

Heavy Metal Ion Adsorbent in Aqueous Solution: A Review on Chitosan and Chitosan Composites

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ARTICLE INFO

Article history:

Received 15 January 2024

Received in revised form 20 February 2024

Accepted 29 March 2024

Available online 30 April 2024

Keywords:

Environmental pollution; heavy metal ions; adsorption; chitosan; modification

ABSTRACT

One prominent contributor to environmental degradation is the presence of heavy metal ions, which are commonly introduced into ecosystems through diverse channels, including mining operations, paint manufacturing, battery production, and agricultural practises. The unprocessed waste from these industries typically undergoes direct discharge into rivers and seas, thereby adversely influencing the potable water supply and marine ecosystem. This results in the accumulation of heavy metals within living organisms and subsequent environmental degradation that poses a significant risk to human health, as their prolonged consumption can harm various organs and the nervous system. Therefore, the United States Environmental Protection Agency (US-EPA) and the World Health Organisation (WHO) have established stringent regulations regarding the allowable levels of heavy metal intake, particularly concerning potable water consumption. The imperative lies in surmounting this challenge by discovering the optimal technique for extracting heavy metals. This paper examines the primary methods of eliminating heavy metal ions using chitosan-based materials. Specifically, the methods under scrutiny include electrocoagulation (E.C.), bioremediation, ion-exchange, membrane filtration (purification), and adsorption. Adsorption, as a highly efficient method, presents itself as a viable strategy for the remediation of dyes in textile wastewater due to its advantageous characteristics. Notably, this approach obviates the need for extensive spatial requirements and exhibits a commendably swift treatment duration. Chitosan exhibits promising adsorptive characteristics owing to its advantageous physical attributes, including crystalline structure, porous nature, and particle dimensions, all of which synergistically enhance the adsorption capacity of chitosan. Research findings indicate that the adsorption characteristics of chitosan are subject to variation contingent upon the distinct heavy metal ions under consideration and the specific experimental parameters employed. Hence, optimising these parameters becomes imperative to attain enhanced chitosan adsorption capabilities for heavy metal ion removal.

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<https://doi.org/10.37934/armne.18.1.78102>

1. Introduction

The accelerated global expansion of industrial activities exacerbates environmental contamination, especially in marine and soil. These contaminants encompass a wide array of substances, such as minerals, gases, water, soil organic matter, and living organisms. The term "waste" etymology can be traced back to its Latin origin, precisely the word "vastus." This word conveys the concept of leaving behind, abandoning, or failing to construct. The conventional interpretation of waste pertains to materials deemed undesirable or lacking in value [1]. Therefore, wastewater can be defined as the aqueous substance that has undergone prior utilisation for a designated function and is subsequently released into the environment. The wastewater matrix encompasses a diverse array of constituents, including but not limited to human excrement, residual food matter, lipid compounds, and a wide range of chemical substances.

Wastewater is also known to contain heavy metal ions as a constituent. Heavy metal ions, a form of contamination, are derived from diverse sources such as mining activities, paint manufacturing, battery production, and agricultural practises. The unmitigated discharge of waste from these industrial sectors typically finds its way into rivers and oceans, thereby exerting adverse impacts on potable water supplies and ecosystems, particularly those of the marine variety. In this context, the presence of heavy metals within living organisms becomes a concern as they tend to accumulate and subsequently deteriorate the surrounding environment. The presence of various heavy metals, such as chromium (VI), manganese (II), lead (II), arsenic (V), and cadmium (II), in the environment poses significant concerns due to their carcinogenic and toxic properties.

Another contributor to heavy metal contamination in waterways is electronic waste or e-waste. The short lifespan of electronic products has exacerbated the problem. The electronic waste such as wires, cables, printed circuit boards, plugs, and chips, contains toxic heavy metals like Pd, Cu, Sn, Ag, and Zn. The common practice of open-air incinerating electronic waste rather than recycling it has resulted in significant environmental contamination. Furthermore, the waste is frequently dumped directly into the soil and watercourses. E-waste accumulated 53.6 million metric tonnes worldwide in 2019, according to a comprehensive survey. Asia was the biggest source of electronic waste, contributing 24.9 million metric tonnes, while Europe contributed 12.0 million metric tonnes [2].

Heavy metals are commonly denoted as naturally-occurring metallic elements possessing densities exceeding 3 g/cm^3 [3]. The study conducted by Chowdhury *et al.*, [4] shed light on the notable health hazards linked to heavy metal pollutants in developing countries. Based on scientific research, it has been established that arsenic (As) plays a significant role in drinking water contamination, posing a considerable environmental concern. Furthermore, it is important to acknowledge that exposure to heavy metals is an inherent aspect of our surroundings, making it nearly impossible to evade such encounters completely. This issue has significant implications for both terrestrial and aquatic ecosystems, encompassing the well-being of human populations as well as the biodiversity and ecological balance of marine organisms. Biological Oxygen Demand (BOD) and Chemical Oxygen Demand (COD) have been recognised as the principal parameters employed in assessing the extent of water pollution. Regular monitoring of these two parameters is of utmost importance, as they serve as reliable indicators of the contaminants' levels in wastewater [5]. Contaminants, in their various forms, present a significant concern for human well-being due to their inherent capacity to induce detrimental effects on vital organs and the intricate nervous system when persistently ingested over a prolonged duration.

