

Frontiers in Water and Environment



Journal homepage: www.akademiabaru.com/submit/index.php/fwe ISSN: 2785-9029

Recent Developments of Nanocomposite Adsorbents for Removal of Heavy Metals from Water: A Review

Asma Siddiqa¹, M.G. Mostafa^{1,*}

¹ Water Research Lab, Institute of Environmental Science University of Rajshahi, Rajshahi 6205, Bangladesh

ARTICLE INFO	ABSTRACT
Article history: Received 2 January 2025 Received in revised form 3 February 2025 Accepted 3 March 2025 Available online 10 March 2025	Nanotechnology is gaining attention in water treatment process due to its high efficiency and large surface area. This review aimed to study the recently developed nanocomposite adsorbents for removing heavy metals from water. The majority of the nanocomposites achieved maximum adsorption capacity with a moderate pH (5-9), a minimum adsorbent dosage (0.1-2 g/L), and 20-60 min for achieving equilibrium. The study showed that the physisorption occurred at a low mean free energy in contrast to chemisorption, and most of the adsorption processes followed the <i>pseudo</i> -2nd-order kinetics reaction. A comparative analysis of the nanocomposites revealed the selectivity of iron oxide nanocomposites towards the removal of Pb(II) ions;
<i>Keywords:</i> Adsorbent; heavy metals; nanocomposites; removal efficiency; recent development; water treatment	polythiophene-modified chitosan/magnetite nanocomposites could remove 100% of Hg(II) ions. The nanocomposite adsorbents face challenges in large-scale production, regeneration, and economic suitability. The study provides valuable insights for future research on the green synthesis of nanocomposite adsorbents for heavy metal removal processes.

1. Introduction

An increasing number of urbanizations and the changing global environment are forcing the already water-stressed human civilization toward a future where most people will have limited access to fresh water [1]. Various industrial advancements, such as mining operations, energy plants, and climate-related environmental catastrophes, have contributed significantly to the spread of toxic pollutants in aquatic environments. The ecology is adversely affected by pollution in these water bodies, which is also causing severe problems in drinkable water [2]. One of the most harmful water contaminants are heavy metals that harm humans and living things. Heavy metals that are effective at generating water pollution are released, misused, or overused, endangering human health and the ecological balance [3.4].

Drinking water contaminated with heavy metals can negatively impact human health, which can cause harm to the respiratory, circulatory, and renal systems, as well as malformation in developing fetuses, necessitating careful concentration management and effective elimination of less toxic

* Corresponding author. Email address: mgmostafa@ru.ac.bd

https://doi.org/10.37934/fwe.6.1.112

materials [5,6]. Thus, the need of the moment is for a dependable, cost-effective solution that can handle a variety of hazardous contaminants. Many processes, including distillation, reverse osmosis, filtration, ion exchange, coagulation-flocculation, solvent extraction, and adsorption by nanoscale material, have been described as efficient strategies for removing contaminants from wastewater [7,8]. Even though these technologies can remove various pollutants altogether, they usually have drawbacks, including high costs and difficulty.

Since adsorption is a safe, clean, efficient, and technically possible process, considered as most efficient techniques for removing heavy metals from wastewater [9]. Additionally, while utilizing this method, the choice of adsorbent depends on several factors, such as its cost, availability, toxicity, and regeneration capacity [10,11]. Most standard approaches have recently been combined with nanotechnology to accelerate the removal efficiency of heavy metals from water bodies [12]. Wastewater contaminants can be effectively removed using adsorbent-based nanoscale materials [11]. Minerals, activated carbon, agricultural waste, metal oxide nanoparticles, nanocomposites, mixtures of nanoparticles, or nanoparticles attached to more extensive bulk materials, etc., are commonly used as adsorbents due to their simplicity of use, convenience, low cost, and high efficiency concerning the removal of metal ions [13,14]. To create nanomaterials with novel physical and chemical properties for future technological applications, it is necessary to preserve the dispersion and agglomeration of the nanoparticles while manufacturing nanocomposites [15]. Several reviews have recently been published describing the synthesis methods and the effectiveness of different nanocomposite adsorbents. However, to the best of our knowledge, a comprehensive review covering the selectivity of nanocomposite adsorbents towards the removal of specific heavy metals by exploring their optimized adsorption conditions has not been published in one manuscript.

This review examines the latest developments in nanocomposite adsorbents and focuses on the best conditions for maximum adsorption. It also examines their isotherm, kinetics, and thermodynamics. It also compares nanocomposites' selectivity for removing specific heavy metals and provides a guideline for the future use of eco-friendly, cost-effective nanocomposites, such as those made from agricultural waste or minerals, in the water treatment process.

2. Materials and Methods

This review collected data from several published articles using search engines PubMed, Scopus, Google Scholar, Web of Science, and other databases for publications across the last six (06) years, from 2018 to 2023. The following terms were searched using the Boolean operators ('and' and 'or') to search for relevant research publications using the following terms: nanocomposites, heavy metals, water treatment, and adsorption. Initially, the review identified over 300 published papers but finally included 70 directly related to and/or supporting this study title. Here, the study followed the PRISMA Statement for every event of inclusion and exclusion criteria [16]. Figure 1 shows the rate of journal publications. The review explored the recent development of nanocomposite adsorbents. It gave a comparative study of the selectivity of nanocomposites towards the removal of specific heavy metals and provided a guideline for future studies.

