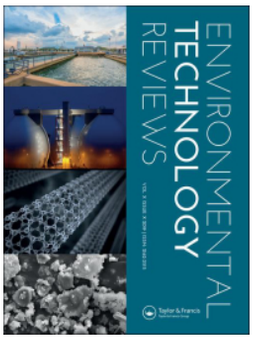

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ISSN: (Print) (Online) Journal homepage: www.tandfonline.com/journals/tetr20

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To cite this article: Arvind Kumar, Riona Indhur, Abdul Gaffar Sheik, Suresh Babu Naidu Krishna, Sheena Kumari & Faizal Bux (2024) A review on conventional and novel adsorbents to boost the sorption capacity of heavy metals: current status, challenges and future outlook, Environmental Technology Reviews, 13:1, 521-543, DOI: 10.1080/21622515.2024.2377801

To link to this article: https://doi.org/10.1080/21622515.2024.2377801

ENVIRONMENTAL TECHNOLOGY REVIEWS
2024, VOL. 13, NO. 1, 521–543
https://doi.org/10.1080/21622515.2024.2377801

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REVIEW ARTICLE

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A review on conventional and novel adsorbents to boost the sorption capacity of heavy metals: current status, challenges and future outlook

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ABSTRACT

Heavy metal contamination in aquatic environments has attracted global concern due to its detrimental impact on living organisms, including toxicity and carcinogenicity. Therefore, initiatives to remove these contaminants from wastewater to protect both the environment and the well-being of humans is imperative. Materials utilized for wastewater treatment in the developing world needs to satisfy the main criterion of being locally available, inexpensive and that do not require additional energy input or modifications. Adsorption is the foremost used technique to eliminate heavy metals due to its attractive features including flexible design, operation, and cost-effectiveness. This review article potentially focusses on the use of novel and conventional adsorbents such as g-C3N4, graphene, cellulosic aerogel, natural occurring soil and minerals, aquatic and terrestrial biomass-based adsorbents for heavy metal adsorption, whilst focusing the discussion on the context of the developing world. This heightened interest in g-C3N4, and graphene-based adsorbents owes their unique properties such as great surface area, impressive chemical and thermal stability, three-dimensional structure, and functionalized derivatives which have shown great adsorption capabilities for heavy metals elimination. Additionally, cellulosic aerogel, natural occurring soil and minerals, aquatic and terrestrial biomass is low cost, easily available and effectively removes heavy metals. Finally, current progress, major challenges and future outlook of novel and conventional adsorbent materials for heavy metal removal are discussed. Herein a roadmap is presented together with recommendations for future research to enhance the successful utilization of bio-sorbents in water purification systems such as wastewater treatment plants.

ARTICLE HISTORY
Received 1 February 2024
Accepted 22 June 2024

KEYWORDS
Heavy metals; CNTs; functionalized graphene oxides; sustainable materials; adsorption

Highlights

- A comprehensive review of low cost and conventional materials for heavy metals removal is presented.
- Advanced materials like CNTs and graphene exhibit excellent sorption capacity of heavy metals.
- Current status, challenges and future outlook of heavy metal adsorption with low cost and conventional materials highlighted.

1. Introduction

Over the past few decades, industries have recorded a quantum jump in industrialization, urbanization and agricultural practices. This has globally catapulted economic growth but concomitantly given rise to severe environmental pollution [1]. The environmental burden is gaining momentum as a result of water

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Supplemental data for this article can be accessed online at https://doi.org/10.1080/21622515.2024.2377801.

insufficiency and water contamination since industrialization, urbanization and agricultural practices lead to disposal of highly polluted wastewater (WW) [2]. This is further exacerbated by the limited availability of water due to diminishing natural water supports [3]. Due to the rapid urbanization, approximately 40% of the global populace are experiencing the detriment of water scarcity [4]. The water demand is growing since fresh water is being used for industrial and agricultural purposes. This has attracted a considerable amount of attention and created significant concern among scientists globally. This concern can be mitigated and potentially solved using reclaimed wastewater as a water supply [5]. Although this re-claimed WW supply may reduce the water burden, it is critical that the constituents be identified, enabling the remediation of the WW. These industrial WWs are primarily riddled with toxic organic pollutants and heavy metals since they have been involved in a plethora of industrial activities such as medicinal, petroleum refineries and pharmaceutical procedures. These activities deposit a variety of waste substances and toxic pollutants into the water [6]. Heavy metals raise specific worries because of their toxicity and carcinogenic properties, coupled with their well-documented detrimental impacts on human well-being [7]. Heavy metal contamination has increased due to the expeditious nature of urbanization and industrialization. This has considerably accelerated rates of movement and mobilization of heavy metals that ae present in the environment. For example, the WW generated by the petroleum refining industries contains inorganic waste consisting of heavy metals and organic waste consisting of hydrocarbons and benzenes. While heavy metals such as Co, Mn, Ni, Hg, Cu, Fe, Pb, Zn, Cd and others are typically found in small quantities, they are recognized as the most harmful and prevalent constituents in wastewater effluent [2]. These toxic heavy elements cannot be processed by the body, resulting in their buildup in soft tissues, posing a threat to human well-being as other effects depicted in Figure 2(b). All industrial wastes, specifically toxic pollutants and heavy metals in WW, are greatly detrimental on both the environment and marine

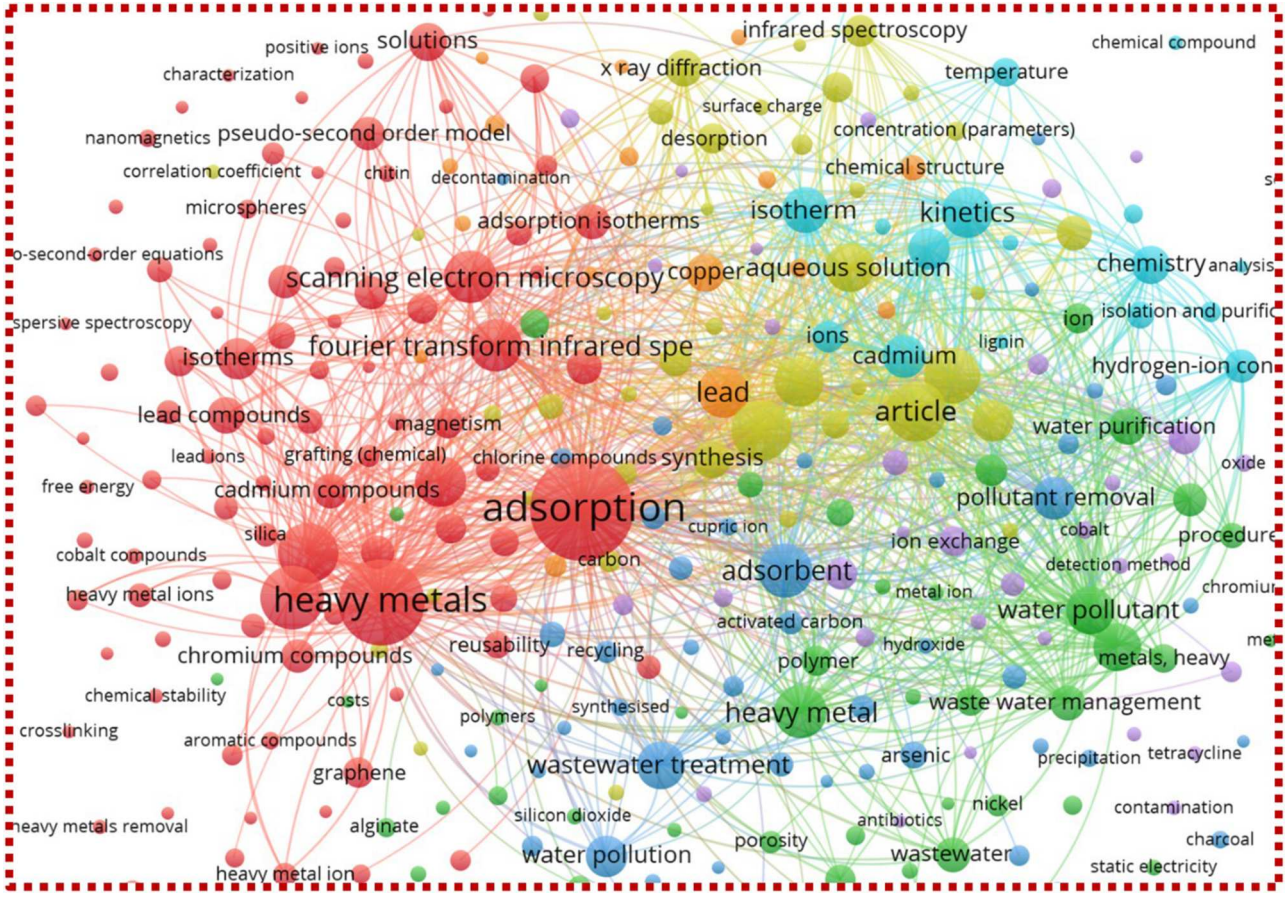
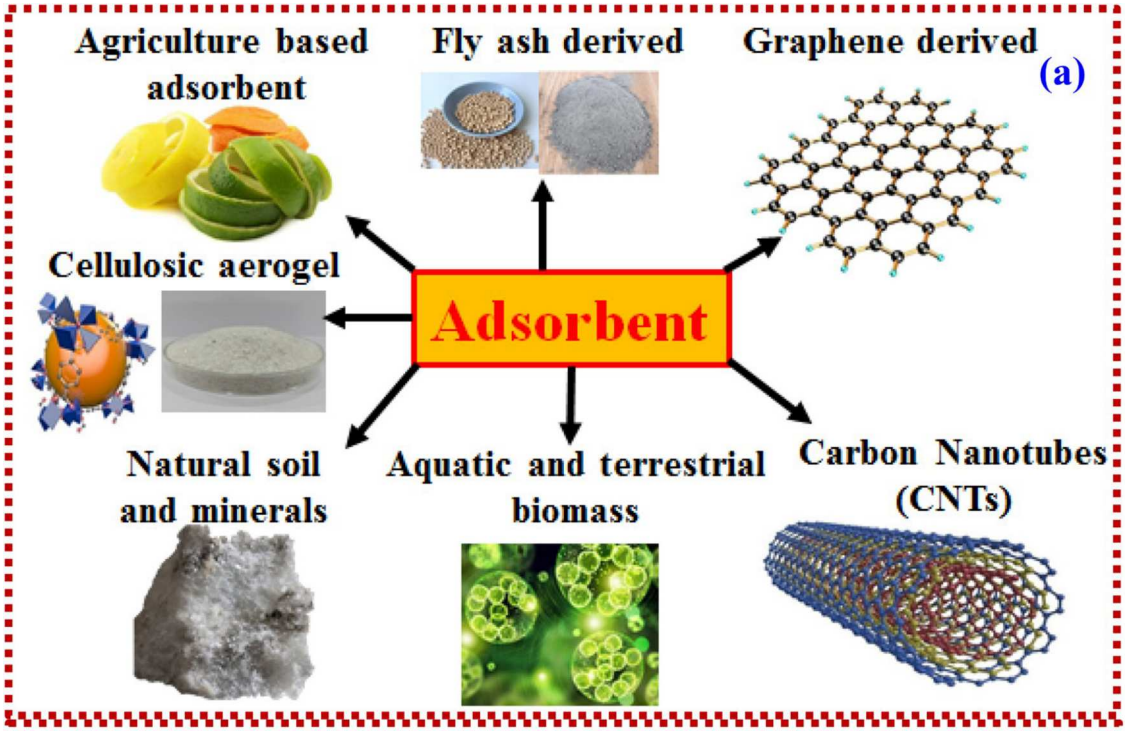