Therefore, the United States Environmental Protection Agency (US-EPA) and the World Health Organisation (WHO) have established regulatory thresholds for acceptable levels of heavy metal consumption, particularly concerning drinking water. The permissible limit refers to the utmost

admissible concentration of a contaminant present in potable water, and these regulations are legally enforceable. Secondary drinking water standards, in contrast, pertain to non-mandatory guidelines established by Water Regulations to assess contaminants present in drinking water that have the potential to induce cosmetic manifestations (such as alterations in skin or tooth pigmentation) or aesthetic manifestations (such as variations in taste, odour, or hue). As an environmental scientist, it is crucial to note that the established threshold for cadmium ion concentration in water is 0.003 mg/L. This limit has been set to prevent potential health risks associated with cadmium exposure, such as renal toxicity, hypertension, and kidney damage [6]. Prolonged exposure to elevated levels of copper (Cu) in the range of 2.5 mg/L or higher has been observed to induce adverse effects on renal function.

Similarly, the consumption of arsenic has been associated with significant health hazards, such as alterations in pigmentation, gastrointestinal discomfort, and an increased risk of developing cancer [7]. The accumulation of heavy metals has been observed to significantly impact the functioning of hormones and enzymes [8]. This phenomenon disrupts crucial metabolic processes, leading to an imbalance in antioxidation [9]. Table 1 presents additional instances of heavy metal ions and their detrimental impacts, thereby shedding light on the potential environmental repercussions of these substances.

Table 1
 The permissible limit of selected metal ions in drinking water

Metals	Main source	Health effects	Permissible limit (mg/L)	References
Copper (Cu)	Corrosion of household plumbing materials and industrial manufacture	Gastrointestinal distress, and in the long run, experience liver or kidney damage	2.5	[10]
Cadmium (Cd)	Paints, pigments, electroplated parts, synthetic rubber, batteries, plastics and phosphate fertilisers Corrosion of galvanised pipes; erosion of natural deposits; discharge from metal refineries; runoff from waste batteries and paints	Renal toxicity, hypertension, fatigue and kidney damage	0.003	[11]
Mercury (Hg)	Erosion of natural deposits; discharge from refineries and factories; runoff from landfills and croplands	Kidney damage	0.001	[12]
Zinc (Zn)	Tap water, rocks and meat	Stomach cramps, skin irritations, vomiting, nausea, anaemia, trouble in pancreas, generate arteriosclerosis	5.00 (Secondary Drinking water standard)	[13]
Lead (Pb)	Corrosion and chipping of the pipes	Alterations in several biological processes, including cell adhesion, intra- and inter-cellular signalling, protein folding, maturation, apoptosis, ionic transport, enzyme regulation, and neurotransmitter release	0.05	[14,15]

Hexavalent chromium (Cr (VI))	Industrial process: steel, catalysis, cement, textile, fungicide, electroplating and dyeing	Teratogenic (causing birth defects), mutagenic (causing genetic mutations), and carcinogenic (causing cancer) effects	0.1	[16,17]
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Thus, treating the wastewater to remove the contaminants before releasing it to the environment is essential. Various techniques can be applied to remove the heavy metals including membrane filtration, bioremediation, ion-exchange, osmosis, electrochemical and adsorption. The choices of technique greatly depend on the nature of chemicals and types of pollutants. Adsorption is the most versatile process as it is simple to operate and can remove both organic and inorganic materials [18]. Adsorbent based on chitosan and its derivatives attracts many researchers due to their ability to detect and remove heavy metals and dyes even at low concentrations [19].








This article provides an overview of chitosan, its derivatives, and its composite as an aqueous heavy metal adsorbent. Other removal methods will also be discussed, including electrocoagulation (E.C.), bioremediation, ion exchange, and membrane filtration.

2. Chitin and Chitosan

Chitosan is a derivative polymer of chitin. Chitin is the second most abundant natural polymer source widely used in daily life after cellulose. It is a low-cost material and easy to be manipulated in the preparation of chemical-based products. Chitin can be found in crustaceans' shells, insect and mollusc exoskeletons, and fungi's cell walls [20]. Whilst fungi produce six to twelve per cent of the soil's chitin biomass, industrial chitin's primary source is waste from marine food production, specifically crustacean shells such as shrimp and crab. The estimated annual turnover is between 10¹⁰ and 10¹¹ tonnes, making chitin one of the most common biopolymers. Examples of sources of chitin are shown in Table 2.

Table 2
 Sources of chitin for chitosan extraction

Source	Sites where chitin is present	Image	References
Insect	The exoskeleton, cuticle, the membrane between the segments	Beetle	[21-24]
			
		Cockroach	[25-28]
		Cricket	[29-32]
			

		Grasshopper	[33,34]
			
Crustaceans	The membrane between the segments, shell, exoskeleton	Crab	[35-42]
			
		Prawn	[43-49]
			
		Lobster	[50-55]
			
Mollusc	The shells, teeth, stomach plates, and feathers	Squid pen	[56-62]
			
		Oyster	[63-68]
			
		Bamboo shell / razor clam	[69]
			
Fungi	The cell wall, mycelium, stem, and spores	<i>Agaricus bisporus</i> , <i>Auricularia auriculajudae</i> , <i>Aspergillus niger</i> , <i>Mucor rouxii</i> , <i>Rhizopus oryzae</i> , <i>Phycomyces blakesleenaus</i> , <i>Absidia blakesleenaus</i> , <i>Absidia glauca</i> , <i>Absidia coerulea</i> , <i>Lentinus edodes</i>	[70-82]

Chitin can be converted to chitosan in three steps: demineralisation, deproteinization, and deacetylation [83]. In order to remove the pigment, numerous inorganic and organic reagents, including hydrogen peroxide, sodium hypochlorite, and acetone, can be used as an optional decolourisation step, primarily with astaxanthin and β -carotene [84].

