

Natural Adsorbents For Heavy Metal Removal of Industrial Waste Water: Review Study

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Abstract

Heavy metals found in industrial effluent can pose serious threats to the human health and environment, Because of their ecological durability and toxicity. One crucial stage in the wastewater treatment is the removal of these contaminants. Of the several methods employed, adsorption is found to be a cost effective method for heavy metals removal. This procedure depends on sorbents-natural or synthetic-that can attach to metal ions and extract them from water. This study focused on reviewing the role of researchers in finding inexpensive adsorbent materials, environmentally appropriate sorbents, particularly for natural materials and agricultural and industrial wastes. These materials' availability, affordability, and high adsorption efficiency of define them. Demonstrating their capacity to extract heavy metals from effluent from industries. pH, contact time, concentration of metals, temperature, and the adsorbent dose amount were among the influencing parameters that were examined. Every one of these elements is essential in establishing the total adsorption capacity. Among the models most commonly used by researchers to study adsorption behavior, the Langmuir and Freundlich models are the most popular and widely used in environmental and industrial applications. The Langmuir model is characterized by its ability to describe adsorption on a homogeneous surface in a single layer formation, making it ideal for determining the maximum adsorption capacity, which is suitable for many materials with specific adsorption sites. For the reundlich model, is characterized by its flexibility in dealing with heterogeneous surfaces and multilayer adsorption interactions, making it suitable for systems of a complex nature. The popularity of these two models is due to their simplicity and accuracy in representing experimental data in a wide range of applications.

Keywords: Adsorption, Heavy metal, Industrial wastewater, Isotherm.

1. Introduction

One major cause of contamination in the environment is industrial effluent. Because of their toxicity, heavy metals have grown in importance among environmental contaminants [1] Generally speaking, heavy metals are defined as having a density of more than 5 g per cubic centimeter. A sizable number of components fall under this group [2]. Arsenic, chromium,

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manganese, iron, cobalt, copper, nickel, cadmium, zinc, mercury, and lead are the metals that the WHO considers to be of the utmost urgent concern [3]. The toxicity and allowable limits of several metal ions are listed in Table 2 [4]. Heavy metals are present in the environment through a several of processes including the manufacture of batteries, painting, printing, burning coal, sewage effluents, vehicle emissions, mining operations, tanneries, alloy making, and the usage of fossil fuels [5]. Economic and environmental considerations have made the recovery and extraction of heavy metal ions from industrial effluent a viable strategy. Chemical precipitation, reverse osmosis, filtration, electrochemical processes, ion exchange, and adsorption are examples of conventional procedures for eliminating heavy metals from effluents [1]. "Adsorption" is the most popular method among them. Because it uses adsorbent materials, especially ion exchange resins, to remove heavy metal cations, this approach is both economical and environmentally friendly [6]. The change in a particular substance's concentration at the interface relative to neighboring phases is referred to as adsorption. The kinds of phases that come into contact with one another affect how effective this process is [7]. An alternative in the form of cost-effective adsorbents for water treatment has been the subject of extensive research in recent years. Because of their potential adsorption capacities, a variety of natural materials or wastes have been used as adsorbents for the adsorption process; these materials can be used naturally or with certain changes. Long-term results show that employing inexpensive adsorbents to remove heavy metals is more hopeful because there are many readily available, locally accessible materials, such as natural materials, agricultural waste, or industrial by-products, that can be used as inexpensive adsorbents. Finding a range of materials as inexpensive adsorbents is becoming more and more popular, according to earlier studies [8].

S. No.	Heavy metals	Health hazards	Permissible limit (mg/L)(WHO)
1.	Cadmium (Cd)	Kidney damage, renal disorder, human carcinogen, emphysema.	0.003
2.	Mercury (Hg)	Neurological damage, paralysis, blindness, rheumatoid arthritis and anorexia.	0.001
3.	Arsenic (As)	Skin, lung, bladder and kidney cancer, neurological disorder, muscular weakness and nausea.	0.01
4.	Lead (Pb)	Brain damage, anaemia, anorexia, vomiting, disease of circulatory and nervous systems	0.05
5.	Chromium (Cr)	Headache, diarrhoea, nausea, vomiting, carcinogenic, lung tumour	0.05
6.	Cobalt (Co)	Asthma like allergy, damage to the heart, damage to the thyroid and liver, carcinogenic	0.1
7.	Copper (Cu)	Liver damage, Wilson disease, Insomnia.	2.5
8.	Zinc (Zn)	Depression, lethargy, neurological signs, dehydration anaemia and increased thirst.	5
9.	Manganese (Mn)	Syndrome of motor dysfunction, memory loss resembling, Parkinson disease.	0.5
10.	Iron (Fe)	Headache, Brittle nails, Depression, Constipation, Tinnitus, Gastrointestinal complains.	0.3
11.	Nickel (Ni)	Dermatitis, nausea, chronic asthma, coughing and cancer of the lung.	2

Table 2. The harmful effect of heavy metal ions and the World Health Organization's acceptable limits [4].