3. Adsorption Parameters of Different Nanocomposite Adsorbents

There are numerous factors that are considered as adsorption parameters, such as types of heavy metal removed, pH, adsorbent dosage, initial concentration, equilibrium time, and reusability of the nanocomposite adsorbents. Table 1 summarizes the different adsorption parameters of recently

developed nanocomposite adsorbents. The removal of Cd(II) was found to have a maximum adsorption capacity of 14.3 mg/g at a pH of 5 reported by Alnasrawi *et al.*, [17]. Another study showed the removal of Pb(II) and Cd(II) ions using multifunctional cobalt oxide nanocomposites. It showed an increased removal from 20.3% to 83.07% for Pb(II) and from 22.41% to 81.04% for Cd(II) occurred by increasing pH from 2.5 to 8.5, respectively observed by El Mouden *et al.*, [18]. The Cu(II), Pb(II), Cd(II), and Zn(II) ions showed maximum adsorption at pH (5.5 ± 0.5) and gradually decreased adsorption capacity with an increase in pH value observed by Nyirenda *et al.*, [19]. A study showed that Cr(VI) adsorption was conducted in a pH range of 4–9 [20]. The amount of Cr(VI) adsorbed depends on the distribution of $Cr_2O_7^{2^-}$, HCrO₄⁻, and CrO₄^{2^-}, which are controlled by the pH of the solution. The surface of the adsorbent becomes more protonated when the pH is lowered, and the positively charged adsorbent's surface is strongly attracted to these oxyanions of Cr(VI). Hence, the removal efficiency increased with increasing the pH from 4.0 to 6.0, and maximum adsorption showed at a pH of 5 reported by Houng *et al.*, [20].



Fig. 1. Flow diagram of PRISMA on the identification of eligible investigations to final inclusion [16]

The adsorbent dose strongly influences the removal of heavy metals. The heavy metal removal efficiency increases with the increase of the adsorbent dosage [21]. The removal efficiency declined until it reached a saturation value. It is possible that after reaching adsorption equilibrium, some adsorbate might detach from the adsorbent [17].

The Cd²⁺ removal efficiency increases from 38.4% to 77.9% when the adsorbent dose is raised from 0.05 to 0.2 g, and 0.2 g was considered the optimum dosage was observed by Alnasrawi *et al.*, [17]. Since the availability of active adsorption sites was saturated, it caused decreased adsorption capacity [19]. The removal of Cu²⁺, Pb²⁺, Cd²⁺, and Zn²⁺ ions at different adsorbent dosages in the range of 0.1–0.4 g increased the removal percentage of metal ions with an increase in adsorbent

Table 1

dosage [19]. The presence of additional active adsorption sites on the adsorbent's surface explained the increase in percentage removal as the adsorbent dosage increased.

Adsorption parame	eters of t	the nand	ocomposit	e adsorbe	ents			
Nanocomposite adsorbent	Heav y metal s	pH value	Dosage (g/L)	Specific surface area (m ² /g)	Equilibrium time (min)	Initial Concentration (mg/L)	Re- usability	References
Multifunctional cobalt oxide nanocomposite	Pb(II) Cd(II)	8.5	2.8	-	20	50	5 cycles	[18]
Polythiophene modified chitosan/magnetite nanocomposite	Hg(II)	7	2	-	60	100	-	[23]
CuMgAl-layered double hydroxide/montmo rillonite nanocomposite	Cd(II)	5	0.2	77.6	30	50	5 cycles	[17]
Activated carbon- supported silver- silica nanocomposite	Pb(II) Cd(II) Cu(II)	5.5	0.4	-	40	10	2 cycles	[19]
Graphene oxide/polyvinyl alcohol/Fe ₃ O ₄ nanocomposite	Co(II)	5.2	0.5	-	100	100	5 cycles	[24]
Iron oxide-tea waste nanocomposite	Pb(II)	7	0.1	171	4	100	-	[25]
Chitosan/dopamine /Fe ₃ O ₄ nanocomposite	Pb(II) Hg(II) Cu(II)	7	0.1	-	30	50	6 cycles	[21]
Hydroxyapatite/Fe₃ O₄ nanocomposite	Pb(II)	6	1.0	-	50	10	-	[26]
Fe ₃ O ₄ /Talc nanocomposite	Cr(VI)	5	2	77.92	60	10	-	[20]
Magnesium oxide/bentonite nanocomposite	Cd(II)	9	0.2	-	10	40	6 cycles	[27]
Polypyrrole - iron oxide seaweed (PPy-Fe₃O₄-SW) nanocomposite	Pb(II)	5	0.1	-	20	30	5 cycles	[28]
Mesoporous Silica- Graphene Oxide nanocomposite	Pb(II) As(III)	6.8	0.1	858	10	10	4 cycles	[29]

The high surface area of nanocomposite adsorbent favors high adsorption capacity. The adsorbent CuMgAl-LDH/MMt has higher catalytic and adsorption activity for removing Cd(II) ions and a higher surface area of 77.6 m²/g reported by Alnasrawi *et al.*, [17]. Here, the enhanced surface area of iron oxide-tea waste nanocomposite (171 m²/g) showed higher removal efficiency of Pb(II) ions

observed by Khanna *et al.*, [25]. Mesoporous silica-graphene oxide nanocomposite gave maximum removal capacity for the removal of Pb(II) and As(III) for having a higher surface area of 858 m^2/g .