Chitosan has unique characteristics and is a pseudo-natural cationic polymer soluble in aqueous solutions that gives chitosan a relatively high sorption capacity for several metal ions. It has been reported that chitosan can be used in the form of solution, gel, film, or fibre [85]. Biomedical and pharmaceutical sectors exploit the potential of chitosan due to its biodegradability, biocompatibility, antimicrobial, non-toxicity, and anti-tumour properties [86]. Likewise, the Food and Drug Administration (FDA) acknowledges chitosan as Generally Recognised as Safe (GRAS); hence, it highly influences the food and beverage industry. Chitosan has been used in food preservation to preserve their nutritional value and as antimicrobial properties. It is also widely used as edible coating, food packaging, emulsion and encapsulation materials and acts as an agent for enzyme immobilisation

and water purification [87]. Apart from that, due to chitosan's unique characteristics, it can be utilised in many other applications, such as in agriculture [88], cosmetics [89], and clinical applications [90]. Furthermore, the inherent antibacterial property of chitosan makes it a commonly used component in antimicrobial hydrogels [91].

Chitosan, a linear polysaccharide consisting of glucosamine and N-acetyl glucosamine units connected by β (1-4) glycoside bonds [92,93], possesses hydrophilic properties that enable it to form complexes with metal ions. This quality makes chitosan suitable as an adsorbent in aqueous solutions to capture metal ions. The structure of chitosan, which includes hydroxyl (O.H.) and amino (NH_2) groups, facilitates its ability to coordinate and bond with metal ions [94] selectively. Figure 1 shows the chemical structure of chitin and chitosan.

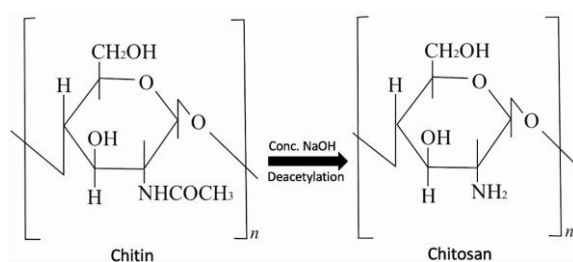


Fig. 1. Chemical structure of chitin and chitosan

Chitosan is a flexible substance that has a wide range of uses. It can be employed in numerous products, such as biocatalysts, antioxidants, antibacterial, and anticancer agents. It is also used in wound healing dressings and as a food preservative. Chitosan is widely recognised as a highly abundant and versatile biopolymer that effectively eliminates pollutants from various water sources [95].

There are three grades of chitosan based on degrees of deacetylation (DD) as proposed by Grégorio Crini [96] technical grade, pure grade and ultra-pure grade. High DD (ultra-pure grade, 90-95%) and high molecular weight (MW) (3×10^5 Da) is suitable for medical applications. The food industry requires chitosan with a DD range of 85-95% and high MW, while wastewater treatments' chitosan is in technical grade, with DD ranging from 70-85% and MW less than (3×10^5 Da). Chitosan with a lower MW has better water solubility, relatively low viscosity and density, and greater cell penetration capacity. Conversely, the higher the MW, the higher the viscosity, adsorption/chelation capacity, and conductivity.

Chitosan's DD and MW differ depending on the raw material used to extract chitin. Chitosan can be extracted from fungi, insects, and crustaceans. In general, insect-derived chitosan has higher purity and longer macromolecular chain lengths. Insects, unlike crustaceans, do not change seasonally. Purifying the insect, on the other hand, is difficult. Furthermore, the chitin content of whole insects is lower than that of crustaceans in general.

Chitosan-based composites, which are part of an environmentally friendly biopolymers class, have recently demonstrated promising capabilities in effectively adsorbing various pollutants, specifically pharmaceuticals like antibiotics, analgesics, antipyretics, and anti-inflammatories. These composites offer a diverse range of potential adsorption mechanisms due to the presence of adsorbent functional groups that have been incorporated into the chitosan material [97].

3. Heavy Metal Ion Removal Methods

Heavy metal ions are removed through various techniques to decrease or eliminate their concentration from diverse sources, including water, wastewater, soil, and industrial effluents.

Determining a method for eliminating heavy metal ions is contingent upon various factors, including the nature and amount of the pollutants, the magnitude of the water or wastewater to be processed, financial implications, and compliance with legal standards. Frequently, a conglomeration of techniques may be utilised to attain effective and all-encompassing elimination of heavy metal ions.

