO             | [153]     |
| Polypyrrole/ <i>m</i> -Phenylenediamine                           | Cr(VI)                                 | 2                        | 555.6                             | PFO             | [154]     |
| PPy/ Cobalt oxide Graphene (COPYGO)                               | Pb (II) & Cd (II)                      | 5.5 Pb (II), 6.1 Cd (II) | 780.363 Pb (II) & 794.188 Cd (II) | PSO             | [155]     |
| PPF@PANi + PPy                                                    | Cr(VI)                                 | 2                        | 510.9                             | PSO             | [156]     |
| PPY/Fe <sub>3</sub> O <sub>4</sub>                                | Cd (II)                                | 6                        | 8.928                             | PFO             | [157]     |
| Poly(pyrrole methane) (PPm) / Fe <sub>3</sub> O <sub>4</sub>      | Cr(VI)                                 | 2                        | 922.49                            | PSO             | [158]     |
| Polypyrrole/nano cellulose (PPy/NC)                               | Cr(VI)                                 | 2                        | 560                               | PSO             | [159]     |
| (Magnetic UiO-66@PPy)                                             | Cr(VI)                                 | 7                        | 259.1                             | PSO             | [160]     |
| PPy-polyethyleneimine (PPy-PEI)                                   | Pb (II)                                | 10                       | 75.60                             | PSO             | [161]     |
| (PPy-PEI)                                                         | Ni (II)                                | 4.5                      |                                   | PFO             | [162]     |
| Maghemite-polypyrrole                                             | CR                                     | 7                        | 269.5                             | PSO             | [163]     |
| Magnetic-Fe <sub>3</sub> O <sub>4</sub> /Polypyrrole/Carbon Black | CR & MB                                | 10 (CR) 8 (MB)           | 227.7 (CR) & 78.6 (MB)            | PSO             | [164]     |
| (PPy-PEI)                                                         | MB                                     | 6.2                      | 183.3                             | PSO             | [165]     |
| PPy/ Fe <sub>3</sub> O <sub>4</sub> /SiO <sub>2</sub>             | CR                                     | 4                        | 361                               | PSO             | [166]     |
| FeWO <sub>4</sub> /PPy                                            | Rose Bengal (RB), Alizarin Red S (ARS) | 6                        | 202.63 (RB) 142.80 (ARS)          | PSO             | [167]     |
| Polypyrrole-based KOH-activated carbon                            | CV                                     | 6.3                      | 497.51                            | PSO             | [168]     |
| PPy - Fe <sub>3</sub> O <sub>4</sub> - SW                         | MB                                     | 10                       | 666.66                            | PSO             | [169]     |
| Polypyrrole@magnetic CS                                           | MO                                     | 4.5                      | 95                                | PSO             | [170]     |
| PPy-Fe <sub>3</sub> O <sub>4</sub> -SW                            | CR                                     | 3                        | 500                               | PSO             | [171]     |
| RGO-CuS-PPy                                                       | Ni(II)                                 | 6.0                      | 166.67                            | PSO             | [172]     |
| PPy/MAA                                                           | Ag (I)                                 | 5.4                      | 714.28                            | PSO             | [173]     |

Note: PSO stands for pseudo 2nd order and PFO stands for pseudo 1st order.

investigated its potential for the removal of hazardous Hg(II) ions from aqueous solutions at concentrations ranging from 100 to 800 ppm. The results showed that high levels of effectiveness in mercury removal occurred, which are primarily due to the mercaptoacetic acid thiol functionality providing strong interactions with the soft acid Hg(II). The kinetic behavior for the adsorption follows pseudo 2nd order with a maximum adsorption capacity up to 1736.8 mg/g at 25 °C as estimated using the Langmuir isotherm. Results indicate that the PPy/MAA (Mercaptoacetic acid) composite may hold much promise for mercury ion remediation. Fig. 9 discusses work on the dynamics of Hg(II) ion adsorption onto an adsorbent. Samples were initially dosed at 25, 50, 100, and 200 mg/L and maintained at 25 °C in this study (Fig. 9(a)). We use a pseudo-second-order model to describe Hg(II) adsorption onto

PPy/MAA composites. The model shows that adsorption is site-dependent, and the amount of Hg(II) adsorbed is proportional to the square of the residual concentration in solution, as depicted in Fig. 9(b). Fig. 9(c) employs the use of an intraparticle diffusion model to see how Hg(II) ions adsorb onto the PPy/MAA. This model will tell us if the diffusive step inside the composite pores affects the global process in terms of kinetic and efficiency of adsorption [174].

Raghunath Das et al. reported environmentally destructive Ag<sup>+</sup> ion remediation response was prepared by synthesizing a PPy derivative containing mercapto groups. FESEM, EDS, HRTEM, STEM, XRD, FTIR, and XPS data ensured that the adsorbent PPy/MAA was produced appropriately, with a strong attraction to the silver ions. The balance adsorption capacity of 714.28 mg/g was accomplished at 25 °C with the



**Fig. 9.** (a) Hg(II) adsorption kinetics at 25 °C with concentrations of metal ions of 25, 50, 100, and 200 mg/L. (b) Model of the Hg(II) ion adsorbed on the PPY/MAA composites that are pseudo-second-order (c) The Hg(II) ions' adsorption onto the PPY/MAA composite using an intraparticle diffusion model. [Reprinted (adapted) with permission from {ACS Sustainable Chem. Eng. 2017, 5, 7524 –7536}. Copyright {2017} American Chemical Society].

pH solution at 5.4. The data fits well into the pseudo 2nd order kinetics to show an efficient removal of silver ions in the experiment. The PPY/MAA composite successfully captured  $\text{Ag}^+$  ions from water solutions and converted them into metallic silver NPs ( $\text{Ag}^0$ ). Fig. 10 shows the adsorption enhancement techniques of  $\text{Ag}^+$  from liquid solutions by PPY/MAA composite visualized. PPY is a CP used with methacrylic acid to have functional groups that increase the ionic affinity of the material (Fig. 10 (a)). The isotherms show how much adsorbate- $\text{Ag}^+$  ions will adsorb onto an adsorbent at a certain temperature equilibrium. Isotherms will tell us how good the composite is and how good it is in removing ions from solutions and this is as summarized in Fig. 10 (b). Also, isotherm plots in Fig. 10(c) represent the composite material PPY/MAA, which has a potential for the adsorption of  $\text{Ag}^+$ . Its properties describe it as a material of suitable quality for real application toward water treatment [175].

Water samples of drinking, ground, river, and generated water qualified as good representatives of water matrices from which PPY effectively preconcentrated, thereby allowing the determination of targeted metal ions. The results indicate the potential applicability of PPY for the recovery and extraction of valuable and less desirable ions on a larger scale [176].