The study observed that most heavy metal ion adsorption occurs in a contact time of 20 min and gradually reaches near equilibrium in 140 min. Adsorption is rapidly achieved at the beginning. For lead and cadmium ions, sorption efficiency quickly increased from 20 to 140 min until reaching a saturation stage in multifunctional cobalt oxide nanocomposites [18]. The contact time considerably impacts calculating kinetic parameters and roughly estimating the equilibrium time. Adsorption tests were conducted at various contact times ranging from 0 to 280 min while maintaining other parameters constant to determine the effect of contact time on the adsorption of Cu^{2+} , Pb^{2+} , Cd^{2+} , and Zn^{2+} ions on the activated carbon-supported silver-silica nanocomposite reported by Nyirenda *et al.*, [19]. In the first 40 min, the percentage removal of Cu^{2+} , Pb^{2+} , Cd^{2+} , and Zn^{2+} ions grew quickly; after that, it reached a steady state. The percentage removal increased as the contact time extended from 0 to 280 minutes, reaching a stable value of 94%, 99%, 90%, and 97% for Cu^{2+} , Pb^{2+} , Cd^{2+} , and Zn^{2+} ions, respectively observed by Kumar *et al.*, [30]. The metal adsorption rate is high initially and then increases slowly until the saturation level reaches equilibrium at 60 minutes was conducted by Zhang *et al.*, [21]. Figure 2 represents the role of nanocomposite adsorbents in environmental remediation.

Investigating the cost-effectiveness and regeneration of nanocomposite adsorbents in practical applications is essential. The nanocomposite adsorbents under consideration could be regenerated by 4-6 cycles, a practical and feasible process. This suggests that the repeated adsorption/desorption process reduces the pollutant level to a lesser volume and also increases the adsorbents' economic viability. The adsorption parameters of the nanocomposite adsorbents are summarized in Table 1.



Fig. 2. Graphical representation of the nanocomposite adsorbent's role in environmental remediation

4. Isotherm, Kinetics and Thermodynamic Studies

In practical applications, isotherm models are essential for predicting every aspect of adsorption behavior and determining the maximum number of contaminants that may be removed from wastewater at a constant temperature. The experimental data were a better fit to the Freundlich isotherm model than the Langmuir, suggesting multilayered adsorption of CuMgAl-layered double hydroxide/montmorillonite nanocomposite for the adsorption of Cd(II) reported by Alnasrawi *et al.*, [17]. Since the mean free energy of surface adsorption was 2.8 kJ/mol, a confirmed physical sorption process (E < 8 kJ/mol) is justified with a low interaction of adsorbate and adsorbent (low k_L value). The activated carbon-supported silver-silica nanocomposite removed metal ions individually and as a multi-ion mixture, following the Freundlich isotherm model with a high value of adsorption intensity constant (n_F) followed by a physical adsorption process. Graphene oxide/polyvinyl alcohol/Fe₃O₄ nanocomposite observed by Nyirenda *et al.*, [19] and Fe₃O₄/Talc nanocomposite followed the monolayer adsorption by the Langmuir isotherm model with a lower isotherm constant (k_L) reported by Huong *et al.*, [20]. The adsorption of Cr(VI) by Fe₃O₄/Talc nanocomposite was a chemisorption process, and an increase of k_F was reported with a temperature rise [20]. The kinetic behavior of different heavy metal ion adsorptions on the selected nanocomposite adsorbents was investigated using the *pseudo*-first-order and *pseudo*-second-order kinetic models. The kinetic data suggested that most adsorbents followed a *pseudo*-second-order kinetic model because the adsorption process depends on the number of active adsorption sites on the adsorbent and the adsorbent ion concentrations.

Thermodynamics studies provide better information on the spontaneity of the adsorption process. Deciding whether a reaction is favorable or not gives more detail about the properties of the adsorbent and adsorbed at equilibrium conditions [31]. In determining the enthalpy, entropy, and Gibb's free energy, which are key terms in every thermodynamic model, the mathematical Eq. (1) to Eq. (3) are usually employed [30].

$$K_d = \frac{C_A}{C_B} \tag{1}$$

$$\Delta G^0 = RT lnk_d \tag{2}$$

$$lnk_d = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT}$$
(3)

where K_d represents the thermodynamic equilibrium constant, where T is the absolute temperature (K), ΔG^o (kJ/mol) is the Gibbs free energy change, ΔH^o (kJ/mol) is the enthalpy change, R is the gas constant (8.314 J/mol/K), and ΔS^o (kJ/mol/K) is the entropy change. The adsorption process's mechanism can also be studied using thermodynamic analysis. Adsorption may be accomplished by physisorption, chemisorption, or a combination of the two, according to the range of ΔG^o values for various adsorbents observed by Ighalo *et al.*, [32]. Table 2 illustrated that the adsorption of different heavy metals onto the surface of various nanocomposite adsorbents is a spontaneous and feasible process, as indicated by Gibb's free energy ($\Delta G^o < 0$).