Conventional techniques for eliminating heavy metals, such as ion exchange, chemical precipitation, coagulation, membrane separation, electrocoagulation, and reverse osmosis, have not been extensively employed due to their low practicality and high expenses. However, every method contributes substantially to the water purification process.

3.1 Electrocoagulation (E.C.)

Coagulation is a process in which pollutants, suspended particles, collide with counter particles and agglomerate to produce an insoluble agglomeration complex [98]. Electrocoagulation (E.C.) refers to the coagulant produced by the electrochemical process, which was first proposed by Vik *et al.*, [99].

In the electrocoagulation (E.C.) technique, coagulation and sedimentation are accomplished via conductors in the reactor as opposed to the addition of chemical coagulants [100,101]. The process employs iron, aluminium, mild steel, or stainless-steel electrodes. When these electrodes are subjected to an electric current, the scarification of the anode generates coagulant formation in the system, resulting in coagulation and flocculation. These ions act as magnets, attracting oppositely charged particles into large masses known as agglomerates [102].

An electrolytic cell with one anode and one cathode is usually used in electrocoagulation. The anode, also referred to as the sacrificial electrode, is made of conductive metallic plates, whereas the cathode can be made of the same or a different material [103]. As a result, effective pollutant removal is achieved [104,105].

3.2 Bioremediation

In order to reduce the rate of heavy metal accumulation, biological remediation (bioremediation) has been developed. It is a cost-effective, adaptable, and efficient method that was initially used to remove petroleum pollution [106]. However, the treatment period for this innovative approach is long and complicated.

In bioremediation, oxidative enzymes are useful for increasing clearance rates. Although these enzymes have exceptional applications in removing pollutants, their poor stability [107] limits their use. Therefore, it may be ineffective with high concentrations of microorganism-harming pollutants. The combination of nanomaterials and bioremediation has an excellent potential for effectiveness and sustainability. The large surface area, transportability, and sequestration properties of nanoparticles make them more cost-effective and expedient than conventional remediation techniques [108].

There are limitations to bioremediation techniques, including soil disturbance, high costs, difficult operation, and secondary contamination [109]. Despite this, researchers have developed bioremediation due to its advantageous properties, which include minimal environmental disturbance, low cost, absence of secondary pollution, in-situ remediation, and simple operation [110].

3.3 Ion-Exchange

Ion exchange is the process of exchanging ions between a substrate and its surrounding environment (wastewater). This process is viable owing to the fact that heavy metals possess varying charges and charge densities [111]. This technique usually uses resin and natural zeolites as the substrate. The resins comprise a cross-linked polymer matrix that allows ions to transfer appropriately. These resins are divided into two categories: synthetic and natural. Zeolites are inexpensive and have a good ability to selectively remove metals [112], whereas synthetic resins are more expensive.

Figure 2 shows the process of ion exchange for heavy metals removal. This method can efficiently remove heavy metal ions from water. However, because this method is pH-sensitive, it cannot be utilised on a large scale.

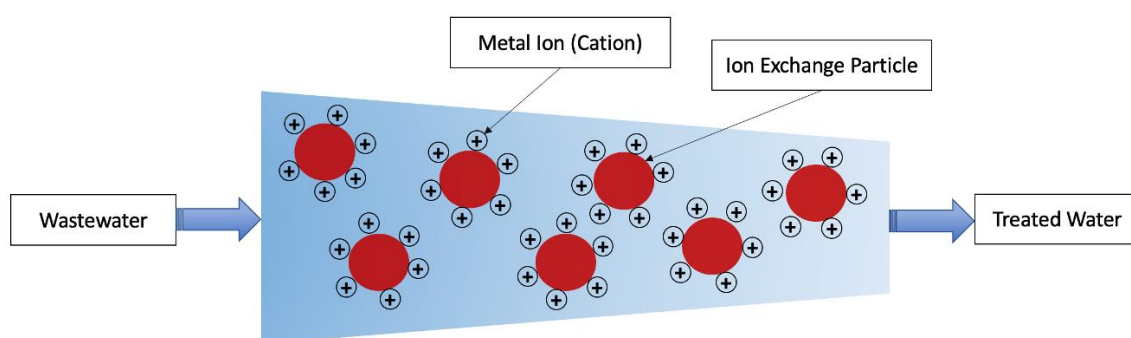


Fig. 2. Removal of heavy metal by ion exchange technique

3.4 Membrane Filtration (Purification)

Membrane filtration is gaining popularity for water purification as a result of its design versatility, scalability, high-quality filtered water, almost negligible footprint, and, in some instances, lower energy consumption. When a driving force is applied across a membrane, sieving and the Donnan effect can separate heavy metal ions based on the ion's size, membrane pore size, solution concentration, and the applied pressure. Studies show that microfiltration (M.F.), ultrafiltration [113], and nanofiltration [114] membranes, as well as reverse osmosis [115], can remove heavy metals ions and other contaminants such as bacteria, protozoa, and dye pigments from water.

In general, there are two types of membrane materials: ceramics and polymers. Ceramic membranes are better suited for industrial wastewater treatment than polymer membranes, which are commonly used in commercial applications. Ceramic membrane possesses excellent chemical resistance and hydrophobicity. However, it is fragile and expensive to construct and operate [116].