#### 4. CS-based NCs for water treatments

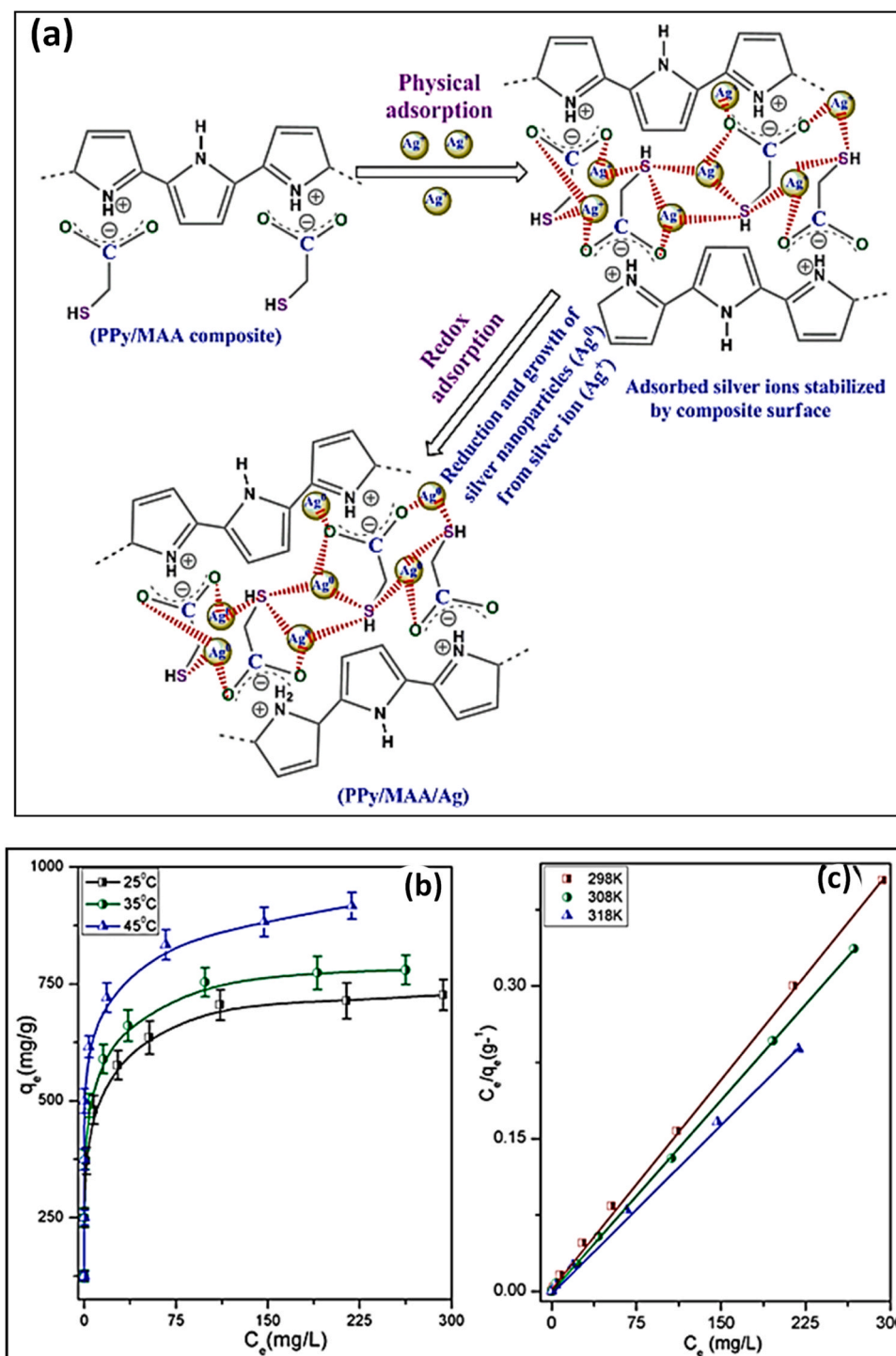
CS is the second-naturally occurring largest polymer, derived from the partial deacetylation of Chitin-a polysaccharide. This exotoxin is a heteropolysaccharide containing D-glucosamine and N-acetyl-D-glucosamine units, which are linked with  $\beta$ -1,4-glycosidic bonds. CS carries free amino and hydroxyl groups, thus fit to formulate functional polymers [177]. CS is widely used as an absorbent for removing different pollutants, like bivalent metal ions and water-soluble organic compounds. These carbohydrate polymers are also known to be low-cost sorbents compared to commercial activated carbons and synthetic ion exchange materials. Chitin is the 2nd abundant natural polymer on earth and may be procured from crustaceans, such as krill, crayfish, and crabs through three stages: (i) acid treatment to break down calcium carbonate, (ii) alkaline extraction to dissolve proteins, and finally, (iii) the cleaning stage, which involves disinfection and color elimination [178, 179]. CS is a very well-known biopolymer that considers its hydrophilic properties, its biodegradability, and its antibacterial character. This biopolymer is very abundant among the naturally occurring types of

biopolymers and is derived from the deacetylation of Chitin. Being highly abundant in amino and hydroxyl groups, CS is advantageous for adsorption due to its molecular structure. It can chelate or adsorb via amine and hydroxyl functional groups to some active sites. It is for this reason that CS is employed widely as a potent biosorbent toward synthetic dyes and heavy metal ions in wastewater without undergoing any elaborate pre-treatment of the dyes, even at low concentration levels, even with various derivatives [180]. CS NCs have different applications, including pollutant removal, photocatalysis, medicine, energy generation, food production, agriculture, and electronics. Their large surface area enhances their capacity to capture pollutants, making them innovative technologies in various fields [181]. One of the challenges against its use in natural CS is that its adsorption power is relatively low and would be enhanced through physical or chemical modification. To overcome this challenge, scientists have resorted to synthesizing CS-based adsorbents whose performance can be relative to the more efficient [182].

CS NPs are ideal for decontaminating polluted water since they can survive extreme conditions without reacting with other chemicals [183]. CS NPs were first made by mixing CS with toluene and glutaraldehyde that connects the CS molecules, using the CS amino groups and the chemical's aldehyde groups. In this method, span 80 was used to keep the mixture stable, and toluene and glutaraldehyde were used to link the CS in a mix of water and oil. The mix was stirred, making tiny droplets that became the base for the NPs. These NPs are taken out of the mix by spinning them in a machine called a centrifuge, then washed many times with different liquids like petroleum ether, acetone, sodium metabisulfite, and water, and finally dried either by taking out the air (vacuum) or freezing [184,185].

Ohya first introduced CS NPs in 1994, pointing out their possible uses in medicine. Specifically, Divya and Jisha [186] showed how ChNPs could be used to deliver 5-fluorouracil, a drug that fights cancer, through the veins. This was done using methods like emulsification and crosslinking. Since then, different ways to make CS NPs have been created, with five methods, i.e., ionotropic gelation, micro-emulsion, emulsification, solvent diffusion, polyelectrolyte complex formation, and the reverse micellar method. Out of these, ionotropic gelation and polyelectrolyte complex formation are the most popular because they are simple and don't require strong forces or harmful solvents [186, 187].

Besides the fact that CS on its own contains different functional



**Fig. 10.** (a) An Overview of designed adsorption techniques for Silver ions onto the PPy/MAA composite". (b)  $\text{Ag}^+$  ion adsorption isotherm onto PPy/MAA composite. (c) Plots illustrating the Langmuir adsorption isotherm. [Reprinted (adapted) with permission from {ACS Sustainable Chem. Eng. 2017, 5, 2711–2724}. Copyright {2017} American Chemical Society].

groups that can easily join with modifications to give any kind of product, the chemical modifications raise its solubility of it which allows for the extension of its usage afield. Showing great biocompatibility, biodegradability, and ease of modification, CS NPs are valuable carriers for drug delivery and vaccine applications for use in vaccine adjuvants, antimicrobials, and tissue engineering [188,189].

CS's most common alterations are graft copolymerization, cross-linking, esterification, carboxymethylation, and etherification [190]. CS cross-linking takes place essentially by reacting amino groups with

cross-linking agents like dialdehydes, including glyoxal and glutaraldehyde, and some organic acids. Through this chemical modification, the properties of CS membranes have been improved significantly concerning proton conductivity. Proton conductivity measured in these cross-linked CS membranes falls within the suitable range for applications in the field of biopolymer membranes [191,192].

#### 4.1. Modification of CS to enhance solubility through carboxylation

CS derivatives have been prepared to enhance their bioactivity and solubility in water. Carboxylation has been one of the standard methods for chemical modification. CS can be readily solubilized in an aqueous solution at room temperature using a mixture of isopropanol and sodium hydroxide. Then, mono-chloroacetic acid and additional isopropanol are added to dilute the formed suspension. The solution's pH value is a critical factor in determining the solubility of this amphoteric CS polymer. Interestingly, even though the O- and N-carboxymethylation can co-occur during the reaction, it is still possible to achieve either modification separately. This control of the response is essential for the tailored derivatives of CS for a specific end [193,194].

An esterification is another established chemical alteration of CS, which also has many scientific studies. Another principle, through the process, the use of acetyl chloride as a solvent instead of  $\text{MeSO}_3\text{H}$  helps predominantly the production of O-acetylated CS instead of N-acetylated CS. The O-acetylation of CS has been demonstrated to be the reason for the improvement of its antifungal efficacy; thereby, it has become one of the most significant changes for its application in antimicrobial agents and antifungal formulations. The property that makes it possible to acetylate only the CS is the advantage that makes it also functional and utilized in biomedical and agricultural areas [195,196].

#### 4.2. Phosphorylation of CS

##### 4.2.1. Development of antimicrobial and Osteo-inductive derivatives

Modifications are crucial because they have displayed antimicrobial and osteo-inductive properties. CS obtains phosphorylation when heat-treated with orthophosphoric acid, which enables the production of modified derivatives with enhanced functional properties that may be appropriate for different applications. Additionally, the reaction of methane sulfonic acid and phosphorus pentoxide might also yield highly effective phosphorylated CS. Such a reaction increases hydroxyl phosphorylation at carbons 3 and 6 of the CS structure, hence enhancing the integrity of the cell wall. The ability to synthesize phosphorylated CS derivatives is of extreme interest in biomedical applications, mainly in improving the performance of biomaterials [186,197]. In environmental projects, making CS, a type of material, better at grabbing metal ions by adding a substance called EDTA has been proven to work well. EDTA is good at forming strong bonds with different metal ions, which helps clean up water with metals in it. This is important for protecting the environment by removing harmful metals from wastewater.

Also, EDTA can change how metals interact with water pollutants, especially metal ions, which might affect how harmful these metals are to them. By making CS better at catching metals with the help of EDTA, we can make it more useful for cleaning up water, which is important for dealing with the problem of metal pollution in water environments. This method helps to quickly take out metal from impure (hard) water and helps in the softening of hard water [198]. CS is a material that can remove heavy metals from water because it has special spots that help it grab onto metal ions using a process called ion exchange and coordination bonds. The ability of CS to do this depends mainly on two things: how big its particles are and how much surface area it has. Normally, CS comes in flakes or powders that are not porous, meaning they have a surface area of about 10 square meters per gram [199].