While most of the adsorption process, in Table 2, the entropy change (ΔS^0) was endothermic in nature because an increase in temperature promotes the adsorption due to increased ion mobility, which allows for easier access to active areas observed by Nassar *et al.*, [33]. The adsorption process of Cd²⁺ on CuMgAl-layered double hydroxide/montmorillonite nanocomposite was exothermic, suggesting that increased temperature reduced the adsorption capacity due to the strong chemical bonds between the adsorbate and adsorbent [17]. The adsorption of Cr(VI) by Fe₃O₄/Talc nanocomposite was also an exothermic reaction, indicating a chemical adsorption process reported by Huong *et al.*, [20]. However, it was found that as temperatures, the adsorbate's experience stronger binding forces to specific sites or reorganize on the surface, reducing their mobility and randomness [34].

Activated carbon-supported silver-silica nanocomposite and graphene oxide/polyvinyl alcohol/Fe₃O₄ nanocomposite showed that there is an increased disorder in the interaction of metal ions with the composites' surfaces as adsorption progresses. Conversely, the adsorption of Cd²⁺ on CuMgAl-layered double hydroxide/montmorillonite nanocomposite and Cr⁶⁺ on Fe₃O₄/Talc adsorbent showed a negative entropy change, which could be attributed to the addition of sorbate to the adsorbent by the chemical adsorption process and lowers the system's degrees of freedom observed by Shan *et al.*, [35].

Table 2

Isotherm, kinetics and thermodynamic study of the nanocomposite adsorbents

Nanocomposite adsorbent	Heavy metal ions	Isotherm	Kinetics	Т(К)	ΔG° (kJ/mol)	Δ <i>H</i> ° (kJ/mol)	Δ <i>S</i> ° (kJ/mol/K)	References
CuMgAl-layered Double hydroxide/ montmorillonite nanocomposite	Cd(II)	Freundlich	<i>Pseudo-</i> 2 nd order	298 303 308 313	-3.02 -1.59 -0.771 -0.225	-57.0	-185	[17]
Activated carbon- supported silver- silica nanocomposite	Cu(II) Pb(II) Cd(II) Zn(II)	Freundlich	Pseudo- 2 nd order	298 308 318 328	-6.85±0.19 -13.4±0.24 -7.59±0.17 -13.7±0.20	30.24±4.79 25.89±1.74 22.81±4.50 39.12±8.70	123.6±16.02 126.4±6.15 95.1±14.33 162.4±27.17	[19]
Graphene oxide/polyvinyl alcohol/Fe ₃ O ₄ nanocomposite	Co(II)	Langmuir	Pseudo- 2 nd order	298 303 313 323	-9.8 -10.07 -10.06 -12.38	22.04	106.47	[24]
Fe ₃ O ₄ /Talc nanocomposite	Cr(VI)	Langmuir Freundlich	<i>Pseudo-</i> 2 nd - order	298 303 313	-0.478 -3.033 -7.796	20.9±418	-0.362	[20]

5. Comparative Study of Nanocomposite Adsorbents

Table 3 showed a comparative study of heavy metal removal efficiency among recently developed nanocomposite adsorbents. The study observed superparamagnetic iron oxide nanoparticles had the greatest affinity towards the removal of Pb(II) from wastewater. Moreover, zeolite, mesoporous silica, hydroxyapatite, and activated carbon had a strong affinity towards Pb(II) removal. The incorporation of some active graphene oxide functional groups, like GO-COO- and GO-O- in GO-SiO₂ improved the adsorption of Pb(II) through a cation exchange reaction. It was found that the maximum 99% of Pb removal efficiency with Q_m (527 mg/g) was due to the presence of –OH, -COOH functional groups [29]. Additionally, the porous structure, along with improved synergistic impact between graphene oxide and SiO₂, provided more dynamic adsorption sites for Pb(II) ions. The seaweed's amorphous polysaccharide embedding matrix, which has a strong affinity for divalent cations, is essentially responsible for the 97.25% removal of Pb(II) (Q_m =333.33mg/g) [28]. Due to its high surface area, the activated carbon-supported silver-silica nanocomposite could remove 94.3% of Pb(II) $(Q_m=81.30 \text{ mg/g})$. Another study revealed that polythiophene-modified chitosan-magnetite nanocomposites could effectively eliminate 100% of Hg(II) ions (Q_m =52.8mg/g) [23]. This is because the materials based on chitosan nanoparticles have a higher capacity to adsorb mercury from aqueous solutions due to their relatively high surface area in combination with the presence of phosphorus atoms having a high affinity for Hg(II) ions, which is regulated by chemical adsorption

(chemisorption). The sorbent and sorbate involved valence forces via the sharing or exchange reaction.