2.

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Objective of study

By examining the variables influencing the adsorption process and evaluating the effectiveness of the materials employed, this study seeks to determine if adsorption techniques can effectively metals removal from industrial- wastewater. By using effective and low cost materials, our research seeks to offer good and ultimate solutions to lessen the heavy metals effect on the environment.

3. Adsorption Techniques

The mass transfer of pollutant from a gas or liquid state to a solid surface, where they are attached by chemical or physical interactions, is known as adsorption. This process involves molecules leaving the solution and attaching themselves to the chemical's surface through chemical and physical bonding [9]. The most popular and successful technique for removing dangerous heavy metals from industrial effluent is adsorption technology. Toxic compounds are transferred to the adsorbent's surface by physical or chemical means (Figure 1). The adsorption process is a cost-effective method with low operating costs that produces less contamination during the hazardous metal extraction process when compared to conventional methods. Adsorption technologies are an environmentally friendly strategy because they allow for sorbent regeneration and repeated use for effective removal [10]. The main characteristics needed for choosing adsorbents are cost-effectiveness, wide pore size, surface area, polar properties of it, and functional groups, they all affect how well adsorption techniques work [11]. When a gasliquid solution accumulates on the surface of a solid-liquid, Adsorption occurs, forming the adsorbate, a molecular structure [12].



Figure 1. (a) Adsorption Mechanism (b) monolayer adsorption (c) multilayer adsorption [10].

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3.1. Type of adsorption

Physical Adsorption: is the process of accumulation of molecules or ions on the surface of a solid due to weak van der Waals forces without forming chemical bonds. It is rapid and depends on temperature and pressure.

Chemical Adsorption: is the process in which ionic bonds are formed between an adsorbent and an adsorbate as a result of a change in the electron density between them. Adsorption models reflect the mechanism of this process, and the most appropriate model is selected based on experimental physical criteria. These criteria include adsorption kinetic studies, adsorption equilibrium, and thermodynamic isotherm analysis, which are used to compare the performance of different models and determine the best one for the system under study [13].

4. Adsorption Isotherm

One important tool for determining the theoretical maximum adsorption capacity and potential interactions between adsorbents and adsorbates is the adsorption isotherm model [14]. Two important adsorption models are the Freundlich and Langmuir isotherms. The Langmuir curve assumes that absorption occurs in a monolayer on a surface with a finite number of homogeneous absorption sites, and assumes that there is no lateral diffusion of the adsorbent across the surface [15]. When describing heterogeneous surfaces, which have a variety of adsorption sites with varying energies, the Freundlich isotherm model is employed [16]. In the case of surface adsorption, the adsorption interaction between surfaces and absorbents is described by the Langmuir isotherm equation. It is predicated on the idea that the absorbent's surface has a limited number of sites and that each site can only absorb a single absorbent molecule. This presumption suggests that the absorbent's surface is uniform, indicating that every site has an equal capacity to adsorb molecules and that the adsorption process can proceed in both directions. The Langmuir isotherm is articulated as [17]:

$$\frac{1}{X/M} = \frac{1}{q_{max}} + \frac{1}{q_{max} b} \frac{1}{C_e}$$

b constant it increases when the size of the molecular increasing; q_{max} represents The maximum amount that an absorbent material can absorb.; X is the weight of the adsorbed substance; M signifies the weight of the adsorbent; and Ce indicates the remaining concentration of the absorbent in the solution at equilibrium.

The equilibrium parameter RL, a dimensionless constant also known as the separation factor or equilibrium parameter, articulates the fundamental characteristics of the Langmuir isotherm [18]:

$$R_{\rm L} = \frac{1}{1 + bC_0} \tag{2}$$

RL indicates whether the isothermal type is inappropriate when $R_1 > 1$, linear when $R_1 = 1$, appropriate when $0 < R_1 < 1$, or irreversible when $R_1 = 0$.

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The equation of Freundlich is an empirical formulation that effectively characterizes a significant amount of adsorption data. The Freundlich isotherm is articulated as [19]:

$$\log q_e = \log K + \frac{1}{n} * \log C_e \tag{3}$$

K and n are constants that change with temperature change.

The Harkins & Jura adsorption isotherm is articulated as follows [20]:

$$\frac{1}{q_e^2} = \frac{B}{A} - \frac{\log C_e}{A} \tag{4}$$

B and A represent the isotherm constants. The presence of a heterogeneous pore distribution clarifies the isotherm equation, which addresses multilayer adsorption. The relationship log Ce and $\frac{1}{a^2}$ Produces a linear graph with a 1/A slope and B/A an intercept [21].

The adsorption isotherm (Halsey) is expressed as follows [22]:

$$\ln C_e = \frac{\ln K}{n_H} - \frac{\ln C_e}{n_H} \tag{5}$$

 n_H , k are the constant of Halsey isotherm.