Figure 1. A comprehensive network of research trends of adsorption of heavy metals and their related topics.



(b)

Heavy Metals	Maximum Permissible Limit (mg/L)		Health effects
	WHO	USEPA	
Pb (II)	0.01	0.05	▪ Affect on kidney, liver and memory slow growth, hearing and anemia in children, Cardiovascular effects increased blood pressure, hypertension
Cu(II)	1.0	0.25	▪ Lung cancer, abdominal pain, Diarrhea, liver toxicity, weakness
Zn(II)	3.0	1.0	▪ Abdominal pain, phyto-toxicity, anemia
Ni(II)	-	0.2	▪ Affect renal functioning, DNA damage, Eczema
Hg(II)	0.01	0.05	▪ Degradation of nervous, digestive and immune systems, lungs, kidneys, gastrointestinal tract
Cr(VI)/(III)	0.05	0.05/0.1	▪ Kidney circulation, lung cancer, dermatitis
Cd(II)	0.003	0.005	▪ Harmful effects on bones, heart, kidney, lungs, and liver of human being
As(III)	0.010	0.05	▪ Carcinogenic and dermatological, cardiorenal and gastrointestinal effects

Figure 2. (a) Classification of various low cost adsorbent materials for heavy metal removal, (b) Permissible limits and health effects of toxic heavy metals

systems. This is because some of these heavy metals affect the regulation of acid–base ions, disrupting the gills' function and harming the fish at the disposal site [2]. In addition, Heavy metal contamination is also a matter of apprehension, since several of the methods for treating and purifying drinking water in developing regions, such as solar disinfection, boiling and chlorination prove ineffectual in eliminating these pollutants [8]. A comprehensive network of research trends of adsorption of heavy metals and

their related topics as shown in Figure 1. Additionally, heavy metals are poisonous to both fauna and flora through bioaccumulation or bio-magnification. While found in low quantities, heavy metals influence critical soil microbial processes that hinder plant metabolism and hinder growth [9]. Furthermore, because of the extensively established adverse consequences of heavy metals on humans' well-being, substantial research has been carried out to develop techniques for eliminating heavy metals from potable water supplies, industrial wastewater, municipal wastewater, and additional water supplies. Various treatment approaches and cutting-edge technologies with high efficacy in removing heavy metals are presently under investigation for decontaminating water from these pollutants. These include electrochemical treatment, solvent extraction, reverse osmosis, adsorption, membrane filtration, ion exchange and chemical precipitation [10]. Nonetheless, within the framework of the developing world, these technologies prove impractical and economically unsustainable. To address water treatment needs in developing regions, the suggested technologies must be readily accessible, constructed by local labourers with limited education, and possess minimal operating and maintenance expenses [11]. This highlights the urgent need for strategies and technologies that may be implemented for the remediation of toxic pollutants and heavy metals in WW that satisfies these criteria. Herein, our primary focus revolves around the utilization of economical materials, often sourced locally, which do not necessitate additional energy or modifications to eliminate heavy metals from water sources. Although providing a comprehensive overview of studies conducted in developing regions on heavy metal removal presents challenges, the goal of this review is to inspect the primary categories of materials readily accessible and employable within the context of the developing world for heavy metal remediation. Adsorption proves highly suitable for removing heavy metals from wastewater due to its noteworthy benefits, such as its cost-effectiveness, simplicity, adaptability, and superior adsorption capacity attributed to its extensive surface area [12]. The main benefit of the adsorption process lies in its reversibility, since adsorbents have the ability to be recycled via regeneration through desorption. Additional key benefits stem from adsorbent synthesis making use of industrial byproducts, natural materials and agricultural waste. as well as simple adsorption mechanism. The adsorption process involves the four major steps as follows: bulk diffusion, film diffusion, pore diffusion, and finally adsorbate attached on the active sites of adsorbent through chemical or physical reactions as mechanism depicted in Figure 3(a). However, the primary limitation associated with employing adsorption methods is the significant expense of the materials involved, restricting their application to laboratory-scale settings. Over recent decades, a plethora of adsorbent nanocomposites have been employed for heavy metals elimination, including agricultural and terrestrial biomass-based materials, functionalized carbon nanotubes (CNTs), carbon nitride, adsorbents derived from cellulosic materials, biochar/layered double hydroxide (LDH) composites, graphene-based materials, magnetic nanorods, boron-based substances, biochar and more.

2. Novel/Innovative and traditional composites for heavy metal elimination

Two branches of adsorption exist: chemical (chemisorption), which is initiated by chemical interactions between the adsorbent and adsorbate, resulting in the formation of ionic or covalent bonds, and physical adsorption, where the rise in adsorbate concentration at the interface occurs due to non-specific van der Waals forces (independent of the substance's nature) [12]. In order to gain a more comprehensive insight into the attributes of adsorbent materials, characterization techniques are illustrated in Fig. S1 (in supplementary file). Adsorbent efficacy in heavy metals elimination from wastewater is contingent upon several factors, including the agitation rate, contact duration, pH, adsorbent quantity, temperature and initial concentration. Additionally, because some occurrences of adsorption are reversible, adsorbents can be regenerated by desorption [12]. Herein, we seek to bridge the gap between cost-effective technologies and the developing world. Therefore, we explore cost-effective adsorbents and classified them in four categories (as depicted in Figure 2(a)) (1) carbon nanotubes (CNTs) and graphene-based adsorbents, (2) functionalized cellulosic aerogel-based, (3) natural occurring soil and mineral-based, and locally occurring biomass-based adsorbent; (4) agriculture, aquatic and terrestrial based adsorbents.

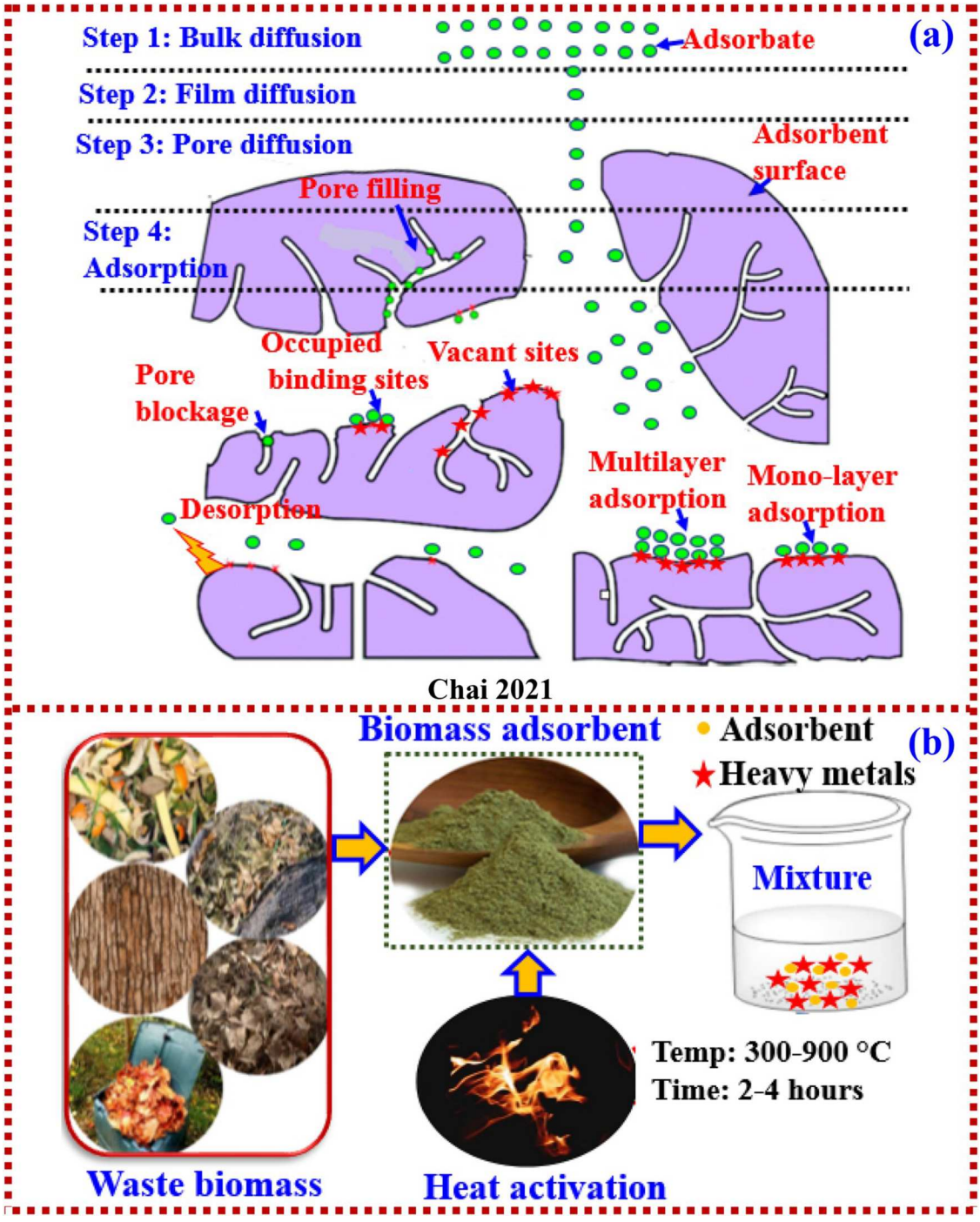


Figure 3. (a) A general overview on the adsorption mechanism, (b) Schematic of bio-adsorbent preparation by heat treatment.