Table 3

Heavy	Nanocomposite	Removal	Q_m	References
metals	adsorbent	efficiency (%)	(mg/g)	
Pb(II)	Multifunctional cobalt oxide nanocomposite	87	55.24	[18]
	Activated carbon-supported silver-silica nanocomposite	94	81.30	[19]
	Chitosan/dopamine/Fe₃O₄ nanocomposite	84	3.6	[21]
	Iron oxide-tea waste nanocomposite	95	18.83	[25]
	Hydroxyapatite/Fe ₃ O ₄ nanocomposite	96	109.89	[26]
	Zeolite-superparamagnetic iron oxide nanocomposite	96	205	[36]
	Polypyrrole - iron oxide seaweed (PPy-Fe ₃ O ₄ - SW) nanocomposite	97	333.33	[28]
	Mesoporous Silica-Graphene Oxide nanocomposite	99	527	[29]
Hg(II) Polythiophene	Polythiophene modified chitosan/magnetite nanocomposites	100	52.8	[23]
	Zeolite-superparamagnetic iron oxide nanocomposite	70	12	[36]
	Chitosan/dopamine/Fe₃O₄ nanocomposite	88	20.2	[21]
Cd(II)	Multifunctional cobalt oxide nanocomposite	82	52.91	[18]
	Activated carbon-supported silver-silica nanocomposite	87	81.97	[19]
	CuMgAl-layered double hydroxide/montmorillonite nanocomposite	95	175	[17]
	Magnesium oxide/bentonite nanocomposite	91	200	[27]
As(III)	Mesoporous Silica-Graphene Oxide nanocomposite	10	30	[29]
	Cellulose-Sn(IV) Bio composite	95	16.64	[37]

The sorbent and sorbate involved valence forces via the sharing or exchange of electrons. The chemical adsorption process has an inverse relationship between the adsorption capacity and the number of active sites on the adsorbent surface [38]. Chitosan/dopamine/Fe₃O₄ nanocomposite could remove 88% of Pb(II) having (Qm = 20.2 mg/g) adsorption capacity due to the presence of an increased number of adsorption sites, which was synthesized by assembling biomimetic polymer (PDA) and chitosan onto magnetic nanoparticles. The removal of Cd(II) ion magnesium oxide/bentonite nanocomposite showed 91% removal efficiency (Qm = 200 mg/g) through cation exchange and precipitation reactions [27]. CuMgAl-layered double hydroxide/montmorillonite nanocomposite showed 95% removal efficiency of Cd (II) ion having (Qm=175 mg/g) due to the presence of the clay mineral of montmorillonite (MMt) having a higher surface area [17]. Activated carbon-supported silver-silica nanocomposite gave 87.7% removal efficiency of Cd(II) removal (Qm=81.97 mg/g) due to the high surface area, physicochemical stability, and porosity of activated carbon, and it was a sustainable adsorbent because silver and silica nanoparticles originated from waste biomass reported by Nyirenda *et al.*, [19].

The increased adsorption capacity of the mesoporous silica-graphene oxide nanocomposite for the removal of As(III) ions was due to its higher surface area, and it was observed ligand interchange among the Si-OH with As(III) ions, which also enhanced the adsorption capacity reported by Barik *et*

al., [29]. The Cellulose-Sn(IV) Bio composite showed a greater adsorption efficiency than conventional cellulose due to modification with Sn(IV) chloride. The chloride ions present on the surface of the Cellulose-Sn(IV) composite may involve ion exchange with the As(III) ion in solution.

6. Limitations, Challenges and Opportunities

Water treatment applications can greatly benefit from the use of recently developed nanocomposite adsorbents like graphene oxide, mesoporous silica, and zeolites. A few obstacles must be overcome to enjoy their advantages. A few of these nanocomposite materials have improved stability and reusability for extended periods, economic viability and sustainability in large-scale manufacturing, and efficient dispersion capacity within the biopolymer matrix [39]. Two important factors that are considered as environmental sustainability by the management of wasted adsorbents and the regeneration of nanocomposite materials. Despite these obstacles, research and development are continuing to offer improved synthesis techniques and sophisticated nanocomposite materials [40]. A combination of membrane filtration technology and magnetic adsorbents can improve water treatment procedures by increasing the efficiency of separation and adsorption capacity [20]. However, there are benefits to rapid and environmentally friendly water filtration systems when multifunctional and reactive biopolymer-based nanocomposites are incorporated into current water treatment technologies [19]. Researchers and industry partners need to work together to develop these strategies further and provide long-term, commercially viable water treatment techniques. The water treatment process will be more effective if alternative methods and real industrial effluent tracking are further investigated.

7. Conclusions

Nanocomposites have been used as an effective tool to remove heavy metals from drinking water and wastewater in recent years and have shown high efficiency due to their small particle size and large surface area. Most of the nanocomposites are crystalline in size and effective for the removal of Pb(II), Cd(II), Cr(VI), Cu(II), Ni(II), As(III), and Zn(II) at moderate pH values ranging from 6 to 8, and a minimum amount of adsorbent dosage was required to suggest the applicability of the adsorbents on a large scale. Most nanocomposites achieved maximum adsorption capacity with a moderate pH (5-9), a minimum adsorbent dosage (0.1-2 g/L), and 20-60 min for equilibrium. Most could be used in 4-6 cycles, suggesting their economic feasibility. Depending on the adsorption intensity constant (n_F) , the adsorbents follow monolayer or multilayer adsorption. Multilayer adsorption on a heterogeneous surface is favored when $n_F>1$, and less favorable or monolayer adsorption occurs when $n_F \leq 1$. Most of the adsorbents follow *pseudo*-2nd-order kinetics. Adsorption may be accomplished by physisorption, chemisorption, or a combination of the two depending on their mean free energy of surface adsorption, Langmuir constant (k_L) and Freundlich constant (n_F). Physisorption phenomena occurred at a mean free energy (E < 8 kJ/mol), low k_L value (weak interaction between adsorbate and adsorbent), and chemisorption occurred with a mean free energy (E>8 kJ/mol) with high k_{L} value (strong interaction between adsorbate and adsorbent). Thermodynamic study of the adsorbents showed the spontaneous nature of the adsorption process. A comparative analysis of the nanocomposite adsorbents revealed that mesoporous silica-graphene oxide nanocomposite could remove 99% of Pb(II) ions. Moreover, iron oxide nanocomposites have greater efficiency towards removing Pb(II) ions. Polythiophene-modified chitosan/magnetite nanocomposites could remove 100% Hg(II) ions. CuMgAl-layered double hydroxide/montmorillonite nanocomposite could remove 95% of Cd(II) ions caused by the higher surface area of clay material. Large-scale applications of the