The Temkin model assumes that the change in heat of absorption occurs in a linear manner. in contrast to the logarithmic relationship described by the Freundlich equation [23]. The sorption heat of all molecules in the layer would diminish linearly with coverage because of sorbate/sorbent interactions. The following expression represents the Temkin isotherm [24]:

$$q_e = B_T (\ln A_T + \ln C_e)$$

Absolute temperature is measured in Kelvin, and the universal gas constant R is expressed as 8.314 J/mol K. B_T Is adsorption heat, while A_T is the constant of equilibrium of association (Lg^{-1}) associated with the highest energy of binding. A_T and B_T are constants that are derived from the slope and intercept of a graph that illustrates the connection between the solute concentration ($\ln C_e$) and the amount of adsorption (q_e).

5. Adsorbents in water purification

Adsorption is an appropriate method for treating industrial wastewater since it is easy to use and reasonably priced. The kind and amount of contaminants present, together with the adsorbent's effectiveness and capacity, all influence the choice of adsorbent. Adsorbents ought to be inexpensive, widely accessible, non-toxic, and quickly regenerable. Water and industrial effluents have been filtered using a variety of adsorbents like biomass, natural material, agricultural waste and residues, and industrial wastes [25], [26].

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5.1. Low-cost adsorbents

Can be classified as illustrated in Figure 2 [27].



5.1.1. Natural adsorbent materials

Many natural materials have adsorbent properties and can be found in large quantities. Although there are many natural adsorbents, chitin, zeolite, clay, peat moss, wood, and coal have been used successfully to remove organic chemicals, colors, and heavy metal ions from water and wastewater [28].

5.1.2. Agricultural wastes

Fruit and vegetable peels, as well as agricultural leftovers, are thrown away as waste materials that have no use. They can function as cost-effective adsorbents with little processing [29]. Because of their distinct structure and chemical properties, lignin and cellulose, which make up the majority of agricultural wastes, make them potential alternative adsorbents. Certain functional groups included in polymer chains, such as phenol, alcohol, aldehyde, ketone, and carboxyl, can remove a variety of impurities from water [30].

5.1.3. Wastes from industry

Heavy metals can be removed from contaminated water using inexpensive adsorbents made from industrial waste, which is a byproduct of several industries. They are affordable and easily obtainable in big quantities. Numerous industrial byproducts, including palm oil ash [31], red mud [32], tea factory waste [33], coffee waste [34], fly ash [35], bagasse ash [36], blast furnace slag, were examined for their potential to remove hazardous materials from water and wastewater. Waste materials' physical properties have an impact on the adsorption process as well [37].

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5.1.4. Techniques for Natural Adsorbent Preparation

- 1. Drying and grinding: Natural raw materials, such as fruit peels or agricultural waste, are cleansed to get rid of contaminants using distilled water. And dried in an oven at suitable temperatures (usually between 50-110°C) to remove moisture. They are ground to fine sizes to increase the effective surface area [38].
- 2. Chemical activation: The raw materials are treated with chemicals such as sulfuric acid or zinc chloride. They are heated at high temperatures to improve their porosity and adsorption capacity [39].
- 3. Thermal activation: The raw materials are heated in an inert atmosphere (such as nitrogen) at high temperatures (up to 800°C). This helps to increase the porosity and surface area available for adsorption [40].
- 4. Surface modification: Certain functional groups, such as amino or carboxylic groups, are added to the surface of the adsorbent. This improves the ability of the material to selectively adsorb certain compounds [41].
- 5. Using natural waste without treatment: In some cases, natural waste is used directly as an adsorbent after washing and drying, without any further treatment. This depends on the nature of the raw material and its adsorption efficiency [42].

6. Parameters affecting adsorption

6.1. pH effect

(Jassim et al., 2014), they employed Iraqi palm tree leaves (leaves burnt) as an adsorbent to extract Cu⁺² from solutions. In a series of batch experiments, they investigated the effect of pH on adsorption to determine the best conditions for copper removal. All the parameters were set(mixing time = 1 hr, shaking rate= 300 rpm, Copper concentration = 100 mg/l). The result is shown in Figure 5. These comparisons showed that this low-cost adsorbent is effective in removing copper from industrial wastewater before it is discharged into the river [43].

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(Hassoon, 2015), they investigated the potential for eliminating heavy metals from an aqueous solution, including copper, cadmium, lead, and chromium. They employed a fixed metal concentration of 1000 ppm and investigated a range of pH values (Table 3). One gram of dried plant material was employed. According to the study, this plant can effectively remove heavy metals from tainted water[44].

				10 J 10 J	1.0			
	Cf	Cd	Cf	Cu	Cf	Pb		Cr
		Removal %		Removal %		Removal %	Cf	Removal %
pH	79.9	92	34.4	96.5	5	99.5	160	84
4								
pH	81	92	31.5	96.8	3.12	99.6	200	80
5								
pH	85	91.5	33.7	96.6	16.8	98.3	200	80
6								
pH	30.1	97	32.2	96.8	5.22	99.4	190	81
8								

Table 3. Effect of pH on the removal rate [44].