The discussion of the performance of aforementioned materials as follows.

2.1. Carbon nano tubes (CNTs) adsorbent

Carbon nanotubes (CNTs) have received an unprecedented amount of interest in the elimination of heavy metals from wastewater due to their layered structure, great specific surface area, hollow and small size, as well as high porosity [13]. Utilizing CNTs has been evidenced to be efficacious in heavy metals elimination [14], toxic pollutants [15] and heavy metal metabolites [16] from wastewater. The adsorption capacity of modified and functionalization on CNTs and MWCNTs has been explored for the elimination of various heavy metals as summarized in

Table 1. Sorption performance of heavy metals media by CNTs and graphene-based adsorbent.

S.No	Adsorbent	Heavy metals	Reaction conditions	Adsorption /Removal	Reference
1	Thiol functionalized MWCNTs	Pb (II)	$[AD]_0$ = 0.25-3 g/L, C_0 = 10 mg/L, pH = 2-5.5, temp = 25 °C, time = 5-40 min	144.9 mg/g	[21]
2	MWCNTs-KOH@NiNPs	Pb (II), As (II), Cd (II)	$[AD]_0$ = 5-30 mg/L, C_0 = 60 mg/L (each), pH = 5, temp = 30 °C, time = 2 h	481 mg/g – Pb (II), 441 mg/g -As (II), 416 mg/g – Cd (II)	[20]
3	MWCNTs	Cd(II) Pb (II)	$[AD]_0$ = 5 mg, C_0 = 1 mg/L, pH = 2-8, temp = 25 °C	13.5 mg/g - Cd(II), 27.9 mg/g -Pb (II),	[65]
4	MWCNTs	Ni (II)	$[AD]_0$ = 0.5 g/L, C_0 = 5 mg/L, pH = 7, temp = 25 °C	115.8 mg/g	[66]
5	Fe_3O_4/MWCNTs-COOH	Cu (II)	$[AD]_0$ = 0.05-0.4 g/L, C_0 = 2-20 mg/L, pH = 7, time = 0-1 h, temp = 25 °C	10.45 mg/g	[19]
6	ZnO- MWCNTs	Cr (VI)	$[AD]_0$ = 100 mg/L, Co = 15 mg/L, pH = 2, temp = 25 °C, time = 2 h	152.2 mg/g	[17]
7	Al_2O_3-MWCNTs	Co (II)	$[AD]_0$ = 1.63 g/L, C_0 = 52.15 mg/L, pH = 10, temp = 25 °C, time = 35.5 min	93% removal	[18]
8	Chitosan coated CNT	Cu (II), Cr (VI)	$[AD]_0$ = 0.5 g/L, C_0 = 20-400 mg/L (each), pH = 3-6, time = 11 h	142.1 mg/g, 123.7 mg/g	[67]
9	Chitosan coated CNT	Cu (II)	$[AD]_0$ = 0.2 g/L, C_0 = 10 mg/L, pH = 2-11, temp = 25-70 °C time = 0-90 min	115.84 mg/g	[22]
10	PHB-CNTs, *P*-CNTs	As, Pb, Cr, Cd, Ni, Cu, Fe, and Zn	$[AD]_0$ = 2-5 g/L, C_0 = 5-167 mg/L, pH = 6.5, time = 10-80 min, temp = 30-70 °C	77-99.5% removal	[68]
11	PAMAM/CNT nanocomposite	As(III), Co(II), Zn(III)	$[AD]_0$ = 0.03 g/L, C_0 = 100 mg/L, pH = 2-7, time = 30 min	432 mg/g, 494 mg/g, 470 mg/g	[69]
12	Fe_3O_4-GO-(o-MWCNTs)	Cd(II), Cu(II), Ni(II)	$[AD]_0$ = 50–330 mg/L, C_0 = 190 mg/L, pH = 5, temp = 25 °C, time = 3 h	625 mg/g, 574 mg/g, 384 mg/g	[30]
13	3-aminopyrazo -le/MWCNTs	Cd(II)	$[AD]_0$ = 50–500 mg/L, C_0 = 10-20 mg, pH = 6-10, temp = 25-45 °C, time = 2 h	83.7% removal	[70]
14	F-MWCNTs	Cu(II)	$[AD]_0$ = 10 mg/L, Co = 20 mg/L, pH = 3, temp = 25 °C, time = 1 h	118.4 mg/g	[71]
15	F-CNT-HOX	As(III) As(V)	$[AD]_0$ = 100 mg/L, C_0 = 0.1-10 mg/L, pH = 6.5, time = 4 h	169 mg/g	[72]
Graphene-based adsorbent					
16	GO-*g* P4VP @PAA hydrogel	Pb (II), Cd (II),	$[AD]_0$ = 0.5-2.5 g/L, C_0 = 1 g/L, pH = 2-6, time = 2.5 h, temp = 25-45 °C	257.28, 175.79 mg/g	[25]
17	Thiol-functionalized graphene oxide	Hg (II)	$[AD]_0$ = 0.2 g/L, C_0 = 10 mg/L, pH = 2-8, time = 36 h, temp = 15-35 °C	49.6 mg/g	[27]
18	Granules GO -Gd_2O_3	Cr (III), Pb (II), As(V)	$[AD]_0$ = 1.4 g/L, C_0 = 1 g/L (each), pH = 4, time = 2 h, temp = 25 °C	29.16 mg/g, 158.23 mg/g, 36.77 mg/g	[73]
19	rGOH	Pb (II), Cd(II)	$[AD]_0$ = 100 mg/L, C_0 = 30 mg/L (each), pH = 4, time = 30 min, temp = 25 °C	250.62, 118.27 mg/g	[74]
20	Magnetic graphene oxide	Pb (II)	$[AD]_0$ = 10-50 mg/L, C_0 = 5-25 mg/L (each), pH = 2-10, time = 5-25 min, temp = 25 °C	98% removal	[28]
21	Graphene oxide-TETA-DAC	Cu (II), Pb (II)	$[AD]_0$ = 10-500 mg/50 mL, C_0 = 100 mg/L, pH = 2-7, time = 1 h, temp = 25 °C	65.1 mg/g, 80.9 mg/g	[75]
22	Graphene oxide function-alized DTC	Pb (II)	$[AD]_0$ = 250 mg/L, C_0 = 20-100 mg/L, pH = 2-5.8, time = 2 h, temp = 25-45 ∘C	132.1 mg/g	[76]
23	3D-MnO_2 nanotubes@rGO oxide	Pb (II)	$[AD]_0$ = 5-250 mg/L, C_0 = 200 mg/L, pH = 5, time = 500 min, temp = 25 ∘C	356.37 mg/g	[77]
24	TI/GO@Fe_3O_4	Pb (II)	$[AD]_0$ = 200 mg/L, C_0 = 100 mg/L, pH = 5, time = 2 h, temp = 20 ∘C	461 mg/g	[29]
25	Graphene oxide/NiO	Cr (VI)	$[AD]_0$ = 0.2 g/L, C_0 = 1/3 mg/mL, pH = 2-11, time = 1 h, temp = 10-50 °C,	198 mg/g	[78]
26	Graphene oxide - MgAl-LDH	Pb (II), Cu (II), Cd (II),	$[AD]_0$ = 0.01-0.15 g, C_0 = 80-300 mg/L, pH = 2-9, time = 6 h, temp = 25 °C	192.3 mg/g, 45.05 mg/g, 23.04 mg/g	[79]
27	Graphene oxide/Fe-Mn (GO/Fe-Mn)	Hg (II)	$[AD]_0$ = 8 mg/L, C_0 = 0.1-5 mg/L, pH = 7, time = 24 h, temp = 25 °C	32.9 mg/g	[80]
28	EDTA-mGO	Pb(II), Hg(II), Cu(II)	$[AD]_0$ = 0.05-0.12 g/L, C_0 = 100 mg/L (each), pH = 1-7, temp = 25-45 °C	508.4 mg/g, 268.4 mg/g, 301.2 mg/g	[81]
29	NH_2-rich polymer/graphene oxide	Cu (II)	$[AD]_0$ = 0.1 g/L, C_0 = 10-210 mg/L, pH = 6, time = 24 h, temp = 20 °C,	349.0 mg/g	[82]

[AD] – Adsorbent dose; C_0 – Metal concentration.