nanocomposite adsorbent faced challenges for stability, extended periods of reusability, economic viability, and environmental sustainability. The results of this study would help the direction of future research in the green synthesis process, considering the commercialization cost, time, and energy consumption for the remediation of heavy metals from drinking and wastewater in a sustainable way.

Acknowledgement

This research was funded by the financial support from the University Grant Commission (UGC), Bangladesh as fellowship.

References

- [1] Anjum, Muzammil, R. Miandad, Muhammad Waqas, F. Gehany, and M. A. Barakat. "Remediation of wastewater using various nano-materials." *Arabian journal of chemistry* 12, no. 8 (2019): 4897-4919. <u>https://doi.org/10.1016/j.arabjc.2016.10.004</u>
- [2] Häder, Donat-P., Anastazia T. Banaszak, Virginia E. Villafañe, Maite A. Narvarte, Raúl A. González, and E. Walter Helbling. "Anthropogenic pollution of aquatic ecosystems: Emerging problems with global implications." *Science of the Total environment* 713 (2020): 136586. <u>https://doi.org//10.1016/j.scitotenv.2020.136586</u>
- [3] Islam, Md Shajedul, and M. G. Mostafa. "Occurrence, source, and mobilization of iron, manganese, and arsenic pollution in shallow aquifer." *Geofluids* 2023, no. 1 (2023): 6628095. <u>https://doi.org/10.1155/2023/6628095</u>
- [4] Islam, Md Zahidul, and M. G. Mostafa. "Iron, manganese, and lead contamination in groundwater of Bangladesh: a review." Water Practice & Technology 19, no. 3 (2024): 745-760. <u>https://doi.org/10.2166/wpt.2024.030</u>
- [5] Emmanuel, Ugwu Chidiebere, Maduka Ignatius Chukwudi, Suru Stephen Monday, and Anakwuo Ikechukwu Anthony. "Human health risk assessment of heavy metals in drinking water sources in three senatorial districts of Anambra State, Nigeria." *Toxicology reports* 9 (2022): 869-875. https://doi.org/10.1016/j.toxrep.2022.04.011
- [6] Mostafa, M. G., SM Helal Uddin, and A. B. M. H. Haque. "Assessment of hydro-geochemistry and groundwater quality of Rajshahi City in Bangladesh." Applied Water Science 7 (2017): 4663-4671. <u>https://doi.org/10.1007/s13201-017-0629-y</u>
- [7] Trivunac, Katarina, and Slavica Stevanovic. "Removal of heavy metal ions from water by complexation-assisted ultrafiltration." Chemosphere 64, no. 3 (2006): 486-491. <u>https://doi.org/10.1016/j.chemosphere.2005.11.073</u>
- [8] Islam, M. R., and M. G. Mostafa. "Adsorption kinetics, isotherms and thermodynamic studies of methyl blue in textile dye effluent on natural clay adsorbent." Sustainable Water Resources Management 8, no. 2 (2022): 52. <u>https://doi.org/10.1007/s40899-022-00640-1</u>
- Khan, Adnan, Syed Badshah, and Claudio Airoldi. "Dithiocarbamated chitosan as a potent biopolymer for toxic cation remediation." Colloids and Surfaces B: Biointerfaces 87, no. 1 (2011): 88-95. https://doi.org/10.1016/j.colsurfb.2011.05.006
- [10] El Messaoudi, Noureddine, Mohammed El Khomri, Yasmine Fernine, Amal Bouich, Abdellah Lacherai, Amane Jada, Farooq Sher, and Eder C. Lima. "Hydrothermally engineered Eriobotrya japonica leaves/MgO nanocomposites with potential applications in wastewater treatment." *Groundwater for Sustainable Development* 16 (2022): 100728. <u>https://doi.org/10.1016/j.gsd.2022.100728</u>
- [11] Mostafa, M. G., Yen-Hua Chen, Jiin-Shuh Jean, Chia-Chuan Liu, and Yao-Chang Lee. "Kinetics and mechanism of arsenate removal by nanosized iron oxide-coated perlite." *Journal of Hazardous Materials* 187, no. 1-3 (2011): 89-95.. <u>https://doi.org/10.1016/j.jhazmat.2010.12.117</u>
- [12] Mostafa, M. G., Yen-Hua Chen, Jiin-Shuh Jean, Chia-Chuan Liu, and Hsisheng Teng. "Adsorption and desorption properties of arsenate onto nano-sized iron-oxide-coated quartz." *Water Science and Technology* 62, no. 2 (2010): 378-386. <u>https://doi.org/10.2166/wst.2010.288</u>
- [13] Almeida, C. A. P., N. A. Debacher, A. J. Downs, L. Cottet, and C. A. D. Mello. "Removal of methylene blue from colored effluents by adsorption on montmorillonite clay." *Journal of colloïd and interface science* 332, no. 1 (2009): 46-53. <u>https://doi.org/10.1016/j.jcis.2008.12.012</u>
- [14] Islam, M. Rafiqul, and M. G. Mostafa. "Characterization of textile dyeing effluent and its treatment using polyaluminum chloride." *Applied Water Science* 10, no. 5 (2020): 1-10. <u>https://doi.org/10.1007/s13201-020-01204-</u> <u>4</u>
- [15] Wu, Wei, Quanguo He, and Changzhong Jiang. "Magnetic iron oxide nanoparticles: synthesis and surface functionalization strategies." *Nanoscale research letters* 3 (2008): 397-415. <u>https://doi.org/10.1007/s11671-008-9174-9</u>