CS NPs onto the surface of fibrous carriers, resulting in the formation of a composite layer. The resultant CS particles immobilized onto the fiber were evaluated for their ability to adsorb anionic dyes. Anionic phthalocyanine dyes, differing by their molecular size and structural properties and bearing a different number of anionic functional groups, were used as model adsorbates. For this reason, the adsorption behavior of these dyes onto CS NPs Fibrous carrier was investigated to understand the mechanisms of interaction and adsorption capacity of CS NPs [200, 201].

#### 4.3. Properties of CS

##### 4.3.1. Deacetylation of CS

Its deacetylation can be done by several chemical and biological methods since it is the main process occurring in the amorphous regions before it can affect the crystalline domains. The chemical methods mainly consist of acidic or alkaline treatments under a nitrogen atmosphere or the addition of sodium borohydride to the solutions of sodium hydroxide, which reduces the side reactions that take place undesirably. On the other hand, enzymatic hydrolysis is ecologically friendly but expensive and hence mainly used for laboratory-scale studies only. Different analytical techniques like acid-base titration, potentiometric titration, conductometric titration,  $^1\text{H}$  NMR spectroscopy, elemental analysis, Fourier transform infrared spectroscopy, ultraviolet spectrophotometric analysis, capillary zone electrophoresis, and Raman spectroscopy have been utilized to date by researchers for the structural determination of CS. It allows the correct characterization of the chemical properties of CS and all its possible applications attributed to it [202].

##### 4.3.2. Classification according to molecular weight

Basically, CS is categorized into four forms according to molecular weight: high molecular weight, medium molecular weight, low molecular weight, and oligochitosan. In this context, the molecular weight of CS was found to affect the physicochemical and antimicrobial properties. The HM-CS inhibited the process of ripening and fruit spoilage more effectively. Although higher MW CS films have many superior film-forming properties and tensile strength, they have reduced antibacterial activity against Gram-negative bacteria, such as *Escherichia coli*, because they cannot diffuse through bacterial membranes. On the other hand, lower MW CS exhibited increased antimicrobial activity against Gram-negative bacteria by passing through microbial cells, thus resulting in the inhibition of mRNA synthesis. Its effectiveness against Gram-positive bacteria is still under study [203,204].

##### 4.3.3. Antimicrobial properties of CS

It was Allan and Hadwiger first to report the excellent antimicrobial properties of CS. CS was then more effective against a broader range of fungi than chitin and also showed broad fungicidal activities afterward. Subsequently, different investigations were done to reveal the mechanism of action underlying CS as an antimicrobial agent; however, no consensus was obtained. The mentioned effects are mainly through the electrostatic attractions of the positively charged amino groups with negatively charged cellular membranes of CS, stimulation of host defense mechanisms, and therefore disruption and scavenging of metals necessary for microbial growth, followed by reduced cell damage due to modification in permeability, creating a polymeric layer. The polycationic property of CS can help disrupt the membrane when the pH is below 6.3, ultimately leading to cell death by the leakage of intracellular components. Such a mechanism is particularly effective against Gram-positive bacteria of *Staphylococcus aureus*. On the other hand, increased lysis and permeability have been reported with Gram-negative bacteria of *Escherichia coli* [205,206].

##### 4.3.4. Self-healing properties of CS

CS-based self-healing materials, in particular self-healing anticorrosion paints, have gained wide interest because of their huge potential to act against corrosion on metallic and automobile surfaces. However, getting an optimal balance between mechanical strength, self-healing properties, and toughness represents one of the most important challenges. Effective methods to improve corrosion protection in these coatings include the incorporation of corrosion inhibitors and the addition of polymerizing agents to repair defects. Noticeably, the CS- and Cerium-based coatings proved to be effective for the protection of aluminum alloy 2024, and 2-Mercaptobenzothiazole (MBT) showed an inhibition effect. Further, chemical grafting efficiently improved the

hydrophobicity of the coatings with polyethylene-alt-maleic anhydride and poly-maleic anhydride-alt-1-octadecene, thus slowing down the ingress of electrolytes. The synergistic effects developed by MBT through active corrosion protection and those of CS by grafting, conferring hydrophobicity of aluminum alloys and hence lesser corrosion were proved [207].

#### 4.4. Properties of CS-based NCs (CSNCs)

Several key properties qualify CS-based NPs for environmental applications. They are biocompatible and biodegradable, ensuring living organisms' safety and minimizing environmental effects in case they degrade without toxic residues [208]. Specifically, high surface area and porosity could considerably improve their ability to absorb water-to-be-treated environment pollutants like heavy metals and dyes [209]. Being positively charged, CS NPs easily attract strongly negatively charged pollutants; hence they have high application potential in water treatment plants [210]. The ability to be functionalized further adds the ability to selectively remove contaminants like pesticides or pharmaceuticals through this technology [211]. In addition, CS NPs are recyclable and reusable multiple times along the purification of impure water [212].

Different properties of CPs includes biocompatible, biodegradability, antimicrobial, cationic nature, pH sensitivity, chemical scalability, antioxidant, large surface area, non-toxic and permeation enhancement properties of CS-based (NPs).

#### 4.5. Methods of preparation of CS NCs

The process known as "ion gelation" involves the cross-linking of CS with a polyanionic compound, typically sodium tripolyphosphate (STPP). Ionic interactions throughout the process result in the formation of NPs [213]. Self-assembly is the process wherein CS molecules assemble themselves in the presence of surfactants or other stabilizers to form CS NPs. Due to this method, hydrophobic and electrostatic interactions will be helpful for the production of NPs [214,215]. The

electrospinning technique was applied to create nanofibers of CS that can subsequently be fabricated into NPs. These nanofibers can effectively adsorb pollutants in water [216,217]. The coacervation technique is used for the separation of CS phases to form NPs. These can be applied in the removal of contaminants like heavy metals and dyes from water [218]. The solvent evaporation method of preparing NPs involves dissolving CS in a solvent and allowing the solvent to evaporate [219]. Different preparation strategies of CS NPs are also shown in Fig. 11.

The electrospinning technique encompasses the dissolution of CS within a solvent. By applying high voltage, one can generate a charged mist, the emitted fibers must undergo drying and then aggregate into NPs. Dissolve CS in an acidic solution, and then introduce a crosslinking agent that triggers gelation that results in the creation of NPs. The coacervation method consists of dissolving CS in an acidic solution, then introducing a non-solvent to trigger phase separation resulting coacervate phase being collected to create and stabilize NPs. The process of making CS-based NPs using the evaporation method involves dissolving CS in an appropriate solvent, applying it to a surface by spraying or casting, and then allowing the solvent to evaporate, resulting in the formation of aggregated NPs.

#### 4.6. Applications of CS-based NCs in water treatment

##### 4.6.1. Heavy metal removal

CS is a very promising adsorbent able to absorb dangerous metal ions from an aqueous medium due to the multiplicity of chelation sites in the form of hydroxyl (-OH) and amino (-NH<sub>2</sub>) groups. These functional groups aid in binding the metal ions through covalent bonding or ion exchange. A lot of focus on the methods of surface modification for the removal enhancement of heavy metals has been devoted to surface modification aimed at enhancing the adsorbent effectiveness of CS [220, 221].

##### 4.6.2. Dye removal

CS-based composites prove particularly efficient in the removal of dyes because of their biocompatibility and large surface areas. NCs