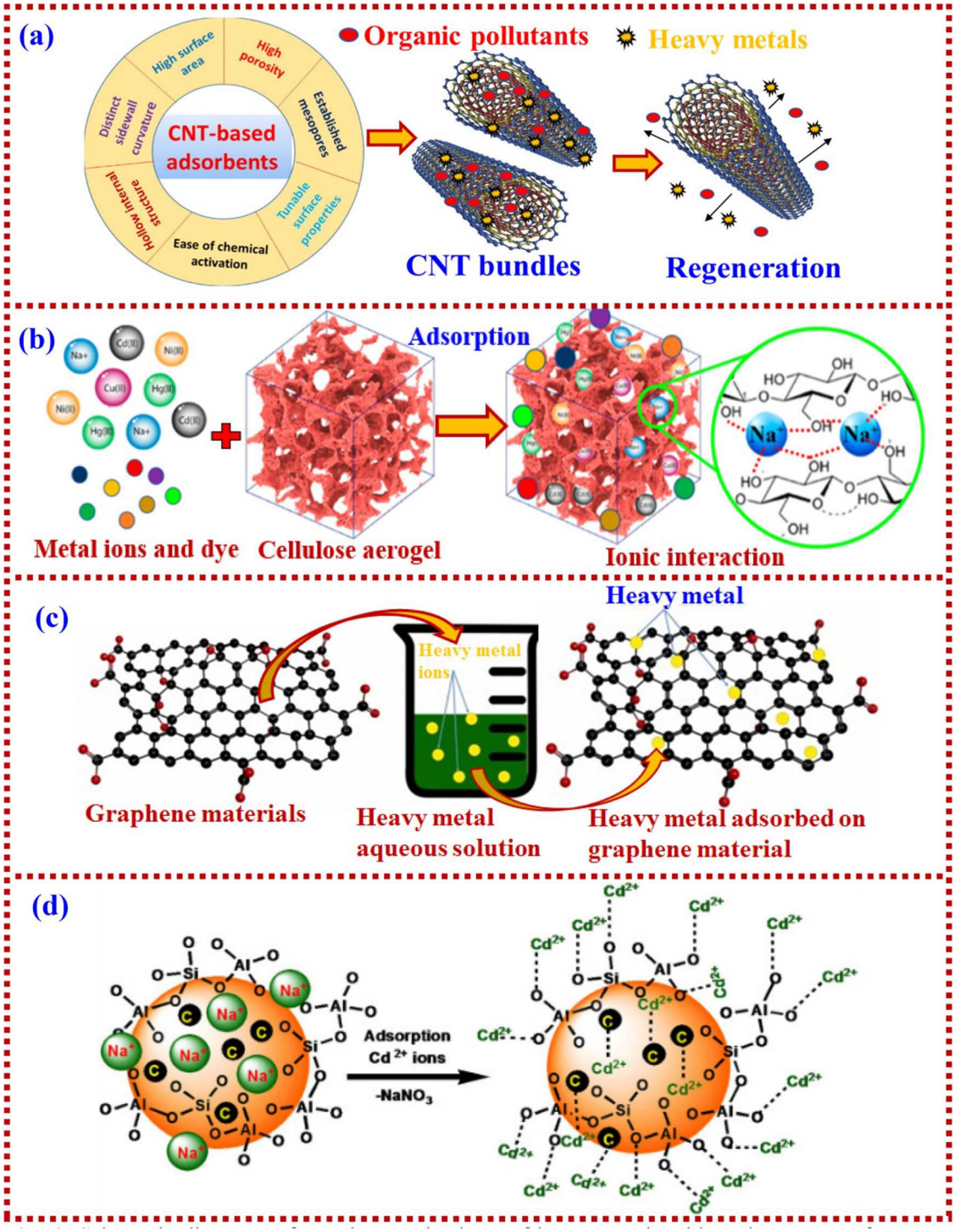


Figure 4. Schematic diagrams of sorption mechanisms of heavy metals with various types of adsorbent materials, (a) CNTs, (b) cellulosic aerogel, (c) graphene, and (d) clay and minerals based.

Table 1. The surface adsorption mechanism of CNTs based adsorbent for heavy metals elimination depicted in Figure 4(a). Recently, authors Wang et al., successfully synthesized the functionalized magnetic Fe_3O_4-GO-(o-MWCNTs) and treated the heavy metal ions Cd (II), Cu (II), and Ni (II) from aqueous media. Under optimum operating condition maximum adsorption of metal ions was observed, i.e. 625, 574, and 384 mg/g, respectively [17]. CNTs have been modified with various non-magnetic metal oxides such as ZnO, Al_2O_3, Fe_3O_4, NiO, as along with bimetallic Pd/Fe-Fe_3O_4 to enhance their attraction to contaminants.

The metal oxide-coated CNTs effectively captured hazardous metal ions, including Co(II), Pb(II), Cu(II), Fe(II), Cd(II) and Cr(VI) from water [18–21]. A new study by authors Egbosiuba et al., synthesized the multiwall various CNT (MWCNTs) based adsorbents labelled MWCNTs-KOH, NiNPs, and MWCNTs-KOH@-NiNPs and utilized for decontamination of Cd(II), As (II) and Pb(II). Among all, adsorbent MWCNTs-KOH@-NiNPs exhibited the greatest surface area of 1242 m^2/g. Herein, they observed the maximum sorption capacity, i.e. 481 mg/g – Pb (II), 441 mg/g – As (II), 416 mg/g – Cd (II) with adsorbent MWCNTs-KOH@-NiNPs under operating condition (adsorbent dose 5–30 mg/L, each metal concentration 60 mg/L, temp 30°C, pH 5, time 2 h) [21]. Chemical oxidation is a common method for modifying carbon nanotubes (CNTs), involving the introduction of oxygen-based functional groups, such as -COOH, -C = O, and –OH. This process enhances the CNTs' ability to attract heavy metal cations [22]. MWCNT-SH functionalized with thiol was synthesized, exhibiting outstanding adsorption capabilities for the elimination of Pb(II). Heavy metal Pb(II) maximum adsorption was noticed to be 145 mg/g under conditions (i.e. pH 5, temp 298 K, Pb(II) initial concentration 10 mg/L, adsorbent dose 2 g/L) [23]. In the case of Cr (VI) adsorption, MWCNTs functionalized with ZnO (ZC) was prepared, which have ZnO nanoparticles grafted on them was synthesized using the hydrothermal method. ZC-10 (MWCNT functionalized with 10 wt%) showed high adsorption affinity, and at pH 2, 94% heavy metals was eliminated [17]. In case of Co(II) removal from aqueous solutions using (MWCNTs) and γ- alumina a hybrid approach employing genetic algorithms and artificial neural networks for modelling was used. RSM optimization stipulates the maximum Co (II) removal ~ 90% under optimum conditions (Co(II) initial concentration of 56.57 mg/L, contact duration 38.6 min, MWCNT dose 1.57 mg/L, pH 10,). while γ-alumina was used, maximum Co (II) removal was observed ~ 93% under optimum conditions (contact period 35.5 min, pH 10, Co(II) initial concentration 52.15 mg/L, γ-alumina dose 1.63 g/L). Additionally, γ-alumina adsorbents and MWCNTs had the highest monolayer adsorption capabilities of 75.78 and 78.94 mg/g, respectively [18]. Apart from that, CNTs are also modified with chitosan based on mercaptoacetic acid locking imine (MALI). Herein, maximum sorption capacity of Cu(II) was noticed to be 115.8 mg/g when the pH was 7, Cu(II) initial concentration 10 mg/L and adsorbent dose 10 mg, within 1 h [24].

2.2 Graphene-based adsorbents

In the realm of heavy metal elimination, modified graphene-based resources have drawn a great amount of attention recently. Physical adsorption and redox reaction are the key benefits of using modified graphene-based materials to remove heavy metals in wastewater. Heavy metals are rapidly taken up through physical adsorption by leveraging their inherent traits, such as their extensive specific surface area. Numerous researchers have utilized graphene-based materials for the purpose of heavy metal eradication as reported in Table 1. The surface adsorption mechanism of graphene-based adsorbent for heavy metals elimination depicted in Figure 4(c). The remediation of highly chemically valent and toxic heavy metals in wastewater involves a redox reaction that reduces the metals valence state following adsorption, as described by Xu and colleagues in their 2022 study [25]. For example, Chi and team synthesized a new surface-ion-imprinted magnetic polymer (IIP) utilizing GO/Fe_3O_4 as support and As(III) as a template. Herein, the IIP has displays great reusability, showcasing a 75% adsorption efficiency even after undergoing five regeneration cycles and reuse.. Moreover, it demonstrates a remarkable selectivity for As(III) even in the presence of other rivalling constituents, including, As (III)/HA, As(III)/SO_4^{2-}, As(III)/Mg(II), As(III)/PO_4^{3-}, As(III)/HCO_3^- and As(III)/Ca(II) [26]. In a study by Zhang et al., demonstrate the GO-g-P4VP@PAA hydrogel showed the excellent adsorbent affinity for Pb(II) 257 mg/g and Cd(II) 175 mg/g. Even after undergoing five cycles, the eradication of Cd(II) and Pb(II) still remained at levels exceeding 80% [27]. In order to functionalization of GO in the presence of N,N′-dicyclohexylcarbodiimide (DCC) and synthesize magnetic nanocomposites of MGO@APhen, Abaszadeh used Fe_3O_4 nanoparticles and 5-amino-1,10-phenanthroline (APhen). These materials demonstrated Outstanding recyclability over six cycles following regeneration with the use of a powerful magnet [28]. Another material, graphene oxide functionalized with thiol groups (GO-SH) was synthesized and employed to efficiently eliminate Hg(II) from water solutions. The majority of Hg(II) was effectively extracted within a

36-hour. Apart from this GO-SH showed the high stability and remained the Hg(II) removal efficiency more than 91.8% over the sixth cycle [29]. To eliminate Pb (II) from water solutions, research was conducted on magnetic graphene oxide (MGO) nanocomposites that were modified with varied amine ligands. The optimal conditions for Pb(II) eradication were Pb(II) initial concentration 20 mg/L, pH 4, time 10 min and adsorbent dose 40 mg. Following that, an exploration was carried out into the variables influencing the adsorption effectiveness, including pH, adsorbent amount, the duration of contact, and concentration of Pb.Pb(II) adsorption capacities of ethylenediamine (EDA), melamine, and monoethanolamine functionalized adsorbents were 98%, 96.34%, and 97.65% removal, respectively. After 5 consecutive cycles of both adsorption and desorption, the Pb(II) adsorption efficiency of the functionalized EDA with MGO (EDA-MGO) adsorbent decreased by 8.8% [30]. The in situ growth approach was used to successfully synthesis TI/GO@Fe_3O_4 for Pb(II) elimination. Oxygen-containing functional groups fixed the metal ions (Fe^{3+}) on the surface of GO and acted as growth sites for Fe_3O_4. Moreover, triisopropanolamine and its products of oxidation (TI), which facilitate Pb(II) ions elimination, enhanced the surface of GO and Fe_3O_4 nanoparticles. Adsorption studies revealed, under ideal circumstances (10 mg TI/GO@Fe_3O_4, 120 min pH 5.0, 50 mL 500 mg/L Pb(II), 293 K), the adsorption cabability of Pb(II) on TI/GO@Fe_3O_4 may reach 461.00 mg/g [31]. Authors Wang and team explore the effect of pH (2-10) on functional hybrid adsorbent γ-PGA-Fe_3O_4-GO-(o-MWCNTs) for the sorption of Cd(II), Cu(II) and Ni(II) and found the maximum adsorption 431.83, 416.45 and 233.11 mg/g, at pHs 8, 5, and 5, as results shown in Fig. S2 (a-d) [17]. Furthermore, Lingamdinne and colleagues' study involved creating a hybrid material known as graphene oxide-based inverse spinel nickel ferrite (GONF) nanocomposite, which was then applied to eliminate heavy metals from water solutions. The GONF exhibited a maximum sorption capacity of 45.50 mg/g for Cr(III) and 25.0 mg/g for Pb(II) [32].