- [16] Islam, Md Shajedul, and Mostafa MG. "Impacts of Climate Change on Global Freshwater Quality and Availability: A Comprehensive Review." Journal of Water and Environment Technology 22, no. 1 (2024): 1-26. https://doi.org/10.2965/jwet.23-036
- [17] Alnasrawi, Fatin A., and Ahmed A. Mohammed. "Enhancement of Cd²⁺ removal on CuMgAl-layered double hydroxide/montmorillonite nanocomposite: Kinetic, isotherm, and thermodynamic studies." *Arabian Journal of Chemistry* 16, no. 2 (2023): 104471. <u>https://doi.org/10.1016/j.arabjc.2022.104471</u>
- [18] El Mouden, Abdelaziz, Noureddine El Messaoudi, Abdelqader El Guerraf, Amal Bouich, Valbonë Mehmeti, Abdellah Lacherai, Amane Jada, and Farooq Sher. "Multifunctional cobalt oxide nanocomposites for efficient removal of heavy metals from aqueous solutions." Chemosphere 317 (2023): 137922. https://doi.org/10.1016/j.chemosphere.2023.137922
- [19] Nyirenda, J., Kalaba, G., & Munyati, O. (2022). Synthesis and characterization of an activated carbon-supported silver-silica nanocomposite for adsorption of heavy metal ions from water. Results in Engineering, 15, 100553. <u>https://doi.org/10.1016/j.rineng.2022.100553</u>
- [20] Thi Huong, Nguyen, Nguyen Ngoc Son, Vo Hoang Phuong, Cong Tien Dung, Pham Thi Mai Huong, and Le Thanh Son. "Synthesis Fe₃O₄/Talc nanocomposite by coprecipition-ultrasonication method and advances in hexavalent chromium removal from aqueous solution." *Adsorption Science & Technology* 38, no. 9-10 (2020): 483-501. <u>https://doi.org/10.1177/0263617420969112</u>
- [21] Zhang, X., Li, B., Han, X., & Wang, N. (2021). Preparation, characterization and application of CS@ PDA@ Fe₃O₄ nanocomposite as a new magnetic nano-adsorber for the removal of metals and dyes in wastewater. RSC advances, 11(38), 23679-23685. <u>https://doi.org/10.1039/D1RA03779J</u>
- [22] Zhang, Xingjing, Baohe Li, Xiaoqian Han, and Nong Wang. "Preparation, characterization and application of CS@ PDA@ Fe₃O₄ nanocomposite as a new magnetic nano-adsorber for the removal of metals and dyes in wastewater." *RSC advances* 11, no. 38 (2021): 23679-23685. <u>https://doi.org/10.1039/D1RA03779J</u>
- [23] Morsi, Rania E., Ahmed M. Al-Sabagh, Yasser M. Moustafa, Said G. ElKholy, and Mostafa S. Sayed. "Polythiophene modified chitosan/magnetite nanocomposites for heavy metals and selective mercury removal." *Egyptian journal* of petroleum 27, no. 4 (2018): 1077-1085. <u>https://doi.org/10.1007/978-981-10-7284-0_5</u>
- [24] Le, Thu Dieu, Luyen Thi Tran, Hue Thi Minh Dang, Thi Thu Huyen Tran, and Hoang Vinh Tran. "Graphene oxide/polyvinyl alcohol/Fe₃O₄ nanocomposite: an efficient adsorbent for Co (II) ion removal." *Journal of Analytical Methods in Chemistry* 2021, no. 1 (2021): 6670913. <u>https://doi.org/10.1155/2021/6670913</u>
- [25] Khanna, Mansi, Ashish Mathur, Ashwani Kumar Dubey, James McLaughlin, Igamcha Moirangthem, Shikha Wadhwa, Devraj Singh, and Ranjit Kumar. "Rapid removal of lead (II) ions from water using iron oxide-tea waste nanocomposite-a kinetic study." *IET nanobiotechnology* 14, no. 4 (2020): 275-280. <u>https://doi.org/10.1049/iet-nbt.2019.0312</u>
- [26] Vahdat, Armin, Behrooz Ghasemi, and Mardali Yousefpour. "Synthesis of hydroxyapatite and hydroxyapatite/Fe3O4 nanocomposite for removal of heavy metals." *Environmental Nanotechnology, Monitoring & Management* 12 (2019): 100233. <u>https://doi.org/10.1016/j.enmm.2019.100233</u>
- [27] Elkhatib, Elsayed A., Mohamed L. Moharem, Ahmed F. Saad, and Farida A. Attia. "Using nano-magnesium oxide/bentonite composite for cadmium removal from industrial wastewater." *Environmental Engineering Research* 28, no. 2 (2022): 210545. <u>https://doi.org/10.4491/eer.2021.545</u>
- [28] Sarojini, Gopalakrishnan, Samikannu Venkateshbabu, and Manivasagan Rajasimman. "Facile synthesis and characterization of polypyrrole-iron oxide–seaweed (PPy-Fe3O4-SW) nanocomposite and its exploration for adsorptive removal of Pb (II) from heavy metal bearing water." *Chemosphere* 278 (2021): 130400. <u>https://doi.org/10.1016/j.chemosphere.2021.130400</u>
- [29] Barik, Bapun, Aniket Kumar, Pratap S. Nayak, L. Satish K. Achary, Lipeeka Rout, and Priyabrat Dash. "Ionic liquid assisted mesoporous silica-graphene oxide nanocomposite synthesis and its application for removal of heavy metal ions from water." *Materials Chemistry and Physics* 239 (2020): 122028. https://doi.org/10.1016/j.matchemphys.2019.122028
- [30] Kumar, P. Senthil, S. Ramalingam, R. V. Abhinaya, K. V. Thiruvengadaravi, P. Baskaralingam, and S. Sivanesan. "Lead (II) adsorption onto sulphuric acid treated cashew nut shell." *Separation Science and Technology* 46, no. 15 (2011): 2436-2449. <u>https://doi.org/10.1080/01496395.2011.590174</u>
- [31] Anastopoulos, Ioannis, Amit Bhatnagar, Bassim H. Hameed, Yong Sik Ok, and Michalis Omirou. "A review on wastederived adsorbents from sugar industry for pollutant removal in water and wastewater." *Journal of Molecular Liquids* 240 (2017): 179-188. <u>https://doi.org/10.1016/j.molliq.2017.05.063</u>
- [32] Ighalo, Joshua O., Chinenye Adaobi Igwegbe, Adewale George Adeniyi, Comfort Abidemi Adeyanju, and Samuel Ogunniyi. "Mitigation of Metronidazole (Flagyl) pollution in aqueous media by adsorption: a review." *Environmental Technology Reviews* 9, no. 1 (2020): 137-148. https://doi.org/10.1080/21622515.2020.1849409