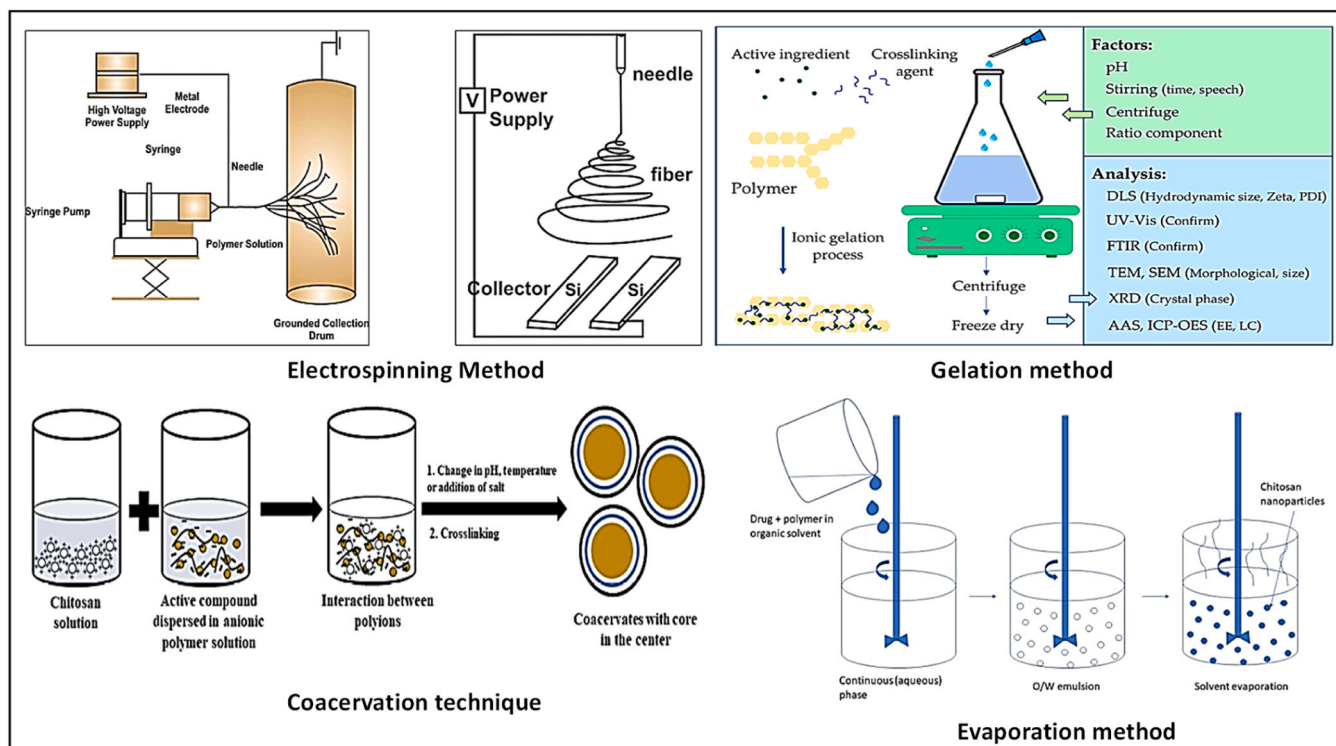


Fig. 11. Different methods of preparation of CS NPs.

impregnated with sodium alginate in conjunction with graphene oxide have recently shown outstanding levels of sorption of dyes. The maximum sorption capacities achieved were 909.1 mg/g for CV and 714.3 mg/g for Congo red. These results enable the use of CS-based NCs in selective dye removal through water treatment processes [222].

#### 4.6.3. Bacterial inactivation

CS-based composites increasingly attract great attention since they can be applied in water treatment, especially for bacterial inactivation. Of particular interest is the CS/TiO<sub>2</sub>-C, N membranes that ensure efficient and long-lasting microbial removal. These membranes have fully removed *Escherichia coli* through photocatalytic processes, thus proving the efficiency of their disinfectant properties. Moreover, they are easy to withdraw from the aqueous media, thus ensuring multiple uses. This case is just one of the many great prospects of CS-based NCs for the improvement of water treatment technology [223].

#### 4.6.4. Flocculation and coagulation

CS possesses several remarkable properties that enable it to function effectively as a flocculant and coagulant in the removal of dissolved impurities. Its long polymer chains, bridging capabilities, and high cationic charge density work together to facilitate the precipitation and aggregation of pollutants, particularly in neutral or alkaline pH conditions. The primary benefits of CS are its nontoxicity and biodegradability, making it environmentally friendly for water treatment applications. Additionally, its excellent chelation behavior enhances its capacity to bind with different pollutants, from dissolved substances to particulate matter. These characteristics have been utilized to develop coagulation and flocculation processes for treating various types of wastewater discharges [224].

#### 4.7. Treating different Pharmaceuticals and Personal Care Products (PPCPs) using CS based NCs

CS based NCs have a prominent potential for the adsorption of PPCPs from wastewater, representing the versatility of these nanomaterials against various contaminants. The increasing presence of PPCPs in aquatic environments lent great concern to the environment and toxicity, thus demanding the development of strategies that would make sure of their effective removal.

The mechanism of adsorption by CS NCs includes several

interactions, such as hydrogen bonding, hydrophobic interactions, and electrostatic attractions, which facilitate PPCPs' attachment to the NPs surface. On account of such a multifunctional adsorption capacity, CS NPs are effective against a wide spectrum of contaminants, encompassing a good variety of organics commonly found in wastewater [225]. A short version of the applications in the form of a flow chart is also depicted in Fig. 12.

#### 4.8. CS-based NCs in dye removal

It has been suggested far back that CS NPs derivatives with improved electrostatic and magnetic properties could be used in wastewater treatment [227]. NPs used in the treatment were found to be applied to both cationic and anionic dyes, including MB, Crystal Violet, Brilliant blue-R, Rh-B, MO, CR, Alizarin red S, and Eosin Y. [228].

To further improve the removal of dye from wastewater, Peng Huang et al. study led to the creation of a cationic polyelectrolyte from CS, which was then combined with the traditional cationic organoclay. With the 10/90 wt. ratio, this hybrid material removed all these classes of anionic dyes while increasing the ability to capture the dye by 370 % and adsorbed the dye five times faster than that of the case with only organoclay. The results of the study, therefore, make it eligible for application to enhance industrial adsorbents since it established that the polyelectrolyte interacts with dyes efficiently and exhibits various adsorption characteristics for different dyes [229]. Fig. 13 represents the steps involved in the elimination of anionic dyes with the help of a composite adsorbent formulated from the cationic polymer N-[(2-hydroxy-3-trimethyl-ammonium)propyl] Chitosan Chloride (HTCC) and organoclay Organically Modified Montmorillonite (OMMT) in water. A schematic diagram to show which section of HTCC can be focused on for optimization to further improve the dye loading capacity of the hybrid adsorbent system is depicted in Fig. 13.

The incorporation of amino-functionalized silicate NPs, OCT-MMT, into the work by Seyed Abolhassan Hosseini improved the removal ability of dyes from electro-spun nanofibrous membranes made of CS/PVA. The optimal incorporation of 2 wt% OCT-MMT improved the porosity and adsorption capacity of the dyes; this effect was detected through an improvement in the rate and efficiency of removal. This ensured that the mechanical and thermal stabilities were improved, and thus these membranes could be reused repeatedly without losing activity. In the case of adsorption, the Langmuir isotherm and pseudo 2nd

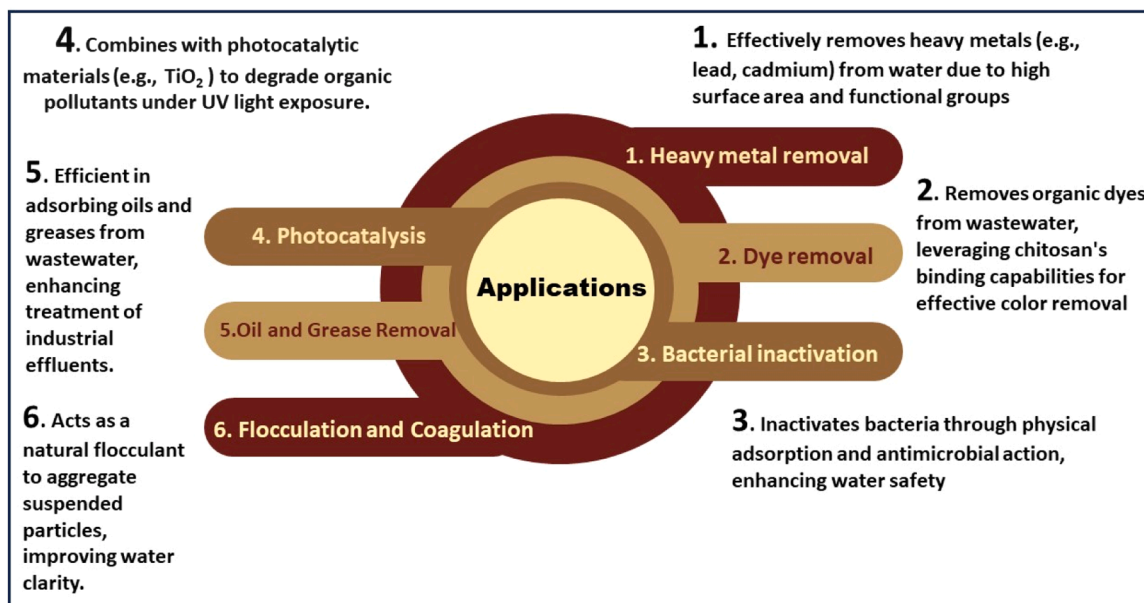
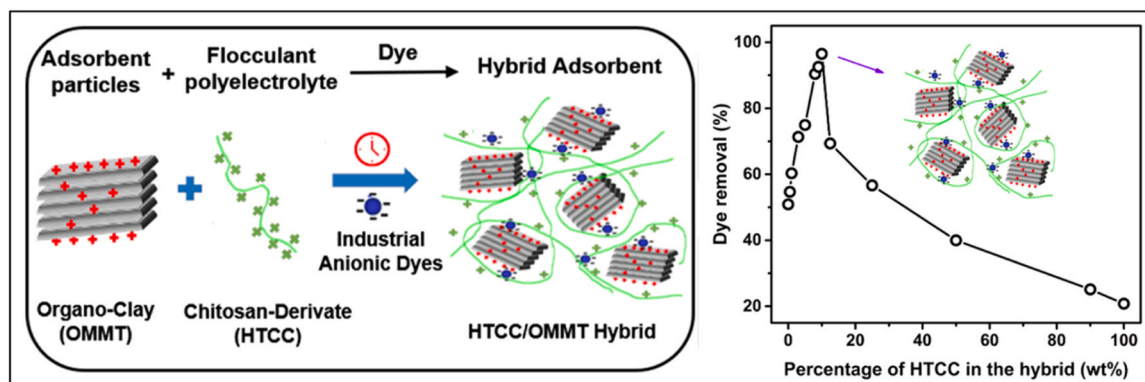


Fig. 12. Applications of CS-based NCs in water purification [226].