2.3 Functionalized cellulosic aerogel-based adsorbent

Several authors have noted that cellulosic aerogel-based adsorbent exhibited the remarkable sorption capability for heavy metals elimination with results summarized in Table S1 (supplementary file). Heavy metals sorption mechanism on cellulosic-based materials depicted in Figure 4(b). A crosslinking process between cellulose filament fibres and bis-acrylamide was used to create a cellulose-based amide-functionalized porous adsorbent at 25°C. This procedure is easy, quick, and affordable, and it has a lot of promise for industrial applications. Due to the produced material's extensive adsorption sites, anionic dyes and copper ions may be removed from aqueous solutions with great efficiency. The maximum adsorption capacities were 751.8, 417.9, and 51.3 mg/g for the dyes Acid Black 1 and Acid Red 18, as well as for copper ions. After five successive recycling trials, regeneration testing revealed that the reduction efficiency for all model contaminants persisted over 92% [33]. $MnFe_2O_4$ was in situ incorporated into the cellulose hydrogel matrix to create $MnFe_2O_4$-Cellulose magnetic composite aerogels (MnCAs) with significant metal ion adsorption capability. The adsorption analysis confirmed that MnCA had a quick adsorption rate and high adsorption capability for heavy metal ions reduction in aqueous solution, that can achieve 63.3 mg/g sorption during 100 min. Mostly, all aerogels showed excellent recyclability and reusability [34]. A green bacterial cellulose graphene oxide composite material was synthesized and characterized. Experiments with desorption and re-adsorption revealed that 0.1 M HCl and 0.1 M HNO_3 may be utilized to recover and reuse both adsorbent and adsorbates with an efficiency of over 90–95% [35]. Novel and innovative aerogel absorbent materials are created through the electrostatic combination of polyethyleneimine (PEI) and nano-fibrillated cellulose (NFC), resulting in the creation of NFC/PEI composite hydrogels (NPHxy) and subsequently aerogels (NPA). NPAs were able to adsorb a maximum of 175.44 mg/g of Cu(II) and 357.14 mg/g of Pb(II), correspondingly [36]. A directional freeze-drying method was used to produce a chitosan (CS)/nanofibrillated cellulose (NFC) aerogel with oriented microchannel architecture. The findings show that Pb(II) has a maximum adsorption capability of 252.6 mg/g, which is significantly greater than the majority of chitosan-based adsorbents described in the literature according to Li and team [37]. Aerogels were produced through a coating process involving the cross-linking of CNFs with polyethylenimine (PEI) and the addition of polydopamine. This coating

method utilized was driven by mussels. From this investigation the corresponding results are reported as, Cu^{2+} and MO both have maximum adsorption capacities of 103.5 and 265.9 mg/g, respectively [38]. The metal adsorption capability of specially manufactured nano-bentonite/Chitosan based aerogel (NCNB) from simulated wastewater has been studied. For Cu, Co, and Cr eradication, the highest adsorption efficiencies were found to be 98.90%, 97.45%, and 99.01%, respectively. It was also revealed that the best pH for maximum removal of Cr, Co, and Cu, was at 2, 4, and 5, respectively [39]. Using graft copolymerization and freeze-drying, cellulose nanocrystals (CNCs) attained from acid hydrolysis of discarded bamboo powder were employed in the creation of aerogels known as cellulose nanocrystal-g-poly (acrylic-acid-co-acrylamide), i.e. CNC-g-P(AA/AM) aerogels. Pb(II) maximum adsorption affinity determined by the Langmuir model was 366.3 mg/g, outperforming the pure CNC aerogel. After five successive adsorption–desorption cycles, the aerogels were noticed to have an 81.3% reduction efficiency [40]. Chen and team [41] created a novel approach for enhancing the anti-hydrolytic potential and mechanical strength of traditional chitosan aerogel (CSA) through bidirectional regeneration of microcrystalline cellulose and chitosan to generate chitosan-microcrystalline cellulose aerogel (CS-MCCA). To increase the rate of CSA adsorption, bentonite (BT) was also added to the regeneration process. This led to the development of the effective adsorbent bentonite-chitosan-microcrystalline cellulose aerogel (BT-CS-MCCA), with maximum Pb(II) adsorption amounts and adsorption equilibrium times of 256.24 mg/g and 60 min, respectively. A composite aerogel comprised of bacterial cellulose (BC) and poly(amidoxime) (PAO) was fabricated through a straightforward and scalable self-assembly method, along with an in-situ oxidation transformation for the purpose of heavy metals elimination. The produced PAO/BC composite aerogel has exceptional sorption capacities for heavy metals, including, and 382.3 mg/g for Cd^{2+}, 457.2 mg/g for Mn^{2+}, 494 mg/g for Zn^{2+}, 509.2 mg/g for Cu^{2+} and 571.5 mg/g for Pb^{2+} surpassing most noted nano-adsorbents [42]). Aerogel composed of porous cellulose nanofibers (CNFs) was created using the directional freeze-drying process. The CNFs aerogel's maximum adsorption capacity was 440.60 mg/g at a concentration of 5–50 mg/L. Since the static adsorption and Langmuir were in good agreement (R^2 - 0.97), it was determined that the adsorption was a homogenous, monolayer process [13].

2.4. Natural occurring soil and minerals-based adsorbents

Given both the economic and technological constraints typically encountered in developing nations, the use of soils and other natural materials for extracting heavy metals from water solutions can represent an efficacious and practical approach. Clay minerals have been found to be used to diminish heavy metal ion concentration. Of these, the most effective and widely used clay minerals for heavy metal sorption in water and soil are attapulgite, montmorillonite and bentonite. Evidence proves that these minerals are efficient in heavy metal sorption in wastewater, soil, water and sludge. These clay minerals confer the benefit of a great specific surface area, large expanding/swelling capability and great ion cation exchange capacity [43]. The sorption efficacy of heavy metals by natural occurring soil and mineral, and locally occurring biomass-based adsorbent, reported in Table 2. The surface adsorption mechanism of natural occurring soil and minerals-based adsorbent for heavy metals removal depicted in Figure 4(d). A study employed the clinoptilolite zeolite based adsorbent for the eradication of metals, i.e. Cr, Pb, Ni, Cu and Cd and obtained the maximum sorption and 5.44–5.79 mg/g, 5.53–5.93 mg/g, 5.92–6.07 mg/g, 6.11–6.13 and 6.21 mg/g [44], respectively. Montmorillonite (MMT), a form of clay, was synthesized at different doses (0.5, 1.0, and 1.5 wt%) to create polysulfone mixed matrix membranes for treating aqueous solutions polluted with Cd and Pb to investigate the effectiveness of heavy metals removal. The range of 1–42 m is used to measure the particle size distribution of MMT. The silica functional group is dominant in the elemental composition of MMT. Compared to the virgin membrane, uptake increased by 83%. The 1.5 MMM membrane has the fastest pure water flow of any membrane at 2500 L/m^2h and is the most porous membrane with a 10% porosity. Measurements of heavy metal eradication reveal that the functionalized membrane can eliminate Pb and Cd to corresponding degrees of 3% and 14%. Furthermore, a solute water flow of greater than 500 L/m^2h is seen in all produced

Table 2. Sorption performance of heavy metals by natural occurring soil and mineral, and locally occurring biomass-based adsorbent.