- [33] Nassar, Nashaat N. "Rapid removal and recovery of Pb (II) from wastewater by magnetic nanoadsorbents." *Journal of hazardous materials* 184, no. 1-3 (2010): 538-546. <u>https://doi.org/10.1016/j.jhazmat.2010.08.069</u>
- [34] Farghali, R. A., M. Sobhi, S. E. Gaber, H. Ibrahim, and E. A. Elshehy. "Adsorption of organochlorine pesticides on modified porous Al30/bentonite: kinetic and thermodynamic studies." *ArabianJournal of Chemistry* 13, no.8 (2020): 6730-6740. <u>https://doi.org/10.1016/j.arabjc.2020.06.027</u>
- [35] Shan, Ran-ran, Liang-guo Yan, Kun Yang, Yuan-feng Hao, and Bin Du. "Adsorption of Cd (II) by Mg–Al–CO₃-and magnetic Fe₃O₄/Mg–Al–CO₃-layered double hydroxides: Kinetic, isothermal, thermodynamic and mechanistic studies." *Journal of hazardous materials* 299 (2015): 42-49. <u>https://doi.org/10.1016/j.jhazmat.2015.06.003</u>
- [36] Nogueira, Helton P., Sergio H. Toma, Alceu T. Silveira Jr, and Koiti Araki. "Zeolite-SPION nanocomposite for ammonium and heavy metals removal from wastewater." *Journal of the Brazilian Chemical Society* 31 (2020): 2342-2350. <u>https://doi.org/10.21577/0103-5053.20200097</u>
- [37] Shekhawat, Anita, Ravin Jugade, Vaishnavi Gomase, Shashikant Kahu, Saravanan Dhandayutham, and Sadanand Pandey. "Adsorptive removal of As(III) by cellulose-Sn (IV) biocomposite." *Journal of Composites Science* 7, no. 1 (2023): 19. <u>https://doi.org/10.3390/jcs7010019</u>
- [38] Figaro, S., J. P. Avril, F. Brouers, A. Ouensanga, and S. Gaspard. "Adsorption studies of molasse's wastewaters on activated carbon: Modelling with a new fractal kinetic equation and evaluation of kinetic models." *Journal of hazardous materials* 161, no. 2-3 (2009): 649-656. <u>https://doi.org/10.1016/j.jhazmat.2008.04.006</u>
- [39] Kolya, Haradhan, and Chun-Won Kang. "Polyvinyl acetate/reduced graphene oxide-poly (diallyl dimethylammonium chloride) composite coated wood surface reveals improved hydrophobicity." Progress in Organic Coatings 156 (2021): 106253. <u>https://doi.org/10.1016/j.porgcoat.2021.106253</u>
- [40] Rouf, Tahrima B., and Jozef L. Kokini. "Biodegradable biopolymer–graphene nanocomposites." Journal of Materials Science 51 (2016): 9915-9945. <u>https://doi.org/10.1007/s10853-016-0238-4</u>