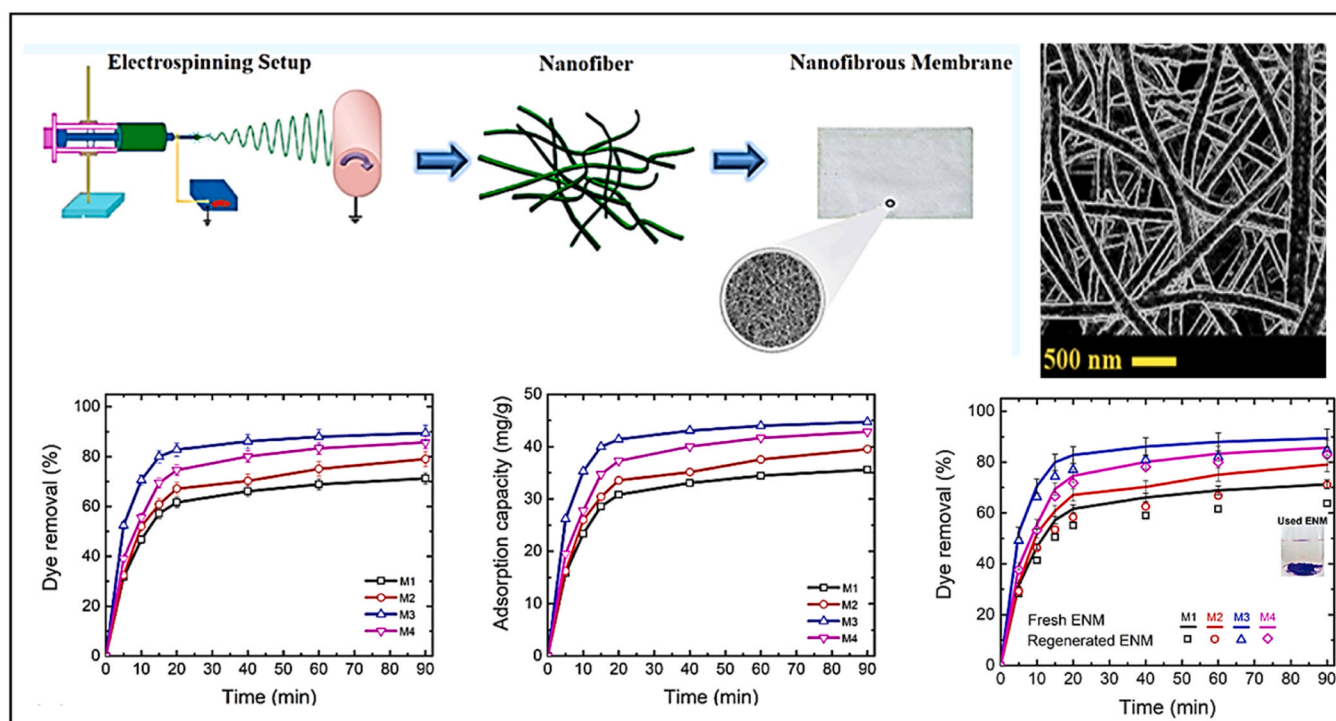


**Fig. 13.** (a) Removal of anionic dye by cationic polyelectrolyte (HTCC)/organo-clay (OMMT) hybrid adsorbent and degradation of hydrolyzed Reactive Black 5 from aqueous solutions (pH 7, 20 C, dye concentration of 100 mg/L, stirred for 26 h), by HTCC/OMMT hybrids with an increasing percentage of HTCC (constant total HTCC/OMMT loading of 80 mg/100 ml). [Reproduced with permission from {ACS Appl. Mater. Interfaces 2019, 11, 11961–11969}. Copyright {2019} American Chemical Society].

order kinetics could describe the best process, an indication of endothermic spontaneous nature [230]. They prepared electro-spun membranes using amino-functionalized montmorillonite NCs material. These NCs were specifically designed to improve adsorption capacity and exhibit strong thermomechanical properties. The synergy of these features enhances its performance across various applications by leveraging the benefits of electrospinning. As reflected in the back-scattered SEM image, NPs of MMT were well dispersed in the structure of the composite fiber of the M4 sample. This further indicates excellent dispersion of the NPs, which improved the overall features of the fibres. Experiments evaluating adsorption efficiency (at a pH level of 7) were executed with a consistent dosage of 0.04 g of OCT-MMT. Additionally, different concentrations of dye in the solution—ranging from 5 to 50 mg/L—were

employed. The impacts of dosage variations on the octadecyl methacrylate montmorillonite emerged as some of the most crucial factors affecting adsorption efficiency for dye removal. However, this situation prompts inquiries regarding the optimal concentration necessary to enhance effectiveness. Although the findings suggest a relationship between dosage and efficiency, the precise mechanisms remain to be completely clarified. Because of these intricacies, the study underscores the necessity for continued research in this domain. The assessment of the reusability of NCs membranes was performed by evaluating their effectiveness in dye removal (specifically) across two separate operational cycles and all this is systematically aggregated in Fig. 14.

Table 4 depicts the efficiency of CS-based NCs in different dye removal. Different types of NCs used, target dye involved, pH,



**Fig. 14.** Amino-functionalized montmorillonite nanocomposite electro-spun membranes created with exceptional adsorption capacity and thermomechanical traits, the back-scattered SEM image indicates that MMT NPs are uniformly integrated within the fiber network of the M4 specimen to be in the range of 500 nm. The impact of OCT-MMT concentration (regarding dye removal) was examined at a pH level of 7, regarding reusability, the evaluation of NCs membranes showed that they displayed consistent dye removal capabilities over two operational cycles. [Reproduced under creative commons open access license from {ACS Omega 2021, 6, 10816–10827}. Copyright {2021} American Chemical Society].

**Table 4**

The efficiency of CS-based NCs in different dye removal.

| Types of NCs                                                                                         | Targeted dye                     | pH               | Adsorption capacity (mg/g) | kinetics  | Reference |
|------------------------------------------------------------------------------------------------------|----------------------------------|------------------|----------------------------|-----------|-----------|
| CS-TiO <sub>2</sub>                                                                                  | MB                               | 6                | 200                        |           | [231]     |
| CS/NiO                                                                                               | Congo Red                        | 6                | 238.1                      | PSO       | [232]     |
| CS/ ZnO                                                                                              | Eriochrome Black T               | 3.5              | 40.9                       | PSO       | [233]     |
| CS/Silica/ZnO                                                                                        | MB                               |                  | 293.3                      | PSO       | [234]     |
| CS/Graphene oxide                                                                                    | MB                               | 7                | 178                        |           | [235]     |
| CS/ZrO <sub>2</sub>                                                                                  | Safranin O                       | 10               | 280.14                     | PFO       | [236]     |
| CS/CuMn <sub>2</sub> O <sub>4</sub>                                                                  | MB                               | 7                | 54.05                      | PFO       | [237]     |
| CS/Fe <sub>2</sub> O <sub>3</sub>                                                                    | Acid blue 113                    | 3–11             | 128                        | PSO       | [238]     |
| CS/Fe <sub>2</sub> O <sub>3</sub> /NiFe <sub>2</sub> O <sub>4</sub>                                  | Methyl Green                     | 8                | 77.22                      | PSO       | [239]     |
| Bentonite/CS/Co <sub>3</sub> O <sub>4</sub>                                                          | Congo Red                        | 2–8              | 303                        | PSO       | [240]     |
| CS/SiO <sub>2</sub> /Carbon nanotubes                                                                | Direct Blue 71, Reactive Blue 19 | 6.8 (DR), 2 (RB) | 61.35 (DB), 97.08 (RB)     | PSO       | [241]     |
| CSGOIO Hydrogel NCs                                                                                  | MB                               | 11               | 36.2                       | PSO       | [242]     |
| CS-benzil/ZnO/Fe <sub>3</sub> O <sub>4</sub>                                                         | RBBR                             | 4                | 620.5                      | PSO       | [243]     |
| CS/TiO <sub>2</sub>                                                                                  | Congo red                        |                  | 32                         | PSO       | [244]     |
| CS-tripolyphosphate/TiO <sub>2</sub>                                                                 | Reactive Orange 16.              | 4–10             | 618.7                      | PSO       | [245]     |
| CS-tripolyphosphate/MgO/Fe <sub>3</sub> O <sub>4</sub> (CTS-TPP/MgO/Fe <sub>3</sub> O <sub>4</sub> ) | Reactive blue 19 (RB 19)         | 4.3              | 120.3                      | PSO       | [246]     |
| CS-TiO <sub>2</sub>                                                                                  | Rose Bengal                      |                  | 79.365                     | PSO       | [247]     |
| Graphene oxide/Chitosan                                                                              | Cu ions                          |                  | 2.54 × 10 <sup>1</sup>     | 1st order | [248]     |
| CS/Co-MCM-15                                                                                         | Methyl Orange (MO)               | 6–8              |                            |           | [249]     |
| CS-glyoxal/TiO <sub>2</sub>                                                                          | Reactive Orange 16 dye (RO16)    | 4                | 390.5                      | PFO       | [250]     |
| CaNiFe <sub>2</sub> O <sub>4</sub> /CS                                                               | MB                               | 7.6              | 89                         |           | [251]     |
| CS/Copper oxide (CS/CuO)                                                                             | MO                               | 6–8              |                            |           | [252]     |
| Ch-PANI-CuO                                                                                          | MO dye                           | 4                |                            |           | [253]     |
| CS                                                                                                   | Acid Blue 25 dye                 | 4–6              | 171.4                      |           | [254]     |
| CH/CMC/GO                                                                                            | Brilliant green, MB              | 7                | 1.89, 122                  | 1st order | [255]     |
| Nano-TiO <sub>2</sub> /MWCNT/CS hydrogel composite beads or NTiO <sub>2</sub> -Cht/MWCNT             | MB                               | 7                | 80.65                      | PSO       | [256]     |
| Ghraphene-ZnO-CS                                                                                     | MB, and AR                       | 4–8              | NA                         | PSO       | [257]     |
| SRC/CS                                                                                               | MB and CR                        | 4 and 2          | 97.08 and 80.9             | PSO       | [258]     |