S.No	Adsorbent	Heavy metals	Reaction conditions	Adsorption /Removal	Reference
1	Clinoptilolite based zeolite-geopolymer	Cr (II), Pb (II), Cu (II), Cd (II) and Ni (II)	$[AD]_0$ = 2 g/L, C_0 = 50 mg/L (each), pH = 5, time = 24 h	5.53-6.21 mg/g,	[39]
2	Montmorillonite Clay	Cd (II)	Flux = 2500 L/m^2h, C_0 = 1 g/L,	3-14% removal	[40]
3	FeOOH-MnO_2/sepiolite/ diatomite	As(V)	$[AD]_0$ = 0.04 g/L, C_0 = 20-200 mg/L, pH = 7, temp = 25 °C, time = 5-90 min	124.3, 148.2 mg/g	[83]
4	Diatomite-chitosan composite	V (V)	$[AD]_0$ = 1.2 g/L, C_0 = 100 mg/L, pH = 5, time = 90 min, temp = 25 °C	96.5% removal	[41]
5	MnO(OH)-modified diatomite	Co, Eu, and Sr	$[AD]_0$ = 1 g/L, C_0 = 0.01-0.8 g/L, pH = 5.5, time = 40 min, temp = 25 °C	16, 28, 64 mg/g	[84]
6	Fly Ash Na-X Zeolite	Pb (II), Zn (II)	$[AD]_0$ = 3 g/L, C_0 = 100 mg/L, pH = 5, time = 10-200 min, temp = 25 °C	332.5, 640 mg/g	[85]
7	Zeolite, bentonite, and steel slag	Fe (II), Cu (II), Pb (II)	$[AD]_0$ = 0.4 g/L, C_0 = 97.7-111.1 mg/L, pH = 7, temp = 25 °C, time = 24 h	45.0 mg/g	[86]
8	Iranian clinoptilolite zeolite	Cd (II), Pb (II)	$[AD]_0$ = 1-10 g/L, C_0 = 0.78-2.18 mg/L, pH = 7, flow rate = 1-10 ml/min, temp = 25°C	85.9%, 98.9%	[87]
9	Mixed pillared clays	Cd (II), Co (II), Cu (II)	$[AD]_0$ = 0.1 g/L, C_0 = 10-100 mg/L, pH = 3-10, time = 24 h, temp = 25-55°C	24.8 mg/g	[88]
10	Bentonite, red earth and volcanic ash	Zn (II), Cu (II) and Ni (II)	$[AD]_0$ = 20-60 mg/L, C_0 = 1-80 mg/L, pH = 2-11, temp = 25-70 °C time = 0-90 min	Ni (II), Cu (II) = 100%, Zn (II) = 90%	[89]
11	Chitosan/diatomaceous earth	Pb (II), Ni (II)	$[AD]_0$ = 1 g/L, C_0 = 50-400 mg/L, pH = 2-8, time = 300 min	175.2, 149.6 mg/g,	[90]
12	Blast furnace slag and fly ash	Pb, Cu, Cd, Cr, Zn	$[AD]_0$ = 0.1-20 g/L, C_0 = 5 mg/L (each), pH = 6.5, temp = 25 °C, time = 24 h	4.3-5.2 mg/g	[91]
13	Zeolite-supported nZVI	Cd (II), Pb (II), As (III)	$[AD]_0$ = 0.5 g/L, C_0 = 50 mg/L (each), pH = 6, temp = 25 °C, time = 10 h	11.5, 48.6 mg/g, 85.3 mg/g	[36]
14	Sepiolite modified nZVI	As (III), Cd (II)	$[AD]_0$ = 1 g/L, C_0 = 5-600 mg/L, pH = 2-9, temp = 25-45 °C, time = 2 h	230.3, 11.37 mg/g	[92]
15	β-FeOOH modification of sepiolite	Cr (VI),	$[AD]_0$ = 400 mg/L, C_0 = 100 mM, pH = 5, time = 1 h	169 mg/g	[93]
16	Zeolite-supported nZVI	Cr (VI), Cd (II)	$[AD]_0$ = 1 g/L, Co = 20 (Cr), 200 (Cd) mg/L, temp = 25 °C, time = 500 min	77% (Cr), 99% (Cd) removal	[94]
Locally available biomass-based adsorbent					
17	Brewed tea waste	Pb (II), Zn (II), Ni (II), Cd (II)	$[AD]_0$ = 0.1-5 g/L, C_0 = 100 mg/L, pH = 2-6, time = 1-150 min, temp = 20 °C	1.19, 1.45, 1.16, 2.48 mg/g	[47]
18	Tobacco leaves coated with iron oxide	Cr (VI), Pb (II), Zn (II)	Flow rate = 5 ml/min, C_0 = 100 mg/L, pH = 2-7, temp = 25 ∘C	92.26%, 75.5%, 89.4%	[45]
19	Oyster shell	Pb (II)	$[AD]_0$ = 0.01-0.5 g/L, C_0 = 700 mg/L, pH = 3-6, time = 0-240 min, temp = 25 ∘C	640 mg/g	[95]
20	Modified cocoa shell adsorbent	Cr (VI), NO_3-	$[AD]_0$ = 4 g/L, C_0 = 50 mg/L, pH = 2-7, time = 3 h, temp = 20 °C	53.2, 31.6 mg/g	[96]
21	Olive stone waste bio -adsorbent	Fe (III)	$[AD]_0$ = 0.1-1 g/L, C_0 = 10-100 mg/L, pH = −8, time = 30-120 h, temp = 20-40 °C	99.5% removal	[97]
22	Chemically treated sheep wool	Au (III), Cu (II)	$[AD]_0$ = 10 mg/15 mL, C_0 = 10 mg/L, pH = 2-5, time = 48 h, temp = 30 °C	0.82, 0.95 mM/g,	[98]
23	Detox tea waste	Cr (VI)	$[AD]_0$ = 0.5-5 g/ 100 mL, C_0 = 1 g/L, pH = 2-7, time = 5-120 min, temp = 20 °C	9.16 mg/g	[99]
24	Oyster shells	Pb (II)	$[AD]_0$ = 1 g/L, C_0 = 0.6 g/L, pH = 2-9, time = 0-1 h, temp = 25 °C	499 mg/g,	[100]
25	Tobacco stem biochar	Pb (II), Cd (II), Cu(II)	$[AD]_0$ = 4 g/L, C_0 = 1 g/L, pH = 2-5.8, time = 12 h, temp = 25-45 ∘C	Cu(II)>Cd(II) >Pb(II)	[101]

[AD] – Adsorbent dose; C_0 – Metal concentration.

membranes. The polysulfone mixed matrix membrane has been effectively modified by MMT inclusion to produce a high-flux membrane with the capacity to treat cadmium and lead-rich water [45]. An in-situ self-assembly technique was used to create a diatomite-chitosan composite (CS-DE-10%) that effectively removed vanadium (V) from wastewater. While diatomite and chitosan alone had a lower removal effectiveness (96.5%) for vanadium, the CS-DE- 10% composite efficiently reduced vanadium toxicity by reducing V (V) to V (IV) since its functional groups offer active atoms. The increased specific surface area and pore volume of the CS-DE-10% adsorbent, which increased by ∼9 and 4 times in comparison to pure chitosan, may have contributed to the better adsorption capacity of the substance. The composite's functional groups OAH, C@O, CAO, NAC@O, and ANH_2 also added to the improved reduction capability [46]. In another study, mixed layer clay was used for Ni removal from wastewater. Herein, the wastewater saw a significant reduction in Ni ions, reaching as high as 75%. Additionally, it was discovered that elevating the pH levels leads to a heightened adsorption rate [47]. Montmorillonite, illite and

kaolinite was employed to eradicate La and Yb from water. The data revealed that montmorillonite was the most effective in heavy metal removal out of the three adsorbents at a pH of 4.5 at room temperature [48]. Zeolite may effectively treat Pb (II), Zn (II), and Ni (II)-polluted soils, as exhibited by Wahba et al. [49]. By modifying montmorillonite with H_2SO_4, Akpomie and Dawodu were able to eliminate the metal species Ni, Pb, Zn, Mn, Cu, Cd, Cu, Mn, Zn, Pb and Ni from automotive effluents. A 50 mL solution with a concentration of heavy metal ions ranging from 0.5 to 2.5 M and a pH range of 2–8 was treated with exactly one gram of the modified clay. The solution was stirred for approximately 180 min at a temperature of 27 °C. Montmorillonite surface area increased from 55.8 to 96.5 m^2/g. The pH 8 produced the greatest adsorption rates of, 14.7% Ni, 12.6% Pb, 11.3% Cd, 12.7% Mn, 10.1% Cu and 18.1% Zn, They came to the conclusion that acid alteration of clay minerals improves the ability of heavy metals adsorption [50]. Heavy metal elimination via soil and other natural minerals/remediation approaches has been the subject of several in-depth reviews. Ongoing exploration in this area is anticipated, particularly since developing countries seek more cost-effective solutions to combat heavy metal pollution. This is driven by the increasing interest in the utilization of abundant, readily accessible soils, minerals, and other organic materials for heavy metal eradication.

2.5. Locally available waste materials-based adsorbents

The efficiency of locally accessible waste material in removing heavy metals from aqueous solutions has been extensively studied as reported in Table 2. In the developing world, different forms of garbage may be created more frequently than others as a result of regional variations in ecosystems, energy sources, farming methods, and cultural behaviours. For example, a study detailed the utilization of tobacco leaves coated with iron oxides in a continuous fixed bed column for adsorbing heavy metals, notably zinc (II), lead (II) and chromium (VI) ions, from industrial wastewater. Under optimal conditions, which included a bed height of 10 cm, flow rate of 5 mL/min and a heavy metal concentration of 100 mg/L, the highest reported adsorption efficiencies were 92.26% for Cr (VI), 75.57% for Pb (II), and 89.36% for Zn (II). Various kinetic models, such as the Thomas, Yoon-Nelson, and bed depth service time models, were applied, and they exhibited strong correlations ($R^2 > 0.95$) [51].

Residual waste materials from vegetable waste may be able to eliminate heavy metals from water sources. This has been exemplified in the work of Li and their research team, where they provided evidence of the effectiveness of mushroom residues in heavy metals eradication. Removal efficiency for Hg (II), Zn(II) and Cu(II) ranged from 39.7% to 81.7% based on an analysis of four distinct kinds of mushroom residues [52]. In another study, authors Zhang et al., synthesized the pectin based composite hydrogel derived from grapefruit peel for the improvement of metal ions sorption, i.e. Mn, Ni, Zn, Cd, Cu, Pb and Fe. Under the optimum operating conditions (e.g. adsorbent dose 1 g/L, time 24 h, pH 6, C_0 50 mg/L, and temp 30 °C) the decreasing order of metals ions adsorption was found Fe > Pb > Cu > Cd > Zn > Ni > Mn [53]. Pb, Zn, Ni and Cd removal efficiencies were investigated utilizing brewed tea trash as a possibly suitable adsorbent, from a simulated watery solution. Throughout the adsorption procedure, the effects of pH levels (2.0–6.0), adsorbent dose (0.1–5.0 g/L), and contact periods (1–150 min.) were studied. The experiment's findings revealed a linear link between the other parameters and an inverse association between pH and the yields of Cd, Ni, Zn and Pb removal. The optimal pH range for removing heavy metals using brewed tea waste was found to be between 4.0 and 5.0. Achieving equilibrium in the adsorption of Cd, Ni, Zn and Pb onto Camellia sinensis took approximately 5, 30, 10 and 2 min, respectively. The adsorbent exhibited a significant capacity to decrease heavy metals from aqueous solutions. Upon application of the Langmuir isotherm model to the equilibrium data, the highest adsorption capacities for Cd, Ni, Zn and Pb were calculated to be and 2.468, 1.163, 1.457, and 1.197 mg/g, respectively [54]. Fly Ash (FA) is a strong and cost-effective adsorbent for dyes and heavy metal elimination as a result of its morphology, charge and surface structure [55]. FA primarily comprises iron oxide, silicon, alumina, and residual carbon, which are favoured by heavy metals as adsorption sites and active centres in the process of dye adsorption. In the context of industrial wastewater, an economical sorbent called

zeolite Linde type A (LTA) was derived from coal fly ash (CFA-ZA) and utilized for Hg(II) eradication. This approach yielded an average removal efficiency of 94% for Hg(II) using CFA-ZA, with the sorption process most accurately presented by the pseudo-second-order (PSO) model and FIMs. The measured sorption capacity was 0.44 mg/g, and it was evident that both physisorption and chemisorption mechanisms were involved in the adsorption of Hg(II) ions onto the surface of CFA-ZA [56]. The microwave-assisted hydrothermal process was utilized to modify municipal solid waste incineration (MSWI) FA to ascertain zeolite to treat heavy metal contaminated wastewater (Ni, Cd, Zn, Pb, Mn, Cd and Cu). The PSO model and LIMs best fit the sorption processes, and 2.0 was the optimum pH for the heavy metals' reduction. The sorption capacities were 27.55 mg/g for Cu, 10.58 mg/g for Cd, 13.61 mg/g for Ni, 50.00 mg/g for Pb, 15.46 mg/g for Mn, and 20.16 mg/g for Zn [57].