Polyethersulfone (PES) and polysulfone (PSf) are two common polymeric membrane materials [117]. PES possess high chemical resistance, thermal stability, and high mechanical strength. However, the hydrophobicity of the PES membrane makes it susceptible to fouling frequently, which can reduce production and shorten the membrane's lifespan [118]. Fouling is primarily caused by the undesired deposition of organic materials, inorganic particles, salts, colloids, and biological growth. Biofouling is another significant problem for membrane systems that is difficult to eliminate due to the microorganisms' ability to replicate [119]. Figure 3 below shows a generic filtration technique.

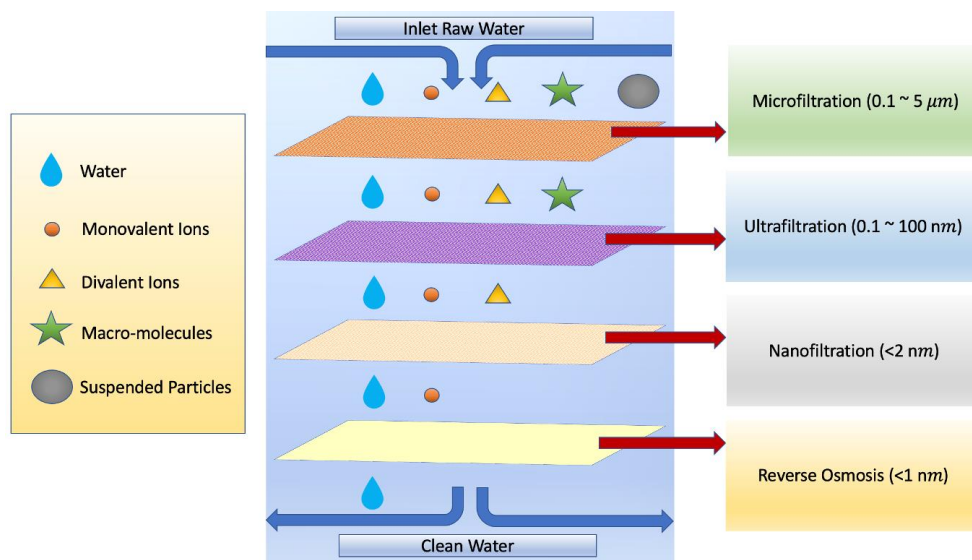


Fig. 3. Generic filtration technique

3.5 Adsorption

Adsorption is the process by which atoms or ions from a gas, liquid, or solid accumulate on a surface. The adsorbates (metal ions) are deposited as a layer on the adsorbent's surface during this procedure. Adsorption outperforms other techniques in terms of low energy consumption, ease of operation, and high removal efficiency. Adsorption can be physical (physisorption) or chemical (chemisorption). Van der Waals forces cause an increase in adsorbate concentration at an interface in physisorption, regardless of the nature of the adsorbate. Conversely, chemisorption is selective and typically irreversible, with covalent or ionic bonds formed between the adsorbate and the adsorbent.

Adsorbent efficacy is influenced by initial concentration, temperature, pH, contact time, stirring speed and competing ions during the process. According to Lakshmi and Ranjitha [120], the present analysis focuses on the adsorption approach for eliminating heavy metals from wastewater with diverse adsorbent materials obtained from natural waste. Many inexpensive adsorbents like sawdust, coal fly ash, activated carbon (A.C.), rice husks, coffee husks, fungi, algae, graphene, and chitosan have been researched to absorb harmful heavy metals from wastewater.

For example, a study by Praipipat *et al.*, [121] examined four different materials which were sawdust powder (S.P.), sawdust powder mixed with iron oxide-hydroxide (SPF), sawdust beads (SPB), and sawdust beads mixed with iron oxide-hydroxide (SPFB) to see their potential as adsorbents in removing lead or RB4 dye by utilising. All the materials are reusable for more than five cycles with high removal rates.

Aouan *et al.*, [122] simulated the mixture of kaolin (K), metakaolin (M.K.), and coal fly ash (CFA) to find the most effective combination of adsorbent materials for removing cationic dyes from wastewater. This approach used artificial intelligence to optimise the properties of adsorbents, eliminating the need to test the various combinations in the laboratory.

Another alternative in adsorbent technology is by using activated carbon (A.C.). However, large-scale implementation can be challenging due to its low effectiveness, lack of availability year-round, instability, limited surface area, and poor ability to absorb CO₂ [123]. The study used composite activated carbon (CAC) prepared by controlled carbonisation and continuous chemical activation of four wastes, such as peanut shell, coffee hush, corn cob, and banana peel. The results showed an

improvement in the removal efficiency and adsorption capacity of water purifying activity compared to the single-biomass-derived activated carbon.

According to Nor *et al.*, [124], waste cooking oil (WCO) shows promise as a viable carbon source for creating a composite material called graphene sand composite (GSC), which can be used to remove pollutants. The study introduces an environment-friendly method for synthesising GSC using WCO as a sustainable carbon source through thermal graphitisation. Results from this study show the effectiveness of GSCWCO as an adsorbent in removing Congo Red dye through batch adsorption with varies concentrations, optimum pH, contact time, and temperature.