**Note:** PSO stands for pseudo 2nd order and PFO stands for pseudo 1st order.

adsorption, and kinetics of the reaction were given in Table 4 [231–258]. CS/TiO<sub>2</sub> NCs show an adsorption capacity of 200 mg/g for adsorbing MB dye at pH 6 [231]. On replacing TiO<sub>2</sub> with NiO NPs, the adsorption capacity improves slightly, that is, 238.1 mg/g for CR dye at the same pH 6 [232]. With ZnO NPs the adsorption capacity of CS-based NCs decreases significantly, i.e., 40.9 for EBT [233]. When Graphene oxide was mixed with CS, the adsorption capacity did not increase significantly, i.e., 178 mg/g [235]. When combining two metal oxide NPs that is, NiO and Fe<sub>3</sub>O<sub>4</sub> NPs, the adsorption capacity of CS-based NCs decreases significantly, which is 77.22 mg/g for MG at pH 8.0 [239]. CS-based NCs show maximum adsorption capacity in the presence of Benzil, ZnO, and Fe<sub>3</sub>O<sub>4</sub> NPs, which is 620 mg/g for Remazol Brilliant Blue R (RBBR) dye at 4 pH [243].

#### 4.9. CS-based NCs in heavy metal removal

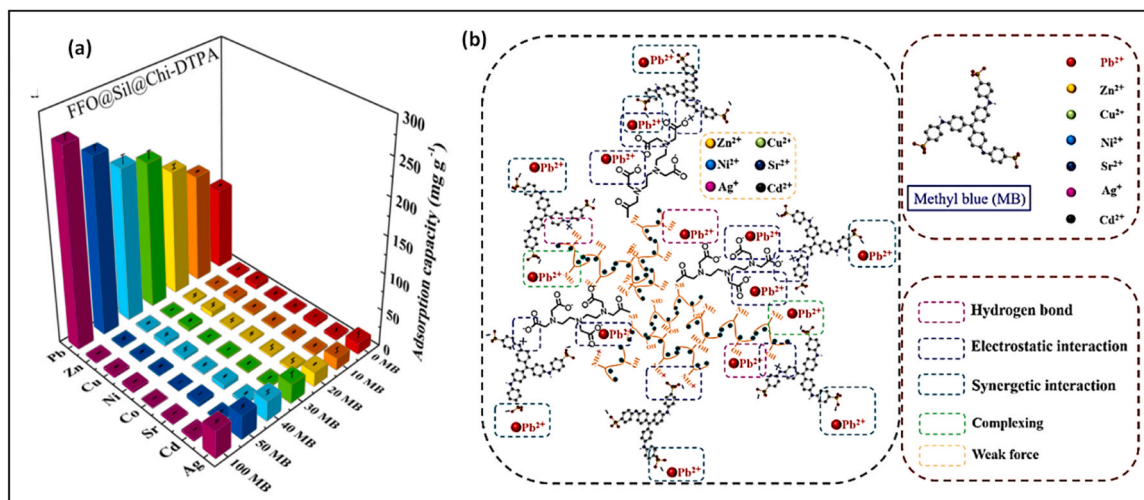
In this respect, the application of CS magnetic materials was studied in adsorption for a variety of metal ions such as Zn<sup>2+</sup>, Pd<sup>2+</sup>, Cd<sup>2+</sup>, Hg<sup>2+</sup>, Mn<sup>2+</sup>, and Fe<sup>3+</sup>. Fatemeh et al. developed a new magnetic nanocomposite based on CS/Al<sub>2</sub>O<sub>3</sub>/Fe<sub>3</sub>O<sub>4</sub>, which is known as EDTA-M-Cs for extracting only zinc (Zn(II)) and cadmium (II) ions from impure water [259]. CS NCs demonstrated appreciable efficiency for adsorption processes, including arsenate, the anthraquinone dye Acid Green 27, Pb(II) ions, Cr(VI), and Cd(II) ions [186].

Under the supervision of Manar El-Sayed Abdel-Raouf, four sorbents for lead and copper removal from simulated solutions were prepared by modifying CS. In the study, the sorbents were tested at their best points of pH 5 using copper and pH 8 for removing lead. Based on the test results, CS/GA/MBA exhibited the highest adsorption values of 95.7 mg/g for copper and 99.15 mg/g for lead. In supporting the pseudo 2nd order kinetics and Langmuir isotherms, it demonstrated chemisorption and a homogeneous spread of metal ions. The modification of topography and roughness of the sample was proved through AFM

analysis to confirm the uptake process of metals and that green sorbents favor both of them [260].

The study by Huang et al. introduces an advanced method for effectively eliminating heavy metals and dyes from complex industrial wastewater. The researchers developed a new magnetic adsorbent, known as diethylene-triamine-penta-acetic acid (DTPA)-modified CS-coated magnetic silica NPs (FFO@Sil@Chi-DTPA), which demonstrates excellent selectivity in adsorbing Pb(II) from multi-metal solutions through anion-synergism. A significant finding of the study is the substantial increase in Pb(II) adsorption capacity by FFO@Sil@Chi-DTPA, particularly as the concentration of methylene blue (MB) in the solution rises. This interaction between MB and Pb(II) highlights the adsorbent's potential for effectively removing heavy metals and dyes, offering an efficient solution for industrial wastewater treatment. Fig. 15a illustrates the strong preference of the FFO@Sil@Chi-DTPA adsorbent for binding to Pb(II) in a mixed system of multiple metal ions and MB. Notably, the adsorption capacity of Pb(II) significantly rises from 111.71 mg/g to 268.01 mg/g as the concentration of MB increases from 0 to 100 mg/L. This indicates that the anionic properties of MB play a critical role in enhancing the selective adsorption of Pb(II) in the complex solution, potentially by facilitating the creation of additional binding sites for Pb(II). The suggested synergistic adsorption mechanism works as follows Pb(II) ions are initially adsorbed onto the adsorbent surface through a combination of interactions, including co-ordination with amino groups, hydrogen bonding with hydroxyl groups, and electrostatic attraction with carboxyl groups. The adsorbed MB molecules further enhance Pb(II) adsorption by creating more selective binding sites, primarily due to their sulfonic acid groups. Additionally, the electrostatic attraction of MB can be reinforced by the Pb(II) ions already adsorbed on the surface, acting as cationic bridges. As depicted in Fig. 15b this results in a comprehensive process for the simultaneous removal of MB and Pb(II) [261].