2.6. Adsorbent derived from agricultural biomass

The exploration of altered agricultural waste for heavy metal removal has garnered substantial attention from researchers in both developed and developing nations. In the context of developing countries, the utilization of agricultural waste often provides an ample and efficient source of adsorbents, effectively utilized for the elimination of varied contaminants from wastewater [58]. Various authors have synthesized the bio-adsorbent employing the agriculture biomass waste (including, vegetable peel, food waste, and another domestic biomass) and obtained remarkable efficacy of heavy metal sorption as summarized in Table 3. The heat treatment technique is a frequently used technique for the preparation of bio-adsorbent since it generates a high surface area available for adsorbate sorption. The schematic diagram of bio-adsorbent synthesized by heat treatment method depicted in Figure 3(b). A study investigated HNO_3 oxidation to alter sugarcane bagasse-derived activated carbon (AC) to determine whether this improved the material's ability to remove Pb. Nitric acid modification enhanced the oxygen-containing functional groups on the adsorbent surfaces, including hydroxyl, carboxyl, carbonyl, and ester. Surface functional groups had an impact on the samples' ability to adsorb Pb(II) ions for removal. The Langmuir model estimated a maximum Pb(II) adsorption capability of ~ 212.31 mg/g. The pseudo-second-order model suited well for all of the samples [59]. The another authors Moubarik and Grimi, tested the adsorption capacity of Cd(II) on sugar cane bagasse based adsorbent and obtained more than 96% removal of Cd(II) under the conditions (i.e. adsorbent dose 0.5 g/L, $[Cd(II)]_0$ 10–30 mg/L, pH 2-11, time 30–80 min, and temp 25–90°C) [60]. An adsorption study was conducted on, sunflower seed hulls, sawdust from pine and residues of corn mixed in water as adsorbents, using synthetic aqueous solutions of Cd(II), Ni(II) and Zn(II) as target metals. Results of this study show that all biomasses are excellent substitutes to synthetic materials, with adsorbent efficacies exceeding 50%. The maximum removal of metals Ni(II), Zn(II), and Cd(II) were observed 71%, 89.2% and 96.6%, under operating conditions, i.e. adsorbent dose 1 g/L, metal concentration 0.18 mmol/L, pH 4–5 and reaction time 24 h [61]. There have been relatively few studies that have proven the efficacy of modified agricultural wastes in removing various pollutants from real-world environmental waters, such as industrial wastewater and municipal wastewater [62]. More than 60 bio-adsorbents are available from agricultural products with best-performing conditions for heavy metal adsorption of dyes, pharmaceuticals, nutrients, and other contaminants. Bio-wastes manufacturing methodologies and protocols must be improved to enhance the adsorption process and enable the design of bio-adsorbents that are more cost-effective, and that can utilize multiple waste substances Aguilar-Rosero et al. [63]. For example, activated carbon (CAC) was manufactured through controlled carbonization and chemical activation of four wastes (such as banana peels, corn cobs, coffee husks and peanut shells). In comparison to the single-biomass-derived activated carbon, the CAC produced at optimal conditions demonstrated 96.2% CO_2 removal efficiency and 8.86 wt% adsorption capacity [64]. A magnetic nanocomposite was synthesized by co-precipitation using peanut hulls, iron (II) sulphate, and iron (III) chloride, an agricultural waste. Under constant ambient parameters, temperature of 25°C and a 200 mL reactor volume, an 82.8% removal efficiency is obtained from experimental studies [65]. The other adsorbent material like hydrogel beads (BPA) made from grapefruit peel, incorporating biochar, pectin, and alginate, were developed

Table 3. Sorption performance of heavy metals media by agriculture, aquatic and terrestrial based adsorbent.

S.No	Adsorbent	Heavy metals	Reaction conditions	Adsorption /Removal	Reference
1	Sugarcane bagasse-based	Pb(II)	$[AD]_0$ = 0.1-0.5 g/L, C_0 = 400 mg/L(Pb), pH = 3-9, time = 2 h	212.3 mg/g	[52]
2	Pine sawdust, sunflower seed	Ni(II), Zn(II) and Cd(II)	$[AD]_0$ = 1 g/L, C_0 = 0.18 mmol/L, pH = 4-5, time = 24 h	RE_{Ni} = 71%, RE_{Zn} = 89.2%, RE_{Cd} = 96.6%	[54]
3	Melon peel	Cu(II), Pb(II) and Cd(II)	$[AD]_0$ = 0.3-3 g/L, C_0 = 10 mg/L (each), pH = 3-9, time = 1 h	77.7 mg/g - Cu(II), 191.9 mg/g -Pb(II), 76.1 mg/g - Pb(II),	[102]
4	Pomelo Peel	Pb(II) and Cu(II)	$[AD]_0$ = 0.25-3 g/L, C_0 = 1.59 g/L(Pb), 3.96 g/L(Cu), pH = 3-9, time = 2 h	205.3 mg/g – Pb (II), 81.9 mg/g - Cu (II),	[103]
5	Grapefruit peel	Cu(II)	$[AD]_0$ = 1 g/L, C_0 = 50 mg/L(Cu), pH = 2-7, time = 24 h, temp = 20 °C	80.6 mg/g - Cu(II),	[46]
6	Peanut hull	Ni^{2+}	$[AD]_0$ = 0.2-0.8 g/L, C_0 = 0-100 mg/L, pH = 2-9, time = 70, temp = 30-50 °C	RE_{Ni} = 82.8%,	[56]
7	Tea waste	Pb^{2+}/ Cd^{2+}	$[AD]_0$ = 2-11 g/L, C_0 = 1 g/L (each), pH = 2-8, time = 1 h, temp = 25 °C	153 / 222 mg/g	[104]
8	Black gram husk	Cr(VI)	$[AD]_0$ = 50–500 mg/L, C_0 = 50-500 mg/L, pH = 1-9, Temp = 20-45 °C, Time = 3 h	215.27 mg/g	[105]
9	Coffee waste	Pb^{2+}	$[AD]_0$ = 0–100 mg/L Co = 25 mg/L, D = 0.010 mg CT = 30 min pH = 7.3 T = 25 °C	41.15 mg/g	[106]
10	Peanut husk	Ni^{2+}	$[AD]_0$ = 0.3–0.7 mg/L Co = 0.3 mg/L, D = 2.5 g/L pH = 6.5	0.1473 mg/g	[64]
11	Corn cob+ petai hull	Pb^{2+}	Co = 400 mg/L initial concentration = 200–500 mg/L	2.230 mg/g	[63]
12	Punica granatum L. peel	Pb^{2+}	$[AD]_0$ = 1000 mg/L, C_0 = 145-200 mg/L, pH = 5.5, Temp = 20-50 °C, Time = 2 h	371.36 mg/g	[107]
13	Banana peel	Mn^{2+}	$[AD]_0$ = 1-6 g/L, C_0 = 100-600 mg/L, pH = 5.5, time = 2 h	5.7306 mg/g	[57]
14	Coconut pith	Ni^{2+}	$[AD]_0$ = 0.25-3 g/L, C_0 = 10-60 mg/L, pH = 2-10, time = 3 h	24.39 mg/g	[108]
15	Coconut-shell biochar	Cd (II)/ Pb (II)	$[AD]_0$ = 1 g/L, C_0 = 100-200 mg/L, pH = 2-8, time = 6 h	more than 90% removal	[109]
16	Agriculture biomass	Pb (II)	$[AD]_0$ = 100 mg/L, C_0 = 100-200 mg/L, pH = 1-7, time = 6 h, T = 25 °C	87.0 mg/g	
17	Papaya leaves, Petai peel	Pb^{2+}	$[AD]_0$ = 100 mg/L, C_0 = 0.59-5.4 g/L, pH = 3-8, time = 2 h	284.35, 36.01 mg/g	[110]
18	Sugar cane bagasse	Cd (II)	$[AD]_0$ = 0.5 g/L, C_0 = 10-30 mg/L, pH = 2-11, time = 30-80 min, temp = 25-90 ∘C	96% removal	[53]
Aquatic and terrestrial based adsorbent					
19	Cassia fistula seed carbon	Cd (II)	$[AD]_0$ = 50–250 mg/0.05 L, C_0 = 10–50 mg/L, pH = 2-10, time = 1.5 h, temp = 30 °C,	68.04 mg/g	[59]
20	Spent mushroom substrates	Cu (II)	$[AD]_0$ = 0.5-4 g/L, C_0 = 25-150 mg/L, pH = 2-6, time = 6 h, temp = 15-35 °C,	65.6 mg/g	[111]
21	Capparis decidua and Ziziphus	Cd (II)	$[AD]_0$ = 1 g/L, C_0 = 249 mg/L, pH = 6, time = 1 h, temp = 15-35 °C,	248.62 mg/g	[60]
22	Alcaligenes faecalis K2	Cd (II)	$[AD]_0$ = 1 g/L, C_0 = 0-125 mg/L, pH = 6-9, time = 0-216 h, temp = 30 °C,	89.5% removal	[61]
23	Cassia fistula seeds	Pb (II)	$[AD]_0$ = 0.10–12 g/L, C_0 = 25-200 mg/L, pH = 2-8, time = 10-60 min, temp = 30-60 °C	129.3 mg/g	[112]
24	Hyacinth roots	Cr (VI)	$[AD]_0$ = 14 g/L, C_0 = 10 mg/L, pH = 3, time = 2 h, temp = 25 °C	1.28 mg/g	[62]
25	Tannery wastewater	Cr (VI)	$[AD]_0$ = 6 g/L, C_0 = 0.45 g/L, pH = 3.9, time = 4 h, temp = 30 °C	97% removal	[113]
26	Pinus halepensis	Cu (II), Pb (II)	$[AD]_0$ = 10 g/L, C_0 = 1-50 mg/L, pH = 1-12, temp = 25 °C	9 mg/g	[114]
27	Four different algae	Hg (II), Cu (II), Zn (II),	$[AD]_0$ = 1 g/L, C_0 = 10 mg/L (each), pH = 6, time = 2 h, temp = 25 °C,	53.8–84.1% removal	[52]