The use of bacterial, fungal, and algal cultures, as well as their components, extracts, or biomolecules, as catalysts for the environmentally-friendly production of nanomaterials for adsorption activities is on the rise [125]. Saravanan *et al.*, [126] also stated that biosorption can be achieved using living and dead bacteria, fungi, and algae. Additionally, agro-waste materials were utilised as bio-sorbents because they have great surface properties, are widely available, and cost-effective. The surface characteristics of these bio-sorbents can be modified by employing various physical and chemical treatments.

Sadiq *et al.*, [127] highlighted the significance of chitosan as a practical adsorbent for removing heavy metals from an aqueous medium due to its valuable properties, availability, and affordability. In addition, hydroxyl and amino functional groups, which increase their chemical reactivity, are essential to adsorb various water contaminants. Chitosan and its derivatives can also be used as adsorbents to remove several other pollutants, including fluorides [128], pigments [129], and phenols [130].

4. Chitosan and Chitosan Composite as Adsorbent

Chitosan and chitosan composites are excellent adsorbents due to their high surface area and ability to form strong bonds with various contaminants. Chitosan is a biopolymer derived from chitin, commonly found in the exoskeletons of crustaceans and insects, as presented in Table 2 above. This unique material has an incredible ability to absorb heavy metals, dyes, and other pollutants from the environment. Chitosan composites, on the other hand, are made by combining chitosan with other materials, such as activated carbon or nanomaterials, to improve their adsorption capacity and selectivity. These composites can be tailored to specific applications, such as removing organic pollutants from wastewater or capturing CO₂ from flue gases [131].

The majority of chitosan adsorbents are submicron to micron-sized powders with an insufficiently porous structure for adsorption and high permeability. The diffusion capacity of the particle restricts mass transfer, thereby decreasing the rate of adsorption. To overcome these limitations, nanoscale adsorbents with increased specific surface area and enhanced efficiency have been developed [132]. Radwan, Abdel Ghaffar, and Ali [133] produced chitosan nanoparticles using gamma radiation, while Shijie *et al.*, [134] prepared chitosan nanoparticles by ionic gelation of chitosan with tripolyphosphate (TPP) anions. The production method for chitosan nanoparticles from gamma radiation is synthesised through a radiation-induced cross-linking process where the size and distribution of chitosan nanoparticles are smaller and narrower than those produced via ionic gelation [135].

On the other hand, chitosan nanoparticles produced from ionic gelation are formed through the interaction of chitosan with a cross-linking agent, typically an ionic solution [136]. Chitosan nanoparticles produced from gamma radiation generally exhibit higher stability due to the cross-linking induced by radiation because the cross-linked structure provides enhanced resistance to degradation and aggregation [137]. Both chitosan nanoparticles produced from gamma radiation and

ionic gelation share commonalities in terms of chitosan as the base material and biomedical applications [138].

Heavy metal ions generally exist in cationic form. Since chitosan film is negatively surface, it can easily attract metal ions towards the film until it reaches dynamic equilibrium. The amount of metal ion absorbed by chitosan film can be determined using Eq. (1).

$$A_{ab} = ((C_o - C_f) / C_o) \times 100\% \quad (1)$$

where A_{ab} is the percentage of adsorption, C_o and C_f are the initial and residual concentrations of metal ions in aqueous solution (mg/L) [139].

Isotherm profiles like Langmuir, Freundlich, and BET model usually represent the adsorption capacity. However, the Langmuir model is the most used isotherm for the adsorption of metal ions and dye adsorption because it denotes chemical adsorption and concentration in the liquid and solid phases [140].

4.1 Magnetic Chitosan Composite

Chitosan has also been used as a carrier for magnetic material to improve the efficiency of absorbing heavy metals in aqueous solution. This magnetic chitosan can attract divalent and hexavalent metal ions and rapidly separate adsorbents from absorbents [141]. Among the most prominent materials to be grafted with chitosan is Iron (II, III) oxide or Fe_3O_4 . For example, hybrid of $MnFe_2O_4$ and chitosan was studied as adsorbents to remove Cu (II) and Cd (II) from polluted water, and they were found to be effective when analysed with Langmuir adsorption isotherm [139]. Many other researchers also show the ability of magnetic chitosan to adsorb Cu^{2+} [142], Cd^{2+} [143], Pb^{2+} [144], and Hg^{2+} [145].

Similar results were achieved when acrylonitrile-PAN-g-Chitosan/ Fe_3O_4 , was studied under various conditions. It has been observed that a material exhibits remarkable reproducibility and reusability, as evidenced by its ability to undergo five adsorption-desorption cycles without experiencing any notable decline in its adsorption capacity for Cu(II) and Ni(II) ions [146]. Another study reported that Fe_3O_4 -graphene oxide-chitosan removes 96.73 % of Zn(II) in 38 min, with adsorption quantity $386.92 \text{ mg}\cdot\text{g}^{-1}$ [147].

On the other hand, a study by Wang *et al.*, [148] reported that a novel magnetic adsorbent consisting of $CoFe_2O_4$ /chitosan (C.S.) composite supported onto an alkalisated MXene (alk-MXene) sheet using a simple hydrothermal and self-assembly methods. This is due to its distinctive surface microstructure, abundant active sites, and varied functional groups.

The two-dimensional transition metal carbide/nitride has become increasingly popular in pollutant elimination. To put it simply, the results obtained from the research focus on the benefits of MXene, $CoFe_2O_4$, and C.S. also suggests a new method for creating magnetic adsorbents based on MXene that can efficiently remove both cationic and anionic dyes from water solutions.