Cellulose-based magnetic beads are environmentally friendly and



**Fig. 15.** (a) Enhanced Binding Capacity of FFO@Sil@Chi-DTPA for Pb(II) with Increasing MB Concentration, (b) Collaborative Elimination of Methylene Blue and Pb (II) via Electrostatic Attraction and Cationic Bridging. [Reprinted (adapted) under kind courtesy from {J. Hazardous Materials, 422, 15 January 2022, 126856}. Copyright {2021} Elsevier].

have been fabricated with the best techniques using extrusion, as presented by research from Xiaogang Luo. These beads were enhanced with carboxyl-functionalized  $\text{Fe}_3\text{O}_4$  NPs and nitric acid-treated activated carbon and evidenced sensitivity and efficiency in the adsorption of  $\text{Cu}_2\text{O}$ ,  $\text{Pb}_2\text{O}$ , and  $\text{Zn}_2\text{O}$ . They possess fast adsorption rates and high

capacity with good reusability, making the process cheaper and scalable for the removal of heavy metals from water. It is mostly because of electrostatic attraction that the adsorption is spontaneous and endothermic [262]. Table 5 depicts CS-based NCs in use for the removal of toxic metals from wastewater [263–296]. Hematite/CS NCs show an

**Table 5**

CS-based NCs in use for the treatment of toxic metal ions from impure water.

| Type of NCs                                              | Heavy metal                                                     | pH                                   | Adsorption capacity (mg/g)                     | Kinetics | Reference |
|----------------------------------------------------------|-----------------------------------------------------------------|--------------------------------------|------------------------------------------------|----------|-----------|
| Hematite/CS                                              | Pb (II), Cu (II), and Cd (II)                                   | 6.4 and 6.5                          | 129.8, 63.2 and 69.9                           | -        | [263]     |
| PVP/CS-SB/ $\text{Fe}_2\text{O}_3$                       | Pb (II) and Hg (II)                                             | 5                                    | 120 and 102.5                                  | -        | [264]     |
| $\text{TiO}_2$ /CSC.Algm. composite sponge               | As (V)                                                          | 5                                    | 202.27                                         | PSO      | [265]     |
| EDTA/CS                                                  | Methyl orange dye                                               | 5                                    | 55.87                                          | PSO      | [266]     |
| Graphene oxide/CS/Ferrite                                | Cr (VI)                                                         | 2                                    | 270.27                                         | PSO      | [267]     |
| Graphene oxide/ $\text{ZnO}$ /CS                         | Pb (II) and Cr (VI)                                             | 5.2                                  | 110.88, 84.5                                   | PSO      | [268]     |
| CS- $\text{MnO}_2$                                       | Cr (VI)                                                         | 2                                    | 61.56                                          |          | [269]     |
| $\text{MnO}_2$ /CS/ $\text{Fe}_3\text{O}_4$              | Zn (II) and Pb (II)                                             | 4–6                                  | 136 and 310.4                                  |          | [270]     |
| ChMGOP- $\text{Co}_3\text{O}_4$                          | As (V)                                                          | 7                                    | 90.9                                           | PSO      | [271]     |
| EDTA-MCS/GO                                              | $\text{Pb}^{2+}$ , $\text{Cu}^{2+}$ , and $\text{As}^{3+}$      | 5, 5.5, and 8                        | 206.52, 207.26, and 42.75                      |          | [272]     |
| $\text{ZnO}$ /CS Core-shell                              | Cu (II), Cd (II), and Pb (II)                                   | Cu (II) 4, Pb (II) 6 and Cd (II) 6.5 | Pb (II) 476.1, Cd (II) 135.1 and Cu (II) 117.6 |          | [273]     |
| Zinc-CS nano-biocomposite                                | Cr (VI)                                                         | 3                                    | 69                                             |          | [274]     |
| CS- $\text{TiO}_2$                                       | Cr(VI)                                                          | 2                                    | 488                                            |          | [275]     |
| $\text{CsC@SiO}_2\text{-@TiO}_2$                         | Cr(VI)                                                          | 3                                    | 182.43                                         |          | [276]     |
| (D- $\text{TiO}_2$ -NC) with CS                          | Cd and Pb                                                       | 6                                    | 147 (Cd) and 145.5 (Pb)                        | PSO      | [277]     |
| CS/ $\text{TiO}_2$ /Ag NPs                               | Cu (II)                                                         | 5                                    | 7.71                                           | PSO      | [278]     |
| Alginate/CS                                              | La (III), Ce (III), Pr (III), Nd (III), Sm (III), Ho (III) ions | 7–8                                  | 128.40–168.44                                  | PSO      | [279]     |
| MCS/CNT                                                  | Pb (II)                                                         | 5                                    | 101                                            | PSO      | [280]     |
| CS/CNT/PDA                                               | Cu (II)                                                         | 7                                    | 112                                            | PSO      | [281]     |
| CS/CMSt                                                  | Cu (II)                                                         | 5                                    | 96                                             | PSO      | [282]     |
| Iron CS Microspheres                                     | As(V)                                                           | 6–9                                  | 120.7                                          | PSO      | [283]     |
| Magnetic CS Coated GO                                    | As (III)                                                        | 7.3                                  | 45.0                                           | PSO      | [284]     |
| CS Coated magnetite                                      | As (III)                                                        | 9                                    | 10.5                                           | PSO      | [285]     |
| Fe (III)-CS                                              | As (III)                                                        | 5.5                                  | 19.7                                           | PSO      | [286]     |
| Amidoxime-Functionalized CS                              | Cu (II), Ni (II)                                                |                                      | 190.7, 128.9                                   | PSO      | [287]     |
| CNTs-CHO-CS composite                                    | Cu (II)                                                         |                                      | 115.84                                         | PSO      | [288]     |
| Magnetic ZIF-67 MOF@Am-CS Beads                          | Cr (VI)                                                         |                                      | 119.1                                          | PSO      | [289]     |
| CS- $\text{MnO}_2$ NCs                                   | Cr (VI)                                                         | 2                                    | 61.56                                          |          | [290]     |
| CS/g- $\text{C}_3\text{N}_4$ / $\text{TiO}_2$ nanofibers | Cr (VI)                                                         | 2                                    | 68.9                                           |          | [291]     |
| Citrate-crosslinked Zn-MOF/CS composite                  | Cr (VI)                                                         | 5                                    | 225                                            | PSO      | [292]     |
| Alginate/Melamine/CS aerogel                             | Metal ions & Radionuclides                                      | 5–9                                  | Variable                                       | PSO      | [293]     |
| CS-pectin gel beads                                      | Cu (II), Cd (II), Hg (II) and Pb (II)                           | 4–9                                  | 169.4, 177.6, 208.5 and 266.5                  | PSO      | [294]     |
| CS-PVA nanofibers                                        | Pb (II) and Cd (II)                                             | 6.8                                  | 266.0 and 148.0                                | PSO      | [295]     |
| CS-VMT composite                                         | Cd (II) and Pb (II)                                             | 4                                    | 58.48 $\text{mg g}^{-1}$ and 166.67            | PSO      | [296]     |

**Note:** PSO stands for pseudo 2nd order, and PFO stands for pseudo 1st order.

adsorption capacity of 129.8 mg/g for Pb (II) at 6 pH, but decrease to 63.2 at 4 pH [263]. Combining chelating agents like EDTA with CS as a low-cost adsorbent for the adsorption of Methyl orange dye from the wastewater increases [266]. The adsorption capacity of CS-based composite increases by using a ZnO/CS core-shell structure that is 476.1 mg/g for Cu(II) at 4 pH and 135.1 for Cd(II) ions at 6 pH [273]. A maximum adsorption capacity was observed for Alginate/Melamine/CS aerogel was 1331.6 mg/g for Pb (II) ions at 5.5 pH [293].

## 5. CS-based NCs in other pollutant removal

Most waterborne diseases are caused by the prevalence of microorganisms like bacteria and fungi in drinking water and other sources, including the removal of heavy metals, pollutants, wastes, and suspended colloidal particles, etc. The necessity to eradicate microorganisms is fundamental. Harmful microbes like bacteria, viruses, and parasites can bring about water pollution. Such harmful microbes are bacteria, in particular, *E. coli*, *Shigella*, and *V. cholerae*. Viruses, for instance, hepatitis A, poliovirus, and Rotavirus. Parasites; among them, *E. histolytica*, Hookworm [297].

Two broad categories of waterborne contaminants are ionic and organic [298]. The two types of ionic contaminants present in water are ions of heavy metals, including lead, copper, selenium, arsenic, mercury, cadmium, chromium, cobalt, etc. Non-metallic ions, including phosphate, nitrate, sulfide, or fluoride [299]. This has been proven to be very efficient in the utilization of CS and its derivatives as adsorbents for the removal of different types of contaminants, including phenols, fluorides, and pigments [300].

Adriana Herrera et al. have replaced CS beads by doping iron oxide (FeO) and titanium dioxide (TiO<sub>2</sub>) NPs through ionic cross-linking. The beading turns porous and increases the bead's surface area. The adsorption studies have been very effective in naphthalene removal, mainly through chemisorption, and the Ch-FeO/TiO<sub>2</sub> beads showed greater capacity. The FeO NPs provided superparamagnetic properties for easy recovery. Although problems of desorption still subsist, the beads were stable with significant interactions with the pollutant, thus an application for photocatalytic remediation with TiO<sub>2</sub> [301]. Table 6 depicts CS-based NCs in the removal of other pollutants [302–311]. A maximum adsorption capacity of 1130 mg/g was shown by CS/GO NCs for Tetracycline dye at 9.0 pH [302]. On methylation of surface groups of CS in the presence of Graphene oxide, its adsorption capacity decreases to 102 mg/g for Rifampicin [303] and on replacing graphene oxide with reduced graphene oxide, its adsorption capacity decreases to 30 mg/g for Cefixime antibiotics [305]. On replacing graphene oxide with Bentonite, CS-based NCs show an adsorption capacity of 465.2 mg/g for Bisphenol-A [309].