[AD] – Adsorbent dose; C_0 – Metal concentration

and employed for the extraction of Cu(II) from a water-based solution. The Freundlich model provided an excellent fit for the adsorption isotherms, and the optimized BPA-9 beads (with a pectin to alginate mass ratio of 10:1 and 0.25% biochar) exhibited an experimental maximum adsorption capacity of approximately 80.6 mg/g at a pH of 6. Interestingly, the BPA beads exhibited effective regeneration capabilities even after five consecutive cycles [46]. Authors Chen et al., synthesized the novel magnetic materials derived from pomelo peel bio-char (MPPB) and effectively utilized for Cu(II) and Pb(II) adsorption from aqueous media. Adsorbent MPPB exhibited the remarkable adsorption capacity 205 mg/g for Pb(II) and 82 mg/g for Cu(II), and best suited by the Langmuir isotherm and pseudo second order kinetic model [40]. The biomass based on banana peels consists of different chemical components such as

hydroxyl groups, phosphate and carboxylic acid which as active sites for heavy ions adsorption from wastewater. Authors Ali synthesized the adsorbent by chemically modified banana peels for Mn(II) reduction from water. The maximum elimination of Mn(II) by grafted banana peels was noticed to be 94%, and the adsorption equilibrium was best suited by the Langmuir isotherm model [57].

2.7. Aquatic and terrestrial biomass-based adsorbent

The most widely used adsorbent materials often encompasses modified carbon synthesized from using zeolites, algal species, plant roots, biomaterials and other materials derived from aquatic and terrestrial biomass-based as reported in Table 3. Yet, because of the expensive nature and specific complexities linked to their amalgamation, there is a rising trend towards the use of minimal effort aquatic and terrestrial-based biomass that can be sufficiently utilized for inorganic contaminant and elimination of heavy metal ions from wastewater [58]. For example, lignocellulose waste biomass cassia fistula seed carbon (CFSC) was investigated and was able to remove Cd (II) ions from saturated liquid samples. Within 80 min of treatment, an effective removal of around 93.2% (w/v) of the Cd (II) ions from a concentration of 10 mg/L was accomplished. The 100 mg/50 mL CFSC dose was determined to be ideal for increased Cd (II) elimination. Apart from that, the highest adsorption of Cd (II) onto CFSC was noticed at pH 6. With three isotherm models, Freundlich, Langmuir, and Dubinin-Radushkevich, investigational trials were evaluated. The measurements produced in this experimental investigation are in good agreement with the Langmuir isotherm model, which predicts a maximum adsorption capability of 68.02 mg/g [59]. To address the elimination of Cd(II) ions from wastewater, researchers examined powdered adsorbents derived from the leaves and branches of Ziziphus mauritiana (ZML) and Capparis decidua (CDB). The impacts of several operational parameters including adsorbent dose,, initial Cd(II) concentration temperature, pH and contact duration, were assessed. When the ideal contact duration, adsorbent dose, pH, and Cd(II) concentration were 6.0, 0.1 g/100 mL, 60 min, and 249 mg/L, respectively, the greatest sorption capability of CDB and ZML for Cd(II) ions at 25°C was 248.62 and 235.65 mg/g, respectively. After six cycles, the produced adsorbents showed good regeneration potential [60]. In order to decontaminate the Cd(II) containing wastewater, an economically viable approaches involved the utilization of screened Alcaligenes faecalis K2 to facilitate the bio mineralization and extraction of Cd(II) from wastewater. The maximum removal of Cd(II) 85.5% were noticed with an initial concentration 75 mg/L [61]. In a study reported by Kumar and Chauhan chemically modified the dried water hyacinth roots (DWHR) based adsorbent and used to remove the Cr (VI). The adsorption batch experiments were executed and obtained the maximum Cr (VI) removal efficiency 95.43% under the optimum operating conditions (i.e. 14 g/L) DWHR dose (size - 150 µm), $[Cr (VI)]_0$ 10 mg/L, agitation speed 200 rpm, temperature 25 ± 5°C, and contact time 2 h [62]. A study by Putri et al., examines the processes of making new biosorbents as well as the absorption of these biosorbents from the the petai hull of Petai hassk and corn cobs of Zea mays L. in order to dissolve lead metal waste (Pb). With a concentration of 400 mg/L, biosorbents H (HNO_3) and D (simplicia) demonstrated maximum adsorption capacity towards Pb (II) at 2.230 and 1.4175 mg/g, respectively [63]. A study was conducted on the biosorption of Ni(II) on modified and natural peanut husks. A sorbent dosage of 2.5 g/L and a pH of the solution 6.5 were found to be effective for the maximum removal of substances from natural peanut husks [64].

3. Current status, challenges and future outlook

Owing to their strong affinity to metal ions, adsorbents made from CNTs, graphene, bio-waste, cellulosic, aquatic and terrestrial biomass, have become increasingly popular as environmentally-friendly and cost-effective materials for heavy metals elimination in recent decades. However, it is important to acknowledge the presence of several challenges and limitations that must be tackled in order to fully harness the remarkable attributes of these adsorbents for actual wastewater treatment. The complexity and variability of environmental systems present a broad spectrum of research opportunities in the future, since there remain numerous issues that require direct resolution.

- Most of the literature has focused on treatments for lab-based wastewater containing a single pollutant, rather than real industrial wastewater. Research should prioritize considering that real wastewater contains various toxic heavy metals in varying amounts. This consideration is vital to evaluate their viability for practical purposes. In the upcoming years, addressing challenges such as economic constraints, revamping bio-adsorbents, and minimizing chemical use is critical as we transition from lab-scale research to commercial adsorption processes.
- Graphitic carbon nitride (g-C_3N_4) and graphene-based adsorbents have not yet achieved realistic applications in wastewater treatment due to the expensive raw materials they require for synthesis. Nevertheless, certain surface modifications are needed in the aforementioned to enhance their efficacy and make them economically viable for heavy metals removal from wastewater.
- Several cellulose-based hydrogels experience a reduction in their adsorption capabilities after regeneration process. Certain studies have indicated that these hydrogels maintain their adsorption capabilities only when regenerated using caustic soda. Additionally, making physical and chemical modifications to cellulose, along with various pretreatment methods, can somewhat improve the adsorption capacity of these cellulose-based hydrogels.
- Currently, the majority of research focuses on using conventional and novel adsorbents in small-scale batches to remove heavy metals from contaminated wastewater. Consequently, it's crucial to investigate whether these affordable bio-adsorbents can be viably employed on a larger commercial scale. The existing methods primarily aim to produce a limited amount of adsorbents for laboratory use in batch applications. These techniques can also be improved by incorporating eco-friendly strategies to minimize the excessive utilization of chemicals and energy, thereby avoiding the need for chemical alterations and further processing.
- The possible harmful environmental impacts of these novel and conventional adsorbents should be thoroughly assessed before they are used in commercial applications. Currently, there is insufficient data about the potential toxicity of adsorbents that have already been employed, as they have adsorbed toxic metal ions onto their surfaces. Consequently, it is imperative to conduct additional research in this area to determine the suitability of these adsorbents for actual water treatment applications. This research is essential to ensure the safety of human health and the preservation of the environment.
- At present, novel adsorbents derived from cellulosic, agriculture and aquatic waste are extensively employed in the elimination of heavy metals. To attain the highest levels of adsorption capacity and selectivity, it is crucial to carefully choose and recognize a suitable biomass residue as an adsorbent that can effectively attract and bind with the target adsorbate. This approach not only ensures the production of cost-effective bio-adsorbents but also contributes to reducing negative environmental consequences such as pollution whilst potentially increasing the adsorption capability.
- Future research should prioritize addressing challenges such as the inadequate elimination of heavy metal contaminants, elevated operational and maintenance expenses, increased energy demands, reduced effectiveness, and the regeneration of novel and conventional adsorbents for subsequent treatment.

4. Conclusions

In this up-to-date review, we provide an overview of conventional and novel adsorbents to boost the sorption of heavy metals. Herein, the focus centered around bridging the gap between cost-effectiveness in the context of the developing world, concentrating on locally available and inexpensive materials that can generate adsorbents in developing countries without compromising removal efficiency. Therefore, the primary emphasis is on emerging potential adsorbents, such as graphene, g-C_3N_4, minerals and clay-based material, and agriculture, aquatic and terrestrial based materials. In comparison to other traditional adsorbents, the application of bio-sorbents in heavy metals eradication from contaminated water has demonstrated superior economic and environmental advantages. Graphene and CNTs-based adsorbents such as Tl/GO@Fe_3O_4 (461 mg/g for Pb), and Fe_3O_4-GO-(MWCNTs) (Cu and Cd, 574–625 mg/g) exhibited the highest adsorption capacity for metal ions adsorption. Other conventional material such as natural

occurring soil and mineral, and locally occurring biomass-based adsorbent reported moderate adsorption capacity for heavy metals removal. Biomass material such as agriculture, aquatic and terrestrial based adsorbent reported relatively less adsorption capacity for heavy metals removal. Factors including surface charge, surface chemistry, the surfactant's pore properties, functional group type, and the altered chemical structure influence heavy metal sorption by CNTs. Additionally, experimental variables including pH, temperature and adsorbate/adsorbent concentration have an impact on the effectiveness of metal adsorption. The majority of the research shown that functionalization has consistently increased CNTs' ability to adsorb metal ions as compared to unfunctionalized CNTs. To modify the surface functionalization of carbon-based adsorbents, metal oxides such Fe_3O_4 are also utilized. However, determining the most suitable metal oxide modifier for heavy metal elimination is highly challenging because of variable conditions, diverse experimental setup, and variations in the source. Fe_3O_4 has, however, been demonstrated to have benefits in terms of recovery following the application of the adsorbent via magnetic separation as in the case of magnetic graphene oxide (Fe_3O_4-GO)(98% removal of Pb (II)).

Disclosure statement

No potential conflict of interest was reported by the author(s).

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