Eltaweil *et al.*, [149] create a novel composite material that effectively removes Cr (VI) ions from water solutions by adsorption, offering high performance and improved recyclability. This composite combines the advantageous adsorption properties of aminated chitosan ($[NH]_2Cs$) derivative and attapulgite (ATP) clay, allowing their benefits to be synergistically utilized. The characterization tools used provided a clear understanding of the favorable thermal and magnetic properties of the attapulgite/magnetic aminated chitosan ($ATP@ [Fe]_3O_4 - [NH]_2Cs$) composite, as well as its larger surface area. Additionally, batch adsorption experiments demonstrated that the highest adsorption capacity was achieved at pH 2 using $ATP@ [Fe]_3O_4 - [NH]_2Cs$ (1:3). The final

findings of the research stated that the $ATP@ [Fe]_3O_4 - [NH]_2Cs$ can be reused for several times which is a beneficial for its application for removing of Cr(VI) ions from polluted water. The researchers concluded that the fabricated $ATP@ [Fe]_3O_4 - [NH]_2Cs$ composite could be applied as sustainable and reusable adsorbent for removing Cr(VI) ions from wastewater.

4.2 Carbon-Based Chitosan Composite

A number of researchers have also investigated the adsorption of heavy metal ions by carbonaceous chitosan. When carbon elements are incorporated into chitosan, its adsorption performance increases due to the formation of a porous structure that generates more ion adsorption sites. The incorporated carbonaceous materials include graphene oxide [150,151], biochar [152], as well as carbon nanotube [153,154].

Generally, chitosan has low acidic resistance but can absorb most heavy metal ions when sufficiently basic. In contrast, ions such as chromium exist as anions, causing the water to become acidic. When dissolved in a low-pH solution, chitosan forms a gel, rendering it incapable of absorbing ions. Thus, the Nowruzzi research group added Poly Vinyl Alcohol (PVA) and activated carbon to raw chitosan to improve its chemical stability in acidic conditions. Activated carbon (A.C.) is known for its large area of porosity, high adsorption capacity and, on its own, able to effectively remove several organic compounds from water. The adsorption rate of chitosan/PVA/AC composites was very fast at the initial stage, and the removal percentage increased with increasing A.C. content. They found that the highest adsorption capacity of chrome was 109.89 mg g^{-1} , and the highest adsorption occurred at $\text{pH} = 2$ [155]. However, earlier studies by Shen *et al.*, [156] showed higher adsorption capacity at 388.60 mg g^{-1} . They synthesised facile hydrothermal carbonisation chitosan that is stable in acid solutions, and the study suggested that its adsorption capacity remains stable to be reused at least five cycles.

Meanwhile, Alshaimi *et al.*, [157] have successfully fabricated a multifunctional composite by synthesising montmorillonite clay and activated carbon. This was achieved by employing chitosan as a crosslinking agent, thereby enhancing the overall performance and functionality of the composite. The experimental findings indicate a notable enhancement in the adsorption capability of the composite material towards Pb^{2+} ions with an increase in pH, up until the threshold of $\text{pH} 5.5$ was attained.

A transformation of graphene oxide nanomaterial (GON) in a nano size to a larger adsorbent was accomplished by linking it with chitosan, a biocompatible crosslinker. This linked material was then added to sand and abbreviated as graphene-chitosan composite (GCS). The adsorption efficiency of GCS was 7.8 mg/g . Chatterjee and Majumder [158] reported that the GCS was capable of being reused up to 10 times for adsorption-regeneration while maintaining a high level of adsorption capacity.

Al-Salman *et al.*, [159] synthesised chitosan/graphene-based nanocomposites utilising a solution-based approach, wherein their exceptional capacity for adsorbing cadmium metal ions was thoroughly examined. Consistent with previous investigations, it was observed that the adsorption rate exhibits an upward trend as the concentration of cadmium ions is increased.

4.3 Polymer and Functional Group Chitosan Composite

Chitosan faces challenges in terms of its effectiveness, ability to be reused, and susceptibility to degradation in acidic conditions. This is because it is a moderately weak base, causing it to lose its ability to adsorb substances. Therefore, extensive research has been conducted to address these

limitations by utilizing techniques such as grafting, adding more amine groups, carboxymethylation, and introducing N and/or O hydroxylation. Additionally, composite formation has been explored as a solution [160].

Rosli *et al.*, [161] demonstrated that chitosan nanofiber membrane cross-linked with poly (vinyl alcohol) (PVA) has the potential to act as a bio-sorbent for the removal of heavy metal ions, especially Pb^{2+} . According to Wu *et al.*, [162], heavy metal ions and organic dyes can be effectively adsorbed and separated when chitosan and PVA are spun with polyvinylpyrrolidone (PVP). A recent study by Rahman *et al.*, [163] stated that activated chitosan-clay bio nano-filter (ACCBNF) is an artificial mixture that was very crystalline and stable when exposed to heat. Additionally, these mixtures have excellent surface activity and can absorb large amounts of Ni^{2+} and Cu^{2+} . This is likely due to the combination of the chitosan molecule's reactive groups and the porous structure of the modified clay.