Specifically, we emphasized that NCs-based adsorbents—such as those derived from CS, CPs, and MO NPs offer several advantages over conventional adsorbents (e.g., activated carbon, silica, clays) in industrial water treatment settings [263–266]. NC-based adsorbents show

enhanced adsorption efficiency. The incorporation of MO and CPs significantly increases the surface area, active sites, and functional group diversity, leading to higher adsorption capacities and faster kinetics [269–273]. NCs-based adsorbents also show tailored selectivity. NCs can be engineered to selectively target specific pollutants (e.g., heavy metals, dyes, pharmaceuticals), which is particularly beneficial in complex industrial effluents [274–278]. NCs-based adsorbents also show multifunctionality. Many NCs not only adsorb pollutants but also possess photocatalytic and antimicrobial properties, enabling multifunctional treatment in a single-step process [279–282]. NCs-based adsorbents also show properties of regeneration and reusability. Properly designed NCs often exhibit good regeneration potential, which enhances cost-effectiveness and reduces secondary waste generation in industrial processes [283]. NC-based adsorbents also show mechanical and chemical stability. Compared to some conventional materials, these NC-based adsorbents offer superior chemical stability under a wide range of pH and temperature conditions commonly encountered in industrial wastewater streams [305–311,315,316,317].

We acknowledged that while these features make NCs promising for industrial-scale use, challenges such as large-scale synthesis, recovery, potential leaching of nanoparticles, and cost-effectiveness must still be addressed.

The potential environmental impact and toxicity of NCs in long-term applications may have a significant effect on the environment. Their long-term environmental fate and potential ecotoxicological effects remain areas of concern. In certain cases, embedded MO NPs (e.g., TiO<sub>2</sub>, ZnO, CuO) may leach into treated water or the environment, potentially causing toxicity to aquatic organisms and bioaccumulation in the food chain. While CS is biodegradable, many CPs and MO are not, which raises concerns about the persistence of composite residues in natural ecosystems after prolonged use. Long-term use of nanocomposites under varying environmental conditions (e.g., pH, UV exposure) may lead to the formation of transformation products with unknown or toxic behavior. There is still limited consensus on standardized methods for assessing the life-cycle environmental impact and toxicity of nanocomposites, particularly under real-world conditions.

## 6. Conclusions

Addressing water pollution is a critical challenge for both environmental sustainability and public health. The growing problem of dye contamination in water necessitates the development of effective and economically viable treatment technologies. The low-cost adsorbents have shown great promise in enhancing wastewater treatment processes, offering a practical solution for pollution control in resource-limited settings. While advancements in treatment technologies such as adsorption, advanced oxidation, and biological methods provide promising solutions, their implementation must consider both technical feasibility and economic viability. Regulatory frameworks and continuous research are essential to ensure the safe use of emerging

**Table 6**

The CS-based NCs for the treatment of other pollutants.

| Nanocomposites                        | Other pollutants                                       | pH       | Adsorption capacity (mg/g)                      | Kinetics         | Reference |
|---------------------------------------|--------------------------------------------------------|----------|-------------------------------------------------|------------------|-----------|
| CS/GO                                 | Tetracycline                                           | 9.0      | 1130                                            | PFO              | [302]     |
| MCS/GO                                | Rifampicin                                             | 5.0      | 102                                             | PSO              | [303]     |
| CS/BC                                 | Ciprofloxacin, Enrofloxacin                            | 7.0, 7.0 | 106, 100                                        | PSO              | [304]     |
| MCS/rGO                               | Cefixime                                               | 6.4      | 30                                              | PSO              | [305]     |
| CS/GO                                 | Ciprofloxacin, Ofloxacin                               | 5        | 5.3, 8.3                                        | PSO              | [306]     |
| CS/MCM-48                             | Phenol                                                 | 8        | 149.25                                          | PSO              | [307]     |
| Fe/CLCh-EDA/<br>β-CD                  | Phenol (PHE), p-nitrophenol (PNP), and p-cresol (PCS). | 6        | PHE of 243.08, PNP of 274.24, and PCS of 298.35 |                  | [308]     |
| Bentonite-CS                          | Bisphenol A (BPA) and pentachlorophenol (PCP)          |          | 465.21 for BPA and 481.91 for PCP               | Pseudo 2nd order | [309]     |
| Cu-BTC@C-PANi                         | 4-chlorophenol, and 4-nitrophenol                      |          | 277 and 324                                     | Pseudo 2nd order | [310]     |
| CS/ZnO-Fe <sub>3</sub> O <sub>4</sub> | Cephalexin (CFX)                                       |          | 81.38                                           | Pseudo 2nd order | [311]     |

**Note:** PSO stands for pseudo 2nd order, and PFO stands for pseudo 1st order.

**Table 7**

Comparative study of CS, CPs, and MO NPs for the removal of environmental pollutants from waste water.

| Material Type                                                               | Strengths                                                                                                                                                         | Weaknesses                                                                                         | Best Applications                                                                                 |
|-----------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------|
| CS                                                                          | Biodegradable and non-toxic- Excellent adsorption capacity for heavy metals and dyes, Abundant functional groups (–OH, –NH <sub>2</sub> ), Renewable and low-cost | Poor mechanical strength- Limited thermal stability, Solubility issues in neutral/basic pH         | Adsorption of heavy metals, dyes, and nutrients, Carrier material in nanocomposites               |
| CPs (e.g., Polyaniline, Polypyrrole)                                        | High electrical conductivity, Redox-active sites for pollutant degradation- Tunable structure, Photocatalytic and electrochemical synergy with other materials    | Prone to degradation under prolonged use, Limited adsorption alone, and often expensive synthesis. | Electrochemical and photocatalytic degradation, Sensor development, and Adsorption in composites. |
| MO NPs (e.g., TiO <sub>2</sub> , ZnO, Fe <sub>3</sub> O <sub>4</sub> , CuO) | High surface area- Strong photocatalytic properties, Magnetic separability (e.g., Fe <sub>3</sub> O <sub>4</sub> ), Effective in redox reactions.                 | Aggregation in aqueous media, Potential cytotoxicity, High production cost in pure form.           | Photocatalytic degradation [312–314], Magnetic separation, Combined adsorption, photocatalysis.   |

technologies, such as NMs, while mitigating any potential risks to human health and the environment. PANi-based NCs, that is PNHM/MnO<sub>2</sub>/Fe<sub>3</sub>O<sub>4</sub> NCs, show a high adsorption capacity of 1142.1 mg/g for adsorbing MG dye at 6.5 pH. Binary metal oxide NPs, that is ZnO and Al<sub>2</sub>O<sub>3</sub> with PANi CP, show an adsorption capacity of 442.8 mg/g for CR dye at 5.5 pH. On replacing PANi with PPy, PPM/Fe<sub>3</sub>O<sub>4</sub> NCs show an adsorption capacity of 922.5 mg/g for Cr(IV) metal ion at pH 2.0. PPy/Fe<sub>3</sub>O<sub>4</sub>/Seaweed-based composite shows an adsorption capacity of 666.6 mg/g for MB dye at pH 10. On replacing CP with CS-based NCs, the adsorption capacity changes significantly. A maximum adsorption capacity is shown by CS based NCs, which is 620.5 mg/g for RBBR dye by the CS/Benzil/ZnO/Fe<sub>3</sub>O<sub>4</sub> NCs. But for metal ions, CS based NCs, that is Alginate/Melamine/CS aerogel, show a maximum adsorption capacity of 1331.6 mg/g for Pb(II) ions. The adsorption capacity of a particular NCs depends on the nature of the MO NPs, their combination, and the pH of the water. Even for the same NCs, the adsorption capacity is different for organic dye and heavy metal ions present in impure water. Overall, the integration of innovative CPs, CS, MO NP-based NCs provides a low-cost and sustainable solution, such as adsorbents and nanotechnology, which is essential for the effective management of water pollution.

### Future perspectives

Future efforts should focus on further research, regulatory oversight, and the scaling up of these technologies to address the global challenge of water contamination and to safeguard water resources for future generations. The economic aspects of the actual experimental setup should be estimated for the economic viability of this technique.

### CRediT authorship contribution statement

**Aarti Tundwal:** Writing–original draft, Formal analysis. **Harish Kumar:** Supervision, Writing - review & editing, Conceptualization. **Prity Yadav:** Validation, Formal analysis. **Versha Yadav:** Validation, Data curation. **Bindu Mangla:** Validation, Formal Analysis.

### Declaration of Generative AI and AI-assisted technologies in the writing process

We declare that help of any AI tool was not taken in writing this review article.

### Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Harish Kumar reports that administrative support was provided by the Central University of Haryana Department of Chemistry. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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