(Al-Saade et al., 2017), the researchers investigated a environmentally friendly method and cost-effective of extracting lead (Pb) from industrial wastewater. To remove lead ions from industrial wastewater, they used a plant from Iraq called Phragmites australis (Pa) and treated it with sodium dodecyl sulfate (SDS) as a biomaterial. They investigated the impact of pH on the removal rate to identify the optimal settings for metal adsorption. Figure 6 displays the findings [45].

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Figure 6. pH effect on removal rate, metal concentration= 20 ppm [45].

(Baby et al., 2019), they investigated the use of adsorption to remove toxic heavy metals from aqueous solutions. Palm kernel shells (PKS), which are produced in vast quantities from palm farms, were utilized by them as an efficient and cost-effective material. To prepare palm kernels, they were cleaned, thoroughly dried, and ground into a powder. Toxic Heavy metals like Cd^{+2} , Zn^{+2} , Pb^{+2} , Cr^{+6} are present in an solution and are eliminated based on a several parameters, including pH. Figure 7 displays the results [46].



Figure 7. pH impact on removal percentage [46].

(Alfaize et al., 2020), they investigated how well light - expanded clay aggregate from the Al-Khawrah River in Basra could remove heavy metal ions like iron, copper, zinc, and chromium from the General Company for Petrochemical Industries' industrial wastewater using the adsorption batching technique. They conducted a study on the effects of process variables like pH. The results are shown in Figure 8. By suitable the experimental data to isotherm models like Freundlich or Langmuir adsorption, they were able to interpret it. According to the findings,

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the adsorption of Cu, Zn, and Cr followed the Langmuir model, and Fe adsorption followed the Freundlich model [47].



Figure 8. Effect of pH on removal (size of particle = 0.15 mm, T = 25 °C, time = 35 min, dose = 1.0 g, initial concentration is 0.02M [47].

(Hummadi, 2021), in batch adsorption tests employing date pits, they investigated the ideal parameters that directly impact the removal of heavy metals such as Zn, Ni, and Cu, from aqueous solution. At a metals concentration of 60 ppm, agitation rate of 300 rpm, and adsorbent dose of 0.12 g per milliliter of an aqueous solution, they investigated the pH effect on removal capacity. In the date pit (Al-Zahdi Iraqi), heavy metals are adsorbed through an exothermic process. The Freundlich model provided an excellent description of the adsorption equilibrium. Figure 9 shows the outcomes [48].



Figure 9. pH impact on removal ability [48].

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(Al-Sareji et al., 2021), to extract copper (Cu) from tainted water, they employed sawdust as an active adsorbent (1.8 units of concentrated H_2SO_4 mixed with 1 unit of sawdust. With a contact time of 150 min and agitation rate of 150 rpm, a concentration of copper of 5 mg per L, and a dose of sawdust of 2 g per L. they conducted the adsorption tests in a batch manner and examined the impact of solution pH on the outcomes. The outcomes are displayed in Figure 10. They have successfully used sawdust as a low-cost sorbent to remove copper [49].



6.1.1 Summary of the pH effect on adsorption efficiency

The effect of pH on adsorption efficiency is an important operational factor that has been studied in many studies. Studies show that pH plays a pivotal role in determining the effectiveness of adsorbents in removing heavy metals, as each metal requires a specific pH value to achieve the highest adsorption efficiency. For example, a study [43] showed that copper adsorption increases significantly at pH 6, while another study [44] found that copper and lead adsorption are similar at pH values between 4 and 8, whileThe maximum adsorption rates for chromium and cadmium occurred at pH 4 and pH 8, respectively. In another study [46], it was proven that lead, chromium, zinc, and cadmium are most effectively adsorbed at pH 9 for all metals. While another study [47] showed that the best adsorption of zinc, iron, chromium, and copper occurs at pH 5, 4, 2, and 4, respectively. Furthermore, further research [48],[49] demonstrated that the optimal adsorption for for zinc and nickel, it occurred at pH 6. Whereas for copper occurred at pH 7,

Based on these results, it can be concluded that pH has a significant effect on the adsorption efficiency, as it contributes to changing the properties of the adsorbent and thus increasing its ability to capture heavy metals under certain conditions.

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6.2. Contact time effect

(Anirudhan & Sreekumari, 2011), the researchers studied the contact time effect on the adsorption of lead (II), copper (II), and mercury (II) from industrial wastewater using activated carbon made from coconut waste as an adsorbent. Adsorption experiments were conducted with varying metal concentrations of 25 - 100 mg per L and 2 g per L of adsorbent. The results showed that increase in the adsorption of metal ions when the contact time increased, and remained constant at 3 h. The results showed that the amount of adsorbed heavy metals increased from 12.4 to 44.4 mg per g for lead (II), from 12.21 to 40.77 mg/g for mercury (II), and from 11.92 to 36.40 mg/g for copper (II) [50].

(Bernard E. et al., 2013) The researchers investigated the possibility of removing Cu, Fe, Zn, and Pb ions from industrial wastewater using coconut shell activated carbon (ACS) as an adsorbent. Zinc chloride was used to activate the chemically produced activated carbon. They investigated The impact of contact time on the adsorption of Cu^{+2} , Fe⁺², Zn⁺² and Pb⁺² from wastewater in a batch adsorption experiment. The dosage of adsorbent was 0.2 g, the mixing rate was 150 rpm and pH was 2. The results are shown in Figure 12 [51].



Figure 12. Contact time Effect on removal rete [51].

(Alfaize et al., 2020), they studied the use of the adsorption batching technique to evaluate the Expanded lightweight clay block capacity extracted from Al-Khawrah River in Basra, to heavy metals removal such as copper, chromium, zinc, , and iron from industrial waste water of the General Company for Petrochemical Industries. They analyzed on the impact of contact time. Figure 16 displays the results. They analyzed the experimental data by fitting them to the isotherm models such as Freundlich and Langmuir adsorption. The results show that the

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adsorption of Cr , Zn and Cu conformed to the Langmuir model, whereas Fe, it accorded to the Freundlich pattern [47].



Figure 16. Effect of time on removal %(T=25 C, size of particle = 0.15 mm, dose = 1.0 g, pH = 2 (Cr), 5 (Zn), and 4.5 for both Cu, and Fe, and the concentration of metal is 0.02M [47].

(Al-Sareji et al., 2021), they used sawdust as an activated adsorbent (1.8 units of concentrated H_2SO_4 mixed with 1 unit of sawdust) to remove copper (Cu) from contaminated water, They used a batch method to do the adsorption tests and looked at how contact time affected the removal rate, at pH 4, agitation speed of 150 rpm, 5 mg per L of concentration of copper, and 2 g per L of sawdust dose. The results as shown in figure 17. They have effectively employed sawdust as an inexpensive sorbent for copper removal [49].



Figure 17. Effect of contact time on removal % [49].

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(Rajakumar et al., 2023), to evaluate the effectiveness of heavy metals removal such as Cr, Pb and Cu from an aqueous solution, they used an adsorption approach. The biochar that the researchers employed was made from Tamarindus indica shells. To obtain the biochar, contaminants were washed away with deionized water, dried in an oven at a specific temperature, and then powdered for additional usage as an adsorbent. They looked into variables that affect adsorption including hydrogen ion, contact time, and adsorbent dosage. The results shown the metals concentration decreased when contact time increasing, as shown in Table 4 [52].

ĺ	Cr removal		Pb re	moval	Cu removal		
	Time	Cr g/L	Time	Pb g/L	Time	Cu g/L	
	(min)		(min)		(min)		
	0	4.2	0	1.5	0	2.8	
[15	3.8	15	1.4	15	2.4	
	30	3.3	30	1.1	30	2.0	
	45	2.9	45	0.98	45	1.6	
	60	2.5	60	0.72	60	1.2	
ġ	75	2.4	75	0.33	75	0.75	
ŝ	90	1.7	90	0.33	90	0.2	
-	105	1.5			105	0.2	
2	120	1.5					

Table 4. Heavy metal removal [52].

(Mohammed & Saleh, 2023), researchers studied the impact of time on the Lead adsorption from industrial waste water generated by the Northern Refineries Company in Iraq .The researchers made a nano-adsorbent material from attapulgite clay. As seen by Table 5, the results demonstrated that the removal rate rises as contact time increases [53].

Tuble et contact time impact on removal rate [ce]	Table 5.	Contact	time i	mpact on	removal	rate	[53].
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Element	pН	Adsorbent	Contact	Pb	Pb(mg/L)	Removal
		dose	time	(mg/L)	After	rate
		(mg/L)	(min)		removal	
	7	200	15	0.2	0.04	80%
Lead(Pb)	7	200	25	0.2	0.032	84%
	7	200	30	0.2	0.024	88%

6.2.1. Summary of the effect of contact time on adsorption efficiency

The effect of contact time is an important operational factor that affects the adsorption efficiency. Research shows that extending the contact time can enhance the adsorbent's capacity to remove heavy metals from the solution. Many studies have shown that adsorption continues to

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increase with time until it reaches an equilibrium point, where the adsorption process stops progressing significantly. For example, a study [50] showed that the adsorption of lead, copper, and mercury continues to increase significantly until it reaches about 3 hours, while another study [51] found that the adsorption of lead, zinc, iron, and copper continues effectively until 60 minutes, after which no significant increase in adsorption occurs. In a third study [47], it was observed that the ideal contact time for the removal of zinc, chromium, iron, and copper was 90 minutes, where the highest adsorption rate was achieved during this time period. On the other hand, study (49) showed that copper adsorption did not reach equilibrium until after 150 minutes, while in another study [52], the ideal time for lead, copper, and chromium adsorption was about 90 minutes.

6.3. Adsorbent dose effect

(Aman et al., 2008), the researchers studied the adsorbent dosage effect on adsorption to remove heavy metal Cu (II) from industrial wastewater. They used potato peel charcoal (PPC) as the adsorbent. The results are shown in Fig. 18 when the concentration of metals was 150 mg per L, hydrogen ion of 6 and contact time of 120 min [54].





(Anwar et al., 2010), they investigated the adsorption of lead and cadmium onto peels of bananas in a batch system. They used flame atomic absorption spectroscopy to determine the metals' concentration. They study the effect of the adsorbent dosage. The findings are displayed in Figure 19 [55].







Figure 19. The impact of adsorbent dosage on heavy metals removal (contact time: 20 min; agitation speed: 100 rpm, metal concentration, 50 µg/ mL) [55].

(Bernard E. et al., 2013), the researchers investigated the possibility of removing Pb, Fe, Zn, and Cu ions from industrial waste water using coconut shell activate carbon (ACS) as an adsorbent. Zinc chloride was used to activate the chemically produced activated carbon. The effect of adsorbent dose on the adsorption of Pb, Zn, Fe and Cu from wastewater was investigated in a batch adsorption experiment. The contact time was 80 min, mixing rate was 150 rpm and pH was 2. Figure 21. Showed The results [51].



Figure 21. Adsorbent dose impact on removal rate [51].

(Jassim et al., 2014), they employed Iraqi palm tree leaves (leaves burnt) as an adsorbent to extract Cu⁺² from aqueous solutions. In batch experiments, they investigated the effect of adsorbent dose on adsorption to determine the optimal conditions for copper removal. All the

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parameters were set (time = 1 h, agitation speed = 300 rpm, Copper concentration = 100 mg/l). The result is shown in figure 22. These comparisons showed that this low-cost adsorbent is effective in removing copper from industrial wastewater before it is discharged into the river [43].



(Baby et al., 2019), they studied the possibility of remove toxic metals from an aqueous solution through adsorption. They used palm kernel shells (PKS) as an economical and effective material that is produced in large quantities from palm farms. Palm kernels were prepared by washing the kernels, drying them well, and grinding them into powder. Depending on Parameters like adsorbent dose, heavy metals in an aqueous solution are removed. Figure 24 displays the findings [46].



Figure 24. The adsorbent dose Effect on removal rate [46].

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(Hummadi, 2021), they conducted a study on the optimum factors that have a direct effect on toxic heavy metal removal like Zn, Cu, and Ni from aqueous solution in batch adsorption studies using date pits (Al-Zahdi Iraqi). They studied the adsorbent dose effect on removal capacity at an initial metal concentration of 60 ppm, particle size of adsorbent of 200 μ m, shaking speed of 300 rpm, pH 6, The process of adsorption of heavy metals in the date pit (Al-Zahdi Iraqi) is exothermic process. Adsorption equilibrium is accurately characterized using the Freundlich isotherm. The findings revealed that copper's adsorption capability and adsorption intensity are superior to those of zinc and nickel. Figure 25 displayed the results [48].



(Al-Sareji et al., 2021), they used sawdust as an activated adsorbent(1.8 units of concentrated H_2SO_4 mixed with 1 unit of sawdust) to remove copper (Cu) from contaminated water, They used a batch method to do the adsorption tests and looked at how the adsorbent dose affected the removal rate, at pH 5, agitation speed of 150 rpm, 5 mg per L of concentration of copper and 2 g per L of sawdust dose and time of 150 m. The results as shown in figure 26. They have effectively employed sawdust as an inexpensive sorbent for copper removal [55].

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Figure 26. Effect of adsorbent dose on removal rate (55).

6.3.1. Summary of the effect of pH on adsorption efficiency

Adsorbent's capacity to extract heavy metals from the solution is mostly determined by how the adsorbent dose affects adsorption efficiency. Studies indicate that increasing the dose of the adsorbent can improve the adsorption capacity until reaching a certain point, where increasing the dose does not lead to a significant improvement in the adsorption efficiency. For example, study [54] showed that increasing the dose of the adsorbent leads to a gradual increase in the adsorption of copper until it reaches a dose of 1g/100ml, after which there is no noticeable improvement in adsorption. While another study [55] showed that lead requires a larger dose of the adsorbent (40g/L) compared to cadmium, which requires 30g/L, where the highest adsorption rate can be reached by increasing the dose until a certain point, until increasing the adsorbent becomes negative and the adsorption rate decreases. The reason is that increasing the dose of the adsorbent significantly may lead to agglomeration, which reduces the surface area available for adsorption. A third study [46] showed that the adsorption of lead, iron, copper, and zinc continued to increase significantly with increasing dose at 1g/100ml. Another study [48] showed that the copper adsorption rate increased with increasing dose of the adsorbent until it reached the best adsorption rate at a dose of 3g/L.

In addition to the various surface characteristics of each adsorbent, modifications to the adsorbents improve their adsorption capacity, which causes variations in dosage concentrations among experiments.

6.4. Metal concentration effect

(Dhabab et al., 2012), researchers have developed a novel technique to effectively extract cadmium ions from industrial wastewater. This process involves utilizing both natural and

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modified dried leaves of Ceratophyllum demersum. This plant is a variety of spices native to Iraq and cultivated along the banks of Iraqi rivers. They studied the metal concentration effect. The following fixed parameters apply time =60min, pH=2.3, temperature=30 °C, and starting concentration= 50 mg/L. The results are shown in figure 27. Based on the batch sorption isotherm results, the Langmuir isotherm was fit for the modified Ceratophyllum demersum, and the Temkin isotherm was fit for the natural ceratophyllum demersum [56].



Figure 27. Effect of metal concentration on removal rate [56].

(Hassoon, 2015), they investigated the potential for eliminating heavy metals from an aqueous solution, including copper, cadmium, lead, and chromium. They employed a fixed pH of 7 and varying the metal concentration (as indicated in Table 6). One gram of dried plant material was employed. According to the study, this plant can effectively remove heavy metals from contaminated water [44].

	Cd	1		Cı	1		Pb)		Cı	r
Ci	Cf	Removal									
		%			%			%			%
250	3.5	98.6	250	30.9	87.6	250	3.51	98.5	250	43.5	82.6
500	47.4	90.5	500	29.9	94	500	3.94	99.9	500	81	83.8
750	5.8	99.2	750	31.1	95.8	750	1.2	99.8	750	151	79.8
1000	92.7	90.7	1000	33.1	96.7	1000	0.43	99.9	1000	212	82

Table 6. Effect of Heavy metal concentration on the removal rate [44].

(Baby et al., 2019), they studied the removal of toxic heavy metals from aqueous solution through adsorption. They used palm kernel shells (PKS) as an economical and effective material that is produced in large quantities from palm farms. Palm kernels were prepared by washing the kernels, drying them well, and grinding them into powder. They study the metals concentration

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effect. Heavy metals present in an aqueous solution like Cr - Pb - Cd and Zn are removed. Figure 28 displays the findings [46].



Figure 28. The impact of metal concentration on removal rate [46].

(Hanane et al., 2020), they investigated the performance of zeolite in removing heavy metals from an industrial effluent. They conducted zeolite characterization using XRD, FTIR, and SEM. They extracted ten heavy metals from an industrial zone aqueous effluent. The data indicate that the metal clearance rates decrease in the following order: The sequence of elements is Cd > Mn > Sn > Fe > Cr > Ni > Pb > Mg > Zn > Cu. The metal cadmium has the highest adsorption rate (86.47%), while copper has the lowest clearance rate (19.27%). The study illustrates that, in addition to its economic utility, zeolite enhances the adsorption process. Table 7 presents the data [57].

Table 7. Removal rates of heavy metals, at contact time 60 min, 0.5 g of zeolite, temperature 25°C [57].

Metal	C _i (mg/L)	C _f (mg/L)	Rate %	C _{Max} (mg/L)
Fe ⁺²	4.543	1.157	74.541	1.00
Cu ⁺²	4.377	3.533	19.269	1.00
Zn^{+2}	0.917	0.643	29.818	2.00
Mn ⁺²	11.377	2.767	76.420	-
Ni ⁺²	21.520	13.150	38.894	2.00
Sn ⁺²	0.533	0.130	75.625	0.10
Mg ⁺²	239.067	154.707	35.287	300.00
Pb ⁺²	0.573	0.370	35.465	0.50
Cd ⁺²	0.397	0.054	86.471	0.10
Cr ⁺²	0.782	0.214	72.665	2.00
Total	284.442	176.742	37.870	-

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(Priva et al., 2022), they investigated the potential of husk from rice (RH) as an adsorbent to remove toxic heavy metals like chromium, lead, and zinc) from aqueous solution. The researchers used activated RH powder as the starting material and treated the adsorbent with HCl of 0.1 N, to generate functional groups and enhance the surface area. They studied the metal concentration effect on the removal rate for solutions containing 25-150 mg per L of Cr- Pb and Zn ions. The results showed that the toxic heavy metal removal rate increased from 60.84 to 90.8% for chromium, from 65.42 to 96.12% for lead, and from 66.83 to 94.36% for zinc. These results appeared when the content of metal decreased from 150 to 25 mg/L at 1 hour and a temperature at 30 °C, while adsorbent dose at 2.5 g/L and of pH 6 [58].

6.4.1. Summary of the effect of Metal concentration on adsorption efficiency

All previous studies that reviewed the initial concentration effect of heavy metals agree that increasing the metal concentration leads to a gradual decrease in adsorption ratio. This is because the active sites on the adsorbent surface are abundant at low concentrations, allowing for greater adsorption. However, as the metal concentration increases, these sites gradually become saturated, reducing the adsorption efficiency. Therefore, the best adsorption occurs at lower concentrations, while the adsorption is lower at higher concentrations due to the gradual saturation of the active sites on the adsorbent surface.

7. Conclusions

The reviewed studies indicate that natural adsorbents, such as agricultural waste, clays, and biochar, provide an environmentally friendly and effective solution to remove heavy metals from industrial wastewater. These materials are abundant and low-cost. Many studies have proven their ability to improve water quality and sustainably reduce pollution manner, especially in comparison to synthetic adsorbents that may have adverse environmental impacts. Researchers have found that chemical modification of natural materials, such as activation with acids, bases, or other reagents, is effective in significantly increasing the removal efficiency of heavy metals. For instance, acid activation can enhance the surface area and porosity of adsorbents, leading to better adsorption performance. They studied several influencing factors, including contact time, pH, initial metal concentration, and the dosage amount of the adsorbent. Each of these factors plays a critical role in determining the overall adsorption capacity. The results showed that these factors have both negative and positive effects on the process of adsorption, as the adsorption percentage increases and decreases according to the variation of these factors, as highlighted in previous studies. There is a noticeable difference in the results depending on how these parameters are controlled and adjusted in experimental setups. This variability highlights the need for standardized testing conditions to allow for better comparison across studies. However, despite these advancements, there are still challenges in improving the adsorption efficiency under different and complex conditions, such as in the presence of competing ions or fluctuating wastewater compositions. Long-term studies are still needed to evaluate the sustainability of

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these solutions, particularly in real-world applications where multiple variables may interact simultaneously. Therefore, developing efficient recycling methods to regenerate adsorbents after use, extending their functional lifespan, and exploring the possibility of using these adsorbent materials multiple times without significant loss of efficiency, especially in their natural state without chemical modification.

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الممتزات الطبيعية لإزالة المعادن الثقيلة من مياه الصرف الصناعي: دراسة مراجعة امير هادي حسن عباس عبد الكاظم خليف على فاضل ناصر

[قسم هندسة البناء والإنشاءات، كلية المسبيب التقنية، جامعة الفرات الأوسط التقنية، بابل، العراق

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الخلاصة

تشكل المعادن الثقيلة الموجودة في النفايات الصناعية تهديدات خطيرة للبيئة والصحة البشرية، وذلك بسبب سميتها ومتانتها البيئية. ومن المراحل الحاسمة في معالجة مياه الصرف الصحي إزالة هذه الملوثات. ومن بين الطرق العديدة المستخدمة، وجد أن الامتصاص هو طريقة فعالة من حيث التكلفة لإزالة المعادن الثقيلة. ويعتمد هذا الإجراء على المواد الماصة - الطبيعية أو الاصطناعية - التي يمكن أن ترتبط بأيونات المعادن وتستخرجها من الماء. ركزت هذه الدراسة على مراجعة دور الباحثين في إيجاد مواد ماصة غير مكلفة ومواد ماصة مناسبة للبيئة، وخاصة للمواد الطبيعية والنفايات الزراعية والصناعية. إن توافر هذه المواد، وقابليتها للتحمل، وكفاءة الامتصاص العالية تحددها. وتناقش هذه الورقة تأثيرات العديد من المتغيرات على آلية الامتصاص بما في ذلك الرقم الهيدروجيني، وجرعة المادة الماصة، ووقت التلامس وتركيز المعدن. والهدف من الدراسة هو تقديم ملخص للدراسات التي استخدمت المواد الطبيعية لإزالة المعادن الثقيلة. واظهار قدرتها على استخراج المعادن الثقيلة من النفايات الصناعية. وقد تم فحص درجة الحموضة وزمن التلامس وتركيز المعادن ودرجة الحرارة وكمية جرعة المادة الماصة، حيث يعتبر كل عنصر من هذه العناصر أساسياً في تحديد سعة الامتصاص الكلية. ومن بين النماذج الأكثر استخداماً من قبل الباحثين لدراسة سلوك الامتصاص، يعتبر نموذجا لانجموير وفرويندليش الأكثر شيوعاً واستخداماً على نطاق واسع في التطبيقات البيئية والصناعية. يتميز نموذج لانجموبر بقدرته على وصف الامتصاص على سطح متجانس في تكوين طبقة واحدة، مما يجعله مثالياً لتحديد أقصبي سعة امتصاص، وهو مناسب للعديد من المواد ذات مواقع الامتصاص المحددة. من ناحية أخرى، يتميز نموذج فرويندليش بمرونته في التعامل مع الأسطح غير المتجانسة وتفاعلات الامتصاص متعددة الطبقات، مما يجعله مناسباً للأنظمة ذات الطبيعة المعقدة. ترجع شعبية هذين النموذجين إلى بساطتهما ودقتهما في تمثيل البيانات التجريبية في مجموعة واسعة من التطبيقات.

الكلمات الدالة: - الامتزاز، المعادن الثقيلة، مياه الصرف الصناعي، المتساوي الحراري.