Another study by Sobhani [164] demonstrated a straightforward and eco-friendly hydrothermal method to produce $CuMn_2O_4$ /chitosan micro/nanocomposite. According to the findings, the micro/nanocomposites that were prepared demonstrated high effectiveness in adsorbing and eliminating methylene blue. Additionally, it can be utilised for up to five cycles of adsorption and desorption with great efficacy.

Meanwhile, Gan *et al.*, [165] reported a new type of adsorbent was fabricated by using chitosan as a base material and adding various functional groups, such as phosphoric acid, amidoxime, and quaternary ammonium groups, to enhance the adsorption speed and ability to capture uranium and vanadium. Overall, chitosan and chitosan-composites are promising adsorbents with a wide range of applications in environmental remediation, water treatment, and other industries.

Table 3 below shows a list of several chitosan-based material in detection and removal of metal ion.

Table 3

List of several chitosan-based material in detection and removal of metal ion

Chitosan/composite)	Metal ions	Main findings	Reference
Chitosan and clay composite	Nickel (II) (Ni^{2+}) and Copper (II) (Cu^{2+})	<ul style="list-style-type: none"> • Adsorption is directly proportional to amount of concentration. • Adsorption is directly proportional to amount of clay. • Number of adsorbents were between 2.57 – 5.98g. • Uppermost adsorption capacity for Cu^{2+} removal is 88.88%. • Uppermost adsorption capacity for Ni^{2+} removal is 81.13%. 	[166]
Chitosan-soya bean husk activated bio-char composite beads	Zirconium cation (Zr^{4+})	<ul style="list-style-type: none"> • Adsorption is directly proportional to amount of bio-char. • Adsorption efficiency is directly proportional to contact time. • As the size of particles increases, the removal efficiency decreases 	[167]
Chitosan membranes	Cu^{2+} , Ni^{2+} , Cadmium (II) (Cd^{2+}), and Lead (II) (Pb^{2+}).	<ul style="list-style-type: none"> • Rate of adsorption increases with time but become steady when approaching equilibrium which is attained in about 24 hours. • Adsorption is directly proportional to amount of concentration. 	[168]
Chitosan as plasma filter	Arsenic (As)	<ul style="list-style-type: none"> • Uppermost As removal efficiency is 76% 	[169]

(Chitosan cryogel)		with only 2-hour contact time with hydrogel.	
		<ul style="list-style-type: none"> • Follows Freundlich isotherm model. • Chitosan composite are reusable. 	
Chitosan coated with oil palm shell-based activated carbon	Methylene blue (MB)	<ul style="list-style-type: none"> • Adsorption is directly proportional to amount of concentration. • Adsorption efficiency is directly proportional to contact time. 	[170]

5. Advantages, Challenges and Future of Chitosan-Based Adsorbent

Chitosan-based adsorbents have several advantages over other types of adsorbents. The advantages of chitosan-based adsorbents are as follows:

- i. Chitosan is a natural and biodegradable polymer derived from chitin, found in the exoskeletons of crustaceans such as shrimp and crabs. This means that chitosan-based adsorbents are environmentally friendly and have low toxicity levels [171].
- ii. Chitosan-based adsorbents are also highly effective at removing heavy metals, organic pollutants, and dyes from wastewater. They can be used in a wide range of applications, including in the food [172], pharmaceutical [173], and textile industries [174].
- iii. These adsorbents have unique properties, such as high surface area, good mechanical properties, and low toxicity, which make them attractive for use in various industries.
- iv. However, there are also some challenges associated with chitosan-based adsorbents. For example:
 - v. They can be expensive to produce and may not be as effective as other types of adsorbents in certain applications [175].
 - vi. In addition, chitosan itself can be challenging to work with due to its insolubility in water and low stability at high pH values.
 - vii. Moreover, the stability of chitosan-based adsorbents can be affected by environmental factors such as humidity, which can impact their shelf life [176].

Chitosan-based adsorbents may be further optimised for improved efficiency and selectivity in their adsorption capabilities. There is also potential for developing new chitosan-based composite materials that could enhance their functionality and performance. Additionally, research is ongoing to explore using chitosan-based adsorbents in emerging areas such as drug delivery and tissue engineering. Overall, future view potential for chitosan-based adsorbents as they continue to be studied and developed for a wide range of applications.

6. Conclusions

Chitosan can be utilised as an adsorbent to discard heavy metal ions while also reducing environmental pollution. Aside from adsorption techniques, this article discusses various chitosan-based heavy metal ion removal methods such as electrocoagulation (E.C.), bioremediation, ion-exchange, and membrane filtration (purification). Adsorption has been identified as the best effluent removal method due to its advantages: a large physical area of application and a short treatment time. As an adsorbent, modified chitosan is more effective compared to unmodified chitosan. However, researchers have found that modifying chitosan by using harmful modification agents might affect the adsorption capacity. It is recommended that further investigation be conducted on

chitosan modification techniques beyond the utilisation of nano-chitosan. Consequently, the characteristics of chitosan that have been modified can be amplified, thereby augmenting its efficacy as an adsorbent and ultimately mitigating potential environmental contamination.

Acknowledgement

The authors are grateful to Universiti Putra Malaysia for the financial support through IPM Putra research grant 9674